Spin Dynamics
in Disordered Systems

Spindynamik in ungeordneten Systemen

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A.1 Spin cluster in a twisted-boundary condition ................................. II
Part I

Spin precession in disordered systems
1 Anomalous relaxation due to heavy–tailed field distributions

1.1 Introduction

Spin precession

Many important experimental techniques rely on the precession of a spin probe $S$ in a local magnetic field $H$,

$$\frac{dS}{dt} = S \times H,$$

whereby, for notational convenience, we identify the field $H$ with the Larmor frequency $\gamma \mu_0 H$ ($\gamma$: gyromagnetic ratio; $\mu_0$: magnetic permeability of free space), $S$ is understood dimensionless and normalized ($|S| = 1$), and the equation as such actually applies to its quantum–mechanical expectation value.

Part of the justification for this classical treatment is that, for obvious practical reasons (signal strength), one typically monitors the polarization of a large ensemble of independent spin probes. Especially for disordered systems, the corresponding averaged quantity $\langle S(t) \rangle$ evidently yields quite ‘indirect’ information on the local field $H(t)$, which can vary in space and fluctuate in time. So the study of the relation between characteristics of the local fields and the observable $\langle S(t) \rangle$ poses an interesting challenge to theory.

Examples of relevant experimental techniques include nuclear and electron magnetic resonance (NMR, ESR; see e.g. [Abr86, Shi92]), muon spin relaxation ($\mu$SR) and the closely related $\beta$NMR (cf. below), but also certain quantum optical measurements, where transitions in two–level systems can effectively be described by an equation of type (1.1) (see e.g. [Bar04]).

Theoretical background and aim of our approach

Quite generally, the dynamics of disordered systems is often governed by spatial and temporal fluctuations that can be described by Gaussian statistics due to the central limit theorem (CLT). However, in an increasing number of systems it is found that anomalous broad Lévy distributions of the fluctuations play a key role [Shi95, Kut99]. Lévy distributions generalize the CLT if the fluctuations arise from a superposition of independent random contributions whose distributions exhibit no finite second moment [Bou90]. Intriguing consequences for
dynamical processes in many different scientific disciplines have been explored, mainly in
the context of anomalous long-range transport phenomena (see e.g. [Kla96]).

Similarly, spin precession was traditionally studied for Gaussian stochastic processes \( H(t) \),
which generically give rise to exponential decays of an initial spin polarization [Kub67, 
Uem85]. This matches the phenomenological approach by Bloch, where field fluctuations
are taken into account by adding to (1.1) relaxation terms (which are linear in the respective
component of \( S \)). The corresponding relaxation rates can be expressed by spectral densities
of the fluctuations [Abr86].

More complex stochastic processes \( H(t) \), on the other hand, may yield anomalous non–
exponential relaxations, to which the standard Bloch approach can no longer be applied
straightforwardly. Specifically, we may distinguish two particularly relevant cases: (i) The
fluctuations exhibit long–range temporal correlations, \( \langle H(t)H(0) \rangle \sim t^{-\nu} \) with \( \nu \leq 1 \) such
that a characteristic correlation time does not exist, and (ii) the field distribution \( \psi(\mathbf{H}) \) of
local fields is anomalously broad, e.g. \( H^2\psi(\mathbf{H}) \sim H^{-1-\eta} \) with \( \eta < 2 \), such that it exhibits
no finite second moment. The first case is often referred to in the analysis of spin glasses
and similar complex systems of interacting magnetic field sources [Ker96, Cam99].

The second case arises quite naturally when considering an ensemble of independent spins
in a system of randomly distributed field sources, which (in the simplest case) may be taken
non–interacting, yet whose contribution to \( H \) decays as \( \sim r^{-\mu} \) with distance \( r \) from the
precessing spin probe \((\mu > d/2, \text{cf. below})\). As an example of practical importance, which
has not been treated comprehensively before, we will refer to \( \mu \text{SR} \) in random assemblies
of superparamagnetic clusters [Bew98, Jac00a], whose magnetic moments reorient with a
thermally activated rate \( \nu \). While the corresponding dipolar fields decay as \( \sim r^{-3} \),
we will more generally consider fields \( \sim r^{-\mu} \) from randomly distributed sources in dimension \( d = 2 \)
and 3.1

So the aim of Part I of the present work is as follows. We will show how broad local
field distributions emerge in this conceptually simple system and how they give rise to rich
anomalous spin relaxation scenarios, whose long–time relaxation behavior is characterized
by power laws and stretched exponentials with exponents depending on \( \mu \) and \( d \). We
will make new predictions for \( \mu \text{SR} \) experiments in the systems referred to above. Due to
the generality of the problem and the simple scaling arguments we invoke, our findings
should also be of importance for related problems, where the precession dynamics plays a
fundamental role.

**Muon spin relaxation**

Before precisely defining our theoretical model, let us briefly digress on our primary
experimental reference. We start with the \( \mu \text{SR} \) technique, which is typically less well–known
than e.g. NMR or ESR. For a comprehensive overview, we refer to [Dal97, Lee99]; a concise
description may be found e.g. in [Ana05].

1Note that the dimension \( d \) refers to the geometrical configuration of clusters, which could be confined
e.g. to a two–dimensional surface \((d = 2)\), whereas we always take spins, (magnetic) moments and fields
to have three components as usual.
Basically, $\mu$SR consists in implanting (i.e. irradiating) muons into a sample and studying the dynamics of their spins via the associated magnetic moment in analogy to the observation of already present nuclear or electronic spins in the case of NMR or ESR.

The muon is a fundamental particle similar to the electron (i.e. a lepton, spin $1/2$), yet $\sim 207$ times the mass of the latter and a similarly higher magnetic moment (yielding a gyromagnetic ratio of $\gamma \simeq 851.5$ MHz/T). As usual, it comes as particle or antiparticle, $\mu^\pm$; however, since the $\mu^-$ (‘heavy electron’) gets captured close to an atomic nucleus, mainly the $\mu^+$ (‘light proton’) is employed in condensed matter applications as considered here, i.e. to probe interstitial magnetic fields. Eventually, the $\mu^+$ decays with a half-life period $\tau_{1/2} \simeq 2.19$ $\mu$s to a positron and two neutrinos,

$$\mu^+ \rightarrow e^+ + \nu_e + \bar{\nu}_\mu. \quad (1.2)$$

Thereby, both the usual muon production process$^2$ and the decay violate parity, so that the produced muons are in fact nearly 100% spin-polarized opposite to their momentum and the $e^+$ is emitted preferentially in direction of the former muon spin.

Thus, as a distinctive feature, the $\mu^+$ possess a definite initial polarization$^3$, whereas in other techniques, the initial polarization has to be achieved by thermal equilibration of the spins (requiring high external fields and/or low temperatures). Moreover, the average polarization $\langle S(t) \rangle$ of an ensemble of muons may be monitored indirectly as asymmetry in the directions of emitted $e^+$ (difference of ‘forward’ to ‘backward’ counts along the respective axes). Practically, the muons may be brought into the sample in form of a pulse or, alternatively, one after another (at intervals $\gg \tau_{1/2}$, so that any detected $e^+$ may still be related unambiguously to the time $t$ its parent $\mu^+$ had to precess).

Similar to NMR or ESR, $\mu$SR may be performed in the presence of static or dynamic external magnetic fields – so the ‘R’ in the acronym $\mu$SR may variably be taken to stand for rotation, relaxation, or resonance. As a specialty, however, it does not necessitate any applied field, hence it may be used in systems which would be strongly influenced by such fields, e.g. superconductors or spin glasses (cf. Part II of the present work). Also, muons allow to probe very weak internal fields $H$, such as of dilute sources. After all, $\mu$SR supplements well other magnetic probe techniques in a frequency range $10^4$–$10^{12}$ Hz.

The abovementioned $\beta$NMR, finally, is almost identical in principle, except that it employs $\beta$–decaying nuclei (e.g. $^8$Li, $^{11}$Be, $^{15}$O) instead of the $\mu^+$.

### Superparamagnetic clusters

As sample material, we consider a system of superparamagnetic clusters embedded randomly (i.e. at random positions) in a non–magnetic matrix, which occupies most of the volume. In the examples cited above, these were cobalt clusters embedded in copper [Bew98] and iron clusters embedded in silver [Jac00a]. The clusters are single domain ferromagnetic particles which, due to anisotropy effects, exhibit distinct easy magnetization axes [Dor97].

$^2$This is the decay of pions, which themselves are obtained by accelerated protons hitting nuclei in a target material.

$^3$Note that the $\mu^+$ are typically stopped fast ($\lesssim 1$ $\mu$s) and mainly by Coulombic processes in the sample.
Reorientations of a cluster’s magnetic moment $\mathbf{m}$ between its preferential directions may occur thermally activated with a rate

$$\nu = \nu_0 \exp \left( -\frac{E_a}{T} \right),$$

where $E_a$ is the anisotropy energy and $\nu_0$ is an attempt frequency. Thereby, $E_a$ is typically proportional to the particle volume times the so-called anisotropy constant $K$, which, among other factors, depends on the particle shape. Besides surface effects, the ferromagnetic material’s internal structure determines the number and symmetry of the easy axes. For the $\mu$SR, it will turn out important whether there are merely two or more preferential directions of the moment $\mathbf{m}$ per cluster (cf. below). Clearly, the study of systems of such fine particles generally is of great technological interest e.g. in view of magnetic recording applications.

The principle of a muon spin relaxation measurement in a sample as described is depicted in fig. 1.1(a); part (b) of the figure shows the heavy–tailed distribution of local field strengths $H$, which will be calculated in the following section. As already suggested by (1.3), we will consider zero–field $\mu$SR (i.e. no external magnetic field) in a dilute assembly of effectively non–interacting clusters. We will comment on these as well as further assumptions of our model and how our findings may be generalized by the end of the present Part I of this work. Also, more details of the concrete systems in view will be given in sec. 1.5, where we will discuss applications of our results to experiments.

For the present, let us add a remark on the relevant time scales: From equating the experimental time window of the $\mu$SR (or, actually, its upper limit), $t_{\text{exp}} \lesssim 10^{\tau_{1/2}}$, to a typical cluster flip time $\nu^{-1}$ as of (1.3), one obtains the so–called blocking temperature $T_B$. Accordingly, for measurements at temperatures $T \ll T_B$, one has to deal with essentially
static local fields, whereas for \( T \gtrsim T_B \), their (temporal) fluctuations become relevant.\(^4\) In our theoretical approach, on the other hand, we will distinguish different regimes of \( \nu^{-1} \) compared to \( W^{-1} \), i.e. the characteristic time scale of the spin precession determined by the field distribution \( \psi(H) \), cf. fig. 1.1(b). So, in sec. 1.5, we shall discuss how these regimes may be matched to the actual time window \( \lesssim t_{\text{exp}} \) in applications.

**Model definition**

To conclude this introductory section, let us state precisely the theoretical model which we will investigate in the following. We place a spin \( S \) at the origin of a \( d \)-dimensional system that contains randomly oriented point–like magnetic clusters with mean number density \( n \) at random positions (Poisson distribution of the cluster number in a volume \( \Delta V \) with mean cluster number \( n \Delta V \)). A cluster with moment \( m \) and position \( r \) is assumed to induce a field contribution \( h = m/r^\mu \) at the probe site. Each moment \( m \) changes its orientation to a set of possible other orientations with the rate \( \nu \). In particular we study two situations: In the first case, only the directions \( m \) and \(-m\) are possible (uniaxial case), while in the second case, each moment \( m \) may exhibit six mutually perpendicular orientations corresponding to a cubic symmetry (multiaxial case; cf. fig. 1.1(a)). Initially, the spin is polarized in the \( z \)-direction, \( S = (0, 0, 1) \). The task is to solve (1.1) for a given cluster configuration and a certain realization of the cluster reorientation process and to average this solution over all possible realizations. By finally averaging over all cluster configurations we obtain the spin polarization \( \langle S_z(t) \rangle \) at time \( t \) as measured in experiment.

### 1.2 Basic analysis and static case

#### 1.2.1 Local field distribution

As first step in the treatment of our model, we calculate the distribution \( \psi(H) \) of local fields \( H \). Note that, on the assumption of randomly oriented moments (at a given instant), this distribution applies to static as well as fluctuating (uni– or multiaxial) cluster moments. For computational convenience, we consider a \( d \)-dimensional spherical system with radius \( R \). The probability density \( \psi_r(r) \) for the position of a cluster is \( \psi_r(r) = 1/V_d R^d \), where \( V_d = (2\pi)^{d/2}/d\Gamma(d/2) \) is the volume of the \( d \)-dimensional unit sphere. The distribution of the field \( h = m/r^\mu \) generated by one cluster then is (for \( h \geq m/R^\mu \))

\[
\psi_h(h) = \int d^d r \psi_r(r) \delta(h - h(r)) = \frac{1}{4\pi} \frac{d}{\mu} \left( \frac{m}{R^\mu} \right)^{d/\mu} h^{-3 - d/\mu}.
\] (1.4)

Depending on the ratio \( d/\mu \), this may yield radically different probability densities \( \psi(H) \) of the total field \( H = \sum_{i=1}^N h(r_i) \) generated by \( N \sim n R^d \gg 1 \) clusters. For \( d/\mu > 2 \), \( h^2 \psi_h(h) \sim h^{-1-d/\mu} \) has a finite second moment \( \langle h^2 \rangle = d/(d - 2\mu)(m/R^\mu)^2 \) which, by proper

\(^4\) As a side remark, we note that for elevated temperatures (far above those considered here) the muons may start to diffuse in the sample.
application of the CLT (to the 3-dimensional, isotropic densities) gives a Gaussian \( \psi(H) \)
with a width diverging in the thermodynamic limit. For the \( \mu SR \), this would imply a
Debye–like relaxation with a relaxation rate depending on the system size (cf. above).

Hence, as advertised in the introduction, we will generically assume \( d/\mu < 2 \) which,
besides, covers the common experimental examples – such as dipolar \( (\mu = 3) \) or still faster
decaying fields in dimension \( d = 2, 3 \). Then, since the second moment \( \langle h^2 \rangle \) nominally
diverges, the CLT is not applicable any more and the summation of field contributions \( h_i \)
instead falls into the realm of the more general so-called Lévy statistics (see e.g. [Bou90, Bar01]).

Explicitly, \( \psi(H) \) may be calculated via its Fourier transform \( \hat{\psi}_H(q) = \hat{\psi}_h(q)^N \). Since for
small \( q \),
\[
\hat{\psi}_h(q) \sim 1 - \frac{d m \mu}{\mu R^d} |q|^d \int_0^\infty du \, u^{-1-d/\mu} \left( 1 - \frac{\sin u}{u} \right),
\]
we obtain in the limit \( R \to \infty, N \to \infty \) with \( n = N/V_d R^d \) fixed,
\[
\hat{\psi}(q) = \exp \left[ - (Wq)^{d/\mu} \right].
\]
The width \( W = C_W m n^{\mu/d} \) is given by the field \( \propto m/(n^{-1/d})^{\mu} \) associated with the mean
cluster distance \( n^{-1/d} \), whereby the constant \( C_W = \mu^{-1}dV_d \int_0^\infty du \, u^{-1-d/\mu}(1-\sin u/u) \) (see
also below).

Taking the inverse transform of (1.6) we obtain the exact result
\[
\psi(H) = \int \frac{d^3q}{(2\pi)^3} \hat{\psi}_H(q) e^{iqH} = \frac{1}{2\pi H} \int_{-\infty}^{\infty} \frac{dq}{2\pi} q \sin(qH) \exp[-(Wq)^{d/\mu}] \quad (1.7)
\]
\[
= -\frac{1}{2\pi H} \text{Re} \frac{\partial}{\partial H} \int_{-\infty}^{\infty} \frac{dq}{2\pi} \hat{\psi}^{\alpha H -(W|q|)^{d/\mu}} = -\frac{1}{2\pi W^2 H} \text{Re} I'_\alpha,0 \left( \frac{H}{W} \right), \quad (1.8)
\]
where \( \text{Re} I'_\alpha,0(u) \) denotes the real part of the derivative of the Lévy stable law \( I_{\alpha,0}(u) = (2\pi)^{-1} \int dk \exp(-iku - |u|^\alpha) \) to the index \((\alpha,0)\) (cf. the last-mentioned references). For
large \( H \),
\[
4\pi H^2 \psi(H) \sim \frac{2 \Gamma(2+d/\mu) \sin(\pi d/2\mu)}{\pi W} \left( \frac{H}{W} \right)^{-1-d/\mu} \quad (1.9)
\]
which, besides clearly qualifying \( \psi(H) \) as a heavy–tailed distribution, implies that the latter
has the same asymptotic behavior as \( \psi_h(h) \). This reflects the fact that, by contrast to the
Gaussian case, the superposition of the field contributions \( h_i \) is dominated by the largest
term.

Indeed, by considering e.g. the most probable values for the 1st, 2nd, 3rd, ... largest
contributions to a sum like our total field, one finds that the Lévy statistics implies a specific
hierarchy for the rank ordered summands (see e.g. [Emb97]); in our case, in particular, this
means that the \( k \)-th nearest cluster typically gives a contribution of order \( k^{\mu/d} \) times smaller
than the closest cluster. Note that such features of the Lévy statistics, which will play an
important role in our treatment of the \( \mu SR \) in presence of fluctuating cluster moments,
arguably were first discussed for a physical system in [Ciz93] in the context of dilute dipolar or RKKY spin glasses with power–law interactions (cf. Part II of the present work).

In the special case $d = \mu$, i.e. in particular for dipolar–like fields in $d = 3$, the integration in (1.8) may be performed explicitly yielding a three-dimensional variant of the Lorentzian (cf. [Kub81]),

$$\psi(H) = \frac{1}{\pi^2} \frac{W}{(W^2 + H^2)^2}$$  \hspace{1cm} (1.10)

with a characteristic width $W = \pi^2/3\mu m$ (for the latter, see also below). The corresponding heavy–tailed distribution of the modulus, $4\pi H^2 \psi(H) \sim H^{-2}$ for $H \gg W$, is shown in the preceding fig. 1.1(b).

In this context, let us point out that while (1.10) implies a simple Lorentz (or Cauchy) distribution $\pi^{-1}W/(W^2 + H_0^2)$ for any single Cartesian component $H_0$ of $H$, knowledge of the latter is not sufficient to recover the full $\psi(H)$. Clearly, this is because, by contrast to a Gaussian distribution, the $H_0$ are correlated random variables; yet, as for any isotropic distribution, the spherical field components are independent variables.

**Supplementary discussion**

In view of concrete applications, our derivation should be refined in two aspects. *First*, real clusters will not be exactly point–like, but have some finite (linear) extent $a$. Alternatively, this length could be understood as the minimum distance from the field source where the field, to a good approximation, takes the assumed algebraic form $h \sim m/r^\mu$.

Accordingly, the distribution $\psi_h(r)$ should be cut off at a minimal distance $a$, meaning that $\psi_h(h)$ of (1.4) will be limited above by a maximum field $h_{\text{max}} = m/a^\mu$ and the second moment $\langle h^2 \rangle$ will in fact be finite. Then, as usual in applications of Lévy statistics, one has to determine whether the heavy–tailed character of the distribution prevails for its envisaged use, or whether it will be dominated by the Gaussian developing by virtue to the CLT.

In our case, explicitly, the integral in (1.5) gets an upper limit $q h_{\text{max}}$. If the latter is much larger than unity, $q h_{\text{max}} \gg 1$, (1.6) will still hold asymptotically, whereas for $q h_{\text{max}} \ll 1$, we obtain $\psi(q) \sim \exp[-(W_0 q)^2]$ with a width $W_0 = (2 - d/\mu)^{-1/2}a d^n h_{\text{max}}^2$. Correspondingly, by evaluating the inverse Fourier transform as in (1.7), we recover the previous $\psi(H)$ of (1.8) asymptotically for small fields $H \ll h_{\text{max}}$, whereas it exhibits a sharper, Gaussian decay for large $H \gtrsim h_{\text{max}}$.

Thereby, the important point is that the crossover occurs for fields $H$ which are large compared to $W$, $H \sim h_{\text{max}} \gg W$, since we always assume $a \ll n^{-1/d}$, i.e. dilute clusters. Hence, such fields have very low probability $\psi(H)$, and the cutoff of the asymptotic behavior (1.9) due to the finite $h_{\text{max}}$ plays no role for the vast majority of cluster configurations.\footnote{Of course, this is obvious intuitively as, in a dilute system, the spin probe will very unlikely be extremely close to one or even several clusters.}

This may be seen explicitly in fig. 1.1(b), where we find the simulated local field distribution $\psi(H)$ (data points; plotted is the distribution of the modulus $H$) to closely follow the ‘idealized’ one of (1.8), (1.9) (gray line; cf. also below) except at very low probabilities –
1 Anomalous relaxation due to heavy–tailed field distributions

even for moderately high values of $a$ as compared to the mean cluster distance $n^{-1/d}$; also indicated are the maximum fields $h_{\text{max}}(a)$ for each $a$.

For the $\mu$SR, in particular, it will become clear from our treatment in the following that any specific effects of a finite $h_{\text{max}} \gg W$ would become perceptible in the decaying polarization $\langle S_z(t) \rangle$ only at very long times (and hence very small values of the latter) usually not accessible in experiments. Therefore, we will concentrate on point–like field sources as noted in the preceding model description. The assumed diluteness (and smallness) of clusters, $a \ll n^{-1/d}$, also keeps us from considering muons stopping within one of them. As for their spatial distribution $\psi_r(r)$, we neglect the possibility of overlapping or extremely close clusters, which eventually may not be taken independent any more (cf. chap. 2).

The second of the abovementioned refinements of our discussion concerns the particularly relevant case of dipolar fields in $d = 3$. The field at the origin due to a magnetic dipole moment $m$ at position $r$ is given by\(^6\)

$$h(r, m) = \frac{3(m \cdot r)r}{r^5} - \frac{m}{r^3}. \quad (1.11)$$

Thus, besides the already considered\(^7\) so-called secular part $h_1 = -m/r^3$, it also contains the non–secular part $h_{\text{II}} = 3(m \cdot r)r/r^5$ depending on the direction $\hat{r} = r/r$ of the dipole’s location (as viewed from the spin probe). Since both parts in (1.11) exhibit the same $r^{-3}$–dependence (and $\psi_h(h)$ will still be isotropic), this should not alter fundamentally our previous result (1.10). In particular, the influence of a potential cutoff $a$ will be as discussed above. However, to allow for a quantitative application to experiments, it is useful to re–calculate $\psi(H)$ precisely for the full dipolar fields.

In brief, one may start from the observation $h_1(h_{\text{II}}, r) = -3(h_1 \cdot \hat{r})\hat{r}$ (implying that $h_{\text{II}}$ are not independent random variables). A careful inspection of the implied angular relationships then yields the conditional probability $\chi(h_{\text{II}} | h_1) = (2\pi b_{\text{II}}^2)^{-1} \delta(h_{\text{II}} + 3 h_1 \cos \vartheta)$, where $\vartheta = \vartheta(h_1, h_{\text{II}})$ is the angle between the two contributions to $h$. As before, due to symmetry, the Fourier transform $\hat{\psi}_h(h)$ depends on $q = |q|$ only and thus can be spherically averaged, $\psi_h(q) = (4\pi)^{-1} \int d\Omega_q \hat{\psi}_h(q)$; whence

$$\hat{\psi}_h(q) = \int d^3h_1 \int d^3h_{\text{II}} \psi_1(h_1) \chi(h_{\text{II}} | h_1) \frac{\sin(q|h_1 + h_{\text{II}}|)}{q|h_1 + h_{\text{II}}|}, \quad (1.12)$$

in which $\psi_1(h_1)$ is given by (1.4) (for $\mu = d = 3$). Putting everything together, i.e. in particular noting that $|h_1 + h_{\text{II}}| = h_1(1 + 3\cos^2 \vartheta)^{1/2}$, we finally obtain (for small $q$)

$$\hat{\psi}_h(q) \sim 1 - \frac{\pi m}{4\hbar^3} \left(1 + \frac{\arcsinh \sqrt{3}}{2\sqrt{3}}\right) q, \quad (1.13)$$
in close analogy to the previous interim result (1.5) (again for $\mu = d = 3$).

\(^6\)Therein, in compliance to our previous choice of units (see sec. 1.1), we identify the magnetic dipole moment $m$ with $\gamma_\mu \mu m/4\pi$.

\(^7\)Note that, for the statistical treatment of randomly oriented clusters, we may formally let $m \mapsto -m$. 
Accordingly, we may practically ‘read off’ our final result for the dipolar fields (1.11), namely that the local field distribution exactly keeps its double-Lorentzian form (1.10), yet with a modified width

\[ W' = \frac{\pi^2}{3} \left( 1 + \frac{\arcsinh \sqrt{3}}{2\sqrt{3}} \right) m n . \]  

(1.14)

Numerically, the modification, i.e. the factor in parenthesis, is \((\ldots) \approx 1.380\). Note that the simulated data in fig. 1.1(b) actually are for full dipolar fields as in (1.11) and we correspondingly took \(W'\) of (1.14) for the theoretical curve (yet, in the illustration, we set \(h_{\text{max}} = m/a^3\), as it is basically its order of magnitude which matters).

### 1.2.2 Spin precession in the static case

With the local field distribution \(\psi(H)\) at our disposal, we may readily calculate the average spin polarization \(\langle S_z(t) \rangle\) in the presence of (effectively) static cluster moments, i.e. \(\nu = 0\).

For a time-independent field \(H\), the solution of the precession equation (1.1) is

\[ S(t) = S_{\|} + S_{\perp}^{(1)} \cos(\mu t) - S_{\perp}^{(2)} \sin(\mu t) \]  

(1.15)

with the projections \(S_{\|} = (S(0) \cdot \hat{H}) \hat{H}, S_{\perp}^{(1)} = S(0) - S_{\|},\) and \(S_{\perp}^{(1)} = S(0) \times \hat{H},\) whereby, according to the notation introduced above, \(\hat{H} \equiv H/|H|\) is the unit direction of the field. For the \(S_z\)-component and the initial condition \(S(0) = (0, 0, 1)\), this gives

\[ S_z(t) = (H_z^2/H^2) + [1 - (H_z^2/H^2)] \cos(\mu t) , \]  

(1.16)

which then is to be averaged over the field distribution \(\psi(H)\) of (1.8).

Since the latter depends on \(H\) only, one finds \(\langle H_z^2/H^2 \rangle = 1/3\) and \(\langle H_z^2/H^2 \cos(\mu t) \rangle = 1/3(\cos(\mu t))\). For this last term, we consider

\[ \langle \cos(\mu t) \rangle = -2 \text{Re} \int_0^\infty du u \cos(Wtu) L_{\mu,0}(u) , \]  

(1.17)

which, after a partial integration, yields

\[ \langle \cos(\mu t) \rangle = 2 \left[ 1 + Wt \frac{\partial}{\partial(Wt)} \right] \text{Re} \int_0^\infty du \cos(Wtu)L_{\mu,0}'(u) \]  

(1.18)

\[ = 2 \left[ 1 + Wt \frac{\partial}{\partial(Wt)} \right] \exp \left[-(Wt)^{d/\mu}\right] = \left[ 1 - \frac{d}{\mu}(Wt)^{d/\mu}\right] \exp \left[-(Wt)^{d/\mu}\right] . \]  

(1.19)

Thus, altogether, we obtain

\[ \langle S_z(t) \rangle = \frac{1}{3} + \frac{2}{3} \left[ 1 - \frac{d}{\mu}(Wt)^{d/\mu}\right] \exp \left[-(Wt)^{d/\mu}\right]. \]  

(1.20)

For \(d/\mu = 1,\) i.e. in particular for dipolar fields in 3 dimensions, this relaxation law is known as Lorentzian Kubo–Toyabe function [Kub67], whereas for \(d/\mu = 2,\) which we have
excluded in our treatment (cf. above), it is the Gaussian Kubo–Toyabe function obtained for a Gaussian distribution of local fields. Moreover, it may be interesting to note that laws of type (1.20) have been used in the literature to describe phenomenologically anomalous μSR line-shapes with $d/\mu \neq 1, 2$ (see e.g. [Wu94]). Let us emphasize, however, that our equation (1.20) is an exact result for the considered dimension $d$ and spatial dependence $\sim r^{-\mu}$ of the fields; hence, it should not be confused with an effective ‘power Kubo–Toyabe function’ [Cro97] which serves as a fitting function – as, in the latter reference, for a superposition of (independent) Gaussian and Lorentzian field distributions.

Concerning the properties of (1.20), note that the polarization reaches a plateau value $\langle S_z(t) \rangle = 1/3$ for times $t \gg W^{-1}$, which can be understood from symmetry arguments. Moreover, between the initial $\langle S_z(0) \rangle = 1$ and the plateau, the polarization goes through a minimum at $t = (1 + \mu/d)W^{-1}$, whose origin may also be recognized by simple arguments: For any local field strength $H$, the corresponding $S_z(t)$ of (1.16) exhibits a first minimum at $t = \pi/H$, so that one might naively expect a minimum of $\langle S_z(t) \rangle$ around $t = \langle \pi/H \rangle = 2 \Gamma(1 + \mu/d)W^{-1}$ – which coincides with the exact value for $d/\mu = 1$. Further extrema of $\langle S_z(t) \rangle$ may not be expected due to the dephasing of the contributions with different $H$, which will rather average to the plateau value $1/3$.

Eventually, the relaxation law (1.20) for several combinations of $d$ and $\mu$ is plotted in the following fig. 1.2(a) and (b) (gray lines) where, as may be expected, we find it to describe the simulated data for slowly fluctuating cluster moments ($\nu \ll W$) at early times $\nu t \ll 1$, i.e. before typically (m)any flips of the latter have occurred.

### 1.3 Case of slowly fluctuating cluster moments

#### General aspects of the dynamic case

To motivate our approach for the case of fluctuating cluster moments, which will be developed systematically in this and the following section, let us briefly outline the intricacies the field fluctuations add to the treatment of the spin precession: First, the precession equation (1.1) has to be solved for a certain realization of the stochastic $H(t)$. In fact, only a formal solution may be given, but not an explicit expression which, as an example, could be averaged straightforwardly over all possible realizations of $H(t)$.\(^8\)

As a second difficulty, the characteristics of the fluctuations $H(t)$ themselves will crucially depend on the specific (spatial) configuration of clusters. As a simple example, note that, at a given instant, a single cluster moment close to the spin probe may well generate the same field $H$ as several more distant ones; yet the dynamics of $H(t)$ due to their individual flips, i.e. a sudden turnover vs. (typically) a more gradual change, will be quite different in the two configurations. In formal terms, the system exhibits quenched disorder (cf. Part II of the present work) and, to obtain $\langle S_z(t) \rangle$, one has to respect the correct order of averages – first over different realizations of $H(t)$ in a given cluster configuration, only then over the latter.

---

\(^8\)In the previous expression (1.15), in particular, any cluster moment flip at a time $t'$ requires the projections $S_{1,2}$ and $S_{1,2}^{(1,2)}$ to be re-calculated from $S(t')$ and the new field $H(t')$.\(^9\)
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In practice, any analytical treatment of the problem demands to devise suitably simplified pictures of the process. As an example, one might consider a factor like \(\exp\left[\frac{i}{2} \int_0^t dt' H(t')\right]\) to mimic the spin precession, i.e. describing \(\mathbf{S}(t)\) by a single phase variable. Interesting model calculations and references to different physical problems, which may genuinely be described by the diffusion and relaxation of a phase, may be found e.g. in [Tal01]. In our case, however, such an approach may not be adequate in general, since \(\mathbf{S}(t)\) clearly evolves on a sphere (in three-dimensional spin space) rather than merely on the unit circle in the complex plane.

A common simplification concerning \(\mathbf{H}(t)\), on the other hand, is the so-called 'strong collision approximation' [Kub54], in which it is assumed that the field is effectively renewed (at once and, in some sense, completely; cf. below) with a rate \(\nu\), i.e. the flip rate (1.3) in our system. By the end of the following section, we will explain how a naive adoption of this approximation will fail, whereas it may be employed more successfully as part of an ansatz which, to some degree, takes into account the abovementioned fluctuations of spatial cluster configurations [Uem85]. In fact, it is particularly in this last aspect where we shall go further by our approach.

As a principle, we distinguish between the two cases of slowly or rapidly fluctuating cluster moments, where \(\nu \ll W\) or \(\nu \gg W\), respectively. In both cases, we employ scaling arguments to derive the typical decay rates \(\tilde{\Gamma}\) of the spin polarization which, however, will reveal specific differences for uni- or multiaxial clusters (cf. sec. 1.1) and also for their possible spatial configurations. To tackle the problem of averaging over the latter, we consider subensembles of configurations that are specified by fixing the distances of the clusters closest to the spin probe. Clearly, this concept is motivated by the (typically) dominant influence of the nearest clusters, i.e. the hierarchy implied by the Lévy statistics as discussed in sec. 1.2.1.

For the computer simulations, results of which will be presented below, we note that they closely follow the model definition given at the end of sec. 1.1. Specifically, the radius \(R\) of the spherical (or circular) system is chosen such that it contains \(N = 200\) clusters; and, essentially to exclude numerical complications, we use a very small \(a \lesssim 0.06 n^{-1/d}\) (cf. fig. 1.1(b)). After each time interval \(\Delta t\) drawn from the Poisson distribution \(\psi_{\Delta t}(\Delta t) = N \nu \exp[-N \nu \Delta t]\), a randomly chosen cluster moment is reversed (uniaxial case) or changed randomly to one of its four preferred directions perpendicular to the previous one (cubic-multipolar case). During \(\Delta t\), \(\mathbf{S}(t)\) is evolved according to (1.15) (including the footnote on p. 12). To accelerate the simulations, we often reduced the number of considered clusters down to the \(N' = 10\) ones nearest in an extreme case. We tested that none of the choices for \(N\) or \(N'\), \(a\), and \(R\) had a perceivable influence on the final averaged spin polarization \(\langle S_z(t) \rangle\). For \(N'\), by the way, this confirms the anticipation that the closest clusters are by far most important in the process.

**Multiaxial cluster moments**

We begin our theoretical treatment with the case \(\nu \ll W\) of slowly fluctuating cluster moments. Then, as noted before, \(\langle S_z(t) \rangle\) will be as in the static case (1.20) for times
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![Graphs](image)

**Figure 1.2:** Spin polarization $\langle S_z(t) \rangle$ as a function of $vt$ in the slowly fluctuating case ($\nu/W = 10^{-4}$) for (a) multiaxial and (b) uniaxial cluster moments, and several $\mu$ and $d$. The symbols refer to the simulations and their assignment is the same in both figures. The gray lines refer to the exact result (1.20), while the black lines are fits according to the long-time behaviors (1.21), (1.26). The inset in (a) shows, on a semi-logarithmic scale, the exponential long-time relaxation of $\langle S_z(t) \rangle$ vs. $vt$ that is almost independent of $d$ and $\mu$ (the solid line is drawn as a guide for the eye). The inset in (b) demonstrates the scaling (1.25) for 4 different radii $r_1 \ll n^{-1/d}$, $r_1 = 1.0$ (+), 1.5 (x), 2.0 (o), and 2.5 (e) in the case $\mu = d = 3, n = 0.01$.

$t \ll \nu^{-1}$, which holds irrespective of the type of clusters; see fig. 1.2(a) and (b). Thus, we shall now consider the dynamic regime $t \gtrsim \nu^{-1}$, in which the temporal field fluctuations let the polarization decay further to zero from the previous plateau value $1/3$.

For the assumed $\nu \ll W$, the field $H$ in the relevant cluster configurations has a magnitude $H \gg \nu$ (other configurations have an exponentially small weight). In a time interval of order $\nu^{-1}$ then, the spin precesses many periods around the local field, whereby $S_z(t)$ oscillates around a mean value $\bar{S}_z(t)$. This, of course, is why the static relaxation function (1.20) develops initially, yet also implies that it will be the changes of $\bar{S}_z(t)$ averaged over many realizations of the cluster dynamics which determine the further decay of spin polarization.

In the multiaxial case, significant changes of $H$, which occur in a time of order $\nu^{-1}$, alter the axis of precession and, by the simplest assumption, $S_z(t)$ relaxes with a rate proportional to $\nu$. Hence we expect an exponential decay

$$\langle S_z(t) \rangle \sim \exp \left[ -\text{const} \, vt \right], \quad (1.21)$$

which is indeed confirmed by our simulations shown in fig. 1.2(a).

In this context, it may be instructive to check whether the preceding simple argument – somewhat in the spirit of the strong collision approximation – may yield even some more information on the decay. Thus, one might suppose that $S_z(t)$ gets reduced by a factor $1/3$ following any major change of $H$ (cf. the plateau value) and the probability for $k$ such
changes up to time $t$ should be given by a Poisson distribution; hence (cf. [Uem81])

$$
\langle S_z(t) \rangle \sim \frac{1}{3} \sum_{k=0}^{\infty} \frac{(\nu t)^k}{k!} e^{-\nu t} \left( \frac{1}{3} \right)^k = \frac{1}{3} e^{-2/3 \nu t}.
$$

(1.22)

However, this is not precisely what we find in our simulations, as e.g. the black line in fig. 1.2(a) has a prefactor 0.28 and a coefficient $-0.95$ for $\nu t$ in the exponential. So, to proceed, one should invoke a refined picture, which eventually could involve subensembles of cluster configurations as discussed in the following. For the present work, though, such quantitative agreement for the ‘standard’ exponential decay (1.21) was not our main interest.

**Uniaxial cluster moments**

Surprisingly at first, the case of uniaxial cluster moments is more subtle and exhibits a quite different decay law. The basic reason, in short, is that a cluster with only one easy axis, if it is rather close to the spin probe, to some degree fixes the orientation of the local field $H$—i.e., of course, up to its sign (in case this close cluster moment flips). Mere changes of the sign of $H$, however, do not alter the form of the ‘precession cone’ depicted in fig. 1.1(a) and, accordingly, also leave invariant the short-time average $\langle S_z(t) \rangle$ introduced above.

To see this in more detail, we decompose the field $H$ into the contribution $h_1 = m/r_1^d$ from the nearest cluster at distance $r_1$ and the contribution $H_1$ from the other clusters, $H = h_1 + H_1$. In the subensemble of all cluster configurations with given $r_1$, the variance of $H_1$ is

$$
\langle H_1^2 | r_1 \rangle = C_H h_1^2 \left( \frac{h_1}{W} \right)^{-d/\mu}
$$

(1.23)

with a constant $C_H = S_t/[2(\mu - d)C_W^{d/\mu}]$. For $r_1 \gg n^{-1/d}$, one has $h_1/W \ll 1$, and $H_1$ dominates over $h_1$. In other words, the local field $H$ has important contributions from others than the closest cluster (with different easy axes), and one encounters basically the same physical situation as in the multiaxial case. For small $r_1 \ll n^{-1/d}$, however, $h_1$ is dominant, so that changes $h_1 \rightarrow -h_1$ indeed essentially revert the direction of precession while leaving $\langle S_z(t) \rangle$ unchanged.

In this situation of small $r_1 \ll n^{-1/d}$, the presence of the contribution $H_1$ causes the axis of the field $H$ (irrespective of its direction) to wobble around the $\pm h_1$–axis with the rate $\nu$ and an angular amplitude of order $H_1/h_1$. The wobbling motion together with the much faster precession leads to a diffusive type of motion of $\langle S_z(t) \rangle$ which, in terms of a scaling argument, may be characterized by a ‘diffusion rate’ $\tilde{\Gamma} \sim (H_1/h_1)^2 \nu$. Correspondingly, we will employ an ansatz $\langle S_z(t) | H_1, h_1 \rangle \sim \exp[-\tilde{\Gamma} t]$ for the decay of the spin polarization in presence of the specified $H_1$ (understood as a characteristic value; cf. below) and $h_1$ (as determined by $r_1$), which may also be viewed as a mean-field treatment on the level of the corresponding subensembles. As will be substantiated in the following, this correctly yields an extremely slow long–time decay of the polarization dominated by those spins which are relatively close to one cluster (large $h_1$) and far away from any other clusters (small $H_1$).
1 Anomalous relaxation due to heavy-tailed field distributions

To extract the asymptotic relaxation of the spin polarization, we consider the subensemble of all cluster configurations with fixed distances \( r_1 \) and \( r_2 \) of the nearest and second nearest cluster to the spin probe. Then, extending the above discussion, we may decompose \( \mathbf{H}_1 \) into the contribution \( h_2 \) from the second nearest cluster and \( \mathbf{H}_2 \) from the more distant ones, \( \mathbf{H}_1 = h_2 + \mathbf{H}_2 \), whereby \( h_2 = m/r_2^2 \) and \( \langle H_2^2 \rangle_{r_1} \) satisfies (1.23) with \( h_1 \) replaced by \( h_2 \). Accordingly, for \( r_1 < r_2 \lesssim n^{-1/d}, \) \( \mathbf{H}_1 \) is dominated by contribution \( h_2 \) due to the second nearest cluster moment, which implies in particular \( H_2^2 \sim m^2/r_2^{2\mu} \); whereas for \( r_2 \gtrsim n^{-1/d}, \) on the other hand, we have a dominant \( \mathbf{H}_2 \) and \( H_2^2 \sim W^{d\mu}(m/r_2^\mu)^{2-d/\mu}. \)

Thereby, evidently, the interest in \( H_2^2 \) (or \( \langle H_2^2 \rangle_{r_1, r_2} \)) is because it enters the above rate \( \tilde{\Gamma} \), which together with the corresponding \( h_1^2 = m^2/r_1^{2\mu} \) becomes

\[
\tilde{\Gamma} = \tilde{\Gamma}(r_1, r_2) \propto \begin{cases} 
(r_1/r_2)^{2\mu} & \text{for } r_1 < r_2 \lesssim n^{-1/d}, \\
2^\mu W^{d\mu}(m/r_2^\mu)^{2-d/\mu} & \text{for } r_2 \gtrsim n^{-1/d}.
\end{cases}
\tag{1.24}
\]

From there, the next step is to average \( \langle S_z(t) \rangle_{r_1, r_2} \sim \exp[-\tilde{\Gamma}(r_1, r_2)t] \) over the distribution of second smallest cluster distances, \( \phi_2(r_2 | r_1) = S_{2mr_2^d-1} \exp[-V_{dn}(r_2^d - r_1^d)] \) for \( r_1 \leq r_2 < \infty \), to get the polarization decay \( \langle S_z(t) \rangle_{r_1} \) for the subensembles with only \( r_1 \) specified. According to the distinction of cases in (1.24), we obtain two contributions: One, \( \int_{r_1}^{\infty} dr_2 \phi_2(r_2 | r_1) \exp[-\text{const}(r_1/r_2)^{2\mu} \nu t], \) which decreases exponentially with time for large \( t \) and thus will turn out subdominant; and a second one \( \int_{r_1}^{\infty} dr_2 \phi_2(r_2 | r_1) \cdot \exp[-\text{const}r_1^{2\mu} W^{d\mu}(m/r_2^\mu)^{2-d/\mu} \nu t], \) which may be evaluated by a saddle point approximation.\(^9\) In detail, for the latter to be applicable for all \( \mu > d/2 \), one should first do a partial integration and then change the integration variable from \( r_2 \) to \( 1/r_2 \). Eventually, neglecting power laws in \( t \), we find

\[
\langle S_z(t) \rangle_{r_1} \sim \exp \left[ V_{dn}r_1^d - \text{const} \left( n^{1/d} r_1 \right)^{2\mu} \nu t \right]^{d/2\mu}
\tag{1.25}
\]

for \( \nu t \gg 1 \) (and \( r_1 \ll n^{-1/d} \)). We have verified this prediction for various \( \mu \) and \( d \) by our simulations. One example (for \( \mu = d = 3 \)) is shown in the inset of fig. 1.2(b).

It is important to note that we would not have got this result (and the following) on basis of the mean variance in the \( r_1 \)-subensemble, \( \langle H_2^2 \rangle_{r_1} \) of (1.23), i.e. if we had not considered explicitly the dependence on \( r_2 \). We shall come back on this point in a more general context in the following section.

Finally, the decay of the overall polarization \( \langle S_z(t) \rangle \) is obtained by averaging (1.25) over the distribution of smallest cluster distances, \( \phi_1(r_1) = S_{dm}r_1^{d-1} \exp[-V_{dm}r_1^d] \) (whereby, for consistency, the result must not rely on large \( r_1 \gtrsim n^{-1/d} \) excluded above). We thus obtain

\[
\langle S_z(t) \rangle \sim (\nu t)^{-d/2\mu},
\tag{1.26}
\]

i.e. a slow power law decay which indeed is in marked contrast to the exponential decay in the uniaxial case. Moreover, we may note that, as argued before, cluster configurations

\[^9\text{Explanations and numerous examples of this and other techniques of asymptotic analysis may be found e.g. in [Ben99].}\]
1.4 Case of rapidly fluctuating cluster moments

Continuing our treatment of the spin precession in time–dependent fields \( \mathbf{H}(t) \), we next discuss the case \( \nu \gg W \) of rapidly fluctuating cluster moments. Thereby, we may adopt many elements of the preceding discussion for ‘slow’, uniaxial moments; in fact, the following ‘fast’ case will even be somewhat simpler, as we have to consider only one level in the hierarchy implied by the Lévy statistics, i.e. only subensembles with respect to the smallest cluster distance \( r_1 \). The difference, as we shall see, lies in the properties of the diffusion process which may be invoked to describe the effects of the precession dynamics on the average polarization.

Indeed, for the assumed \( \nu \gg W \), the field \( \mathbf{H} \) in the statistically relevant cluster configurations has a magnitude \( H \ll \nu \) and the spin rotates only by a small angle in a time interval of order \( \nu^{-1} \). This implies that the concept of a mean value \( \tilde{S}_z(t) \) is not useful any longer, since the phase of the precession matters. Accordingly, reorientations of the nearest cluster moment are effective for the spin relaxation both in the case of uniaxial and multiaxial cluster moments.

However, the small angular changes of the spin between major field changes will still lead to an irregular, diffusive type of motion of its \( z \)-component \( S_z(t) \). In a time \( \nu^{-1} \), the angular change is of order \( H/\nu \), which corresponds to a diffusion rate \( \tilde{\Gamma} \sim (H/\nu)^2 \nu \). Similarly as above, the latter may again be inserted in a tentative exponential ansatz \( \langle S_z(t) | H \rangle \sim \exp[-\tilde{\Gamma} t] \) for the polarization decay in cluster configurations where the field (or its variance) has the specified order of magnitude. Let us note that, at this point, one might use a more detailed picture of the (assumed) diffusion process, such as specifically the diffusion on the surface of a sphere (see e.g. \( \text{Ryt87} \)); yet this would not alter our basic scaling arguments, and it is not clear whether one might thus gain substantially more insight (cf. the discussion for the slow multiaxial case in sec. 1.3).

To characterize the local field due to a certain cluster configuration, we decompose it as before, \( \mathbf{H} = \mathbf{h}_1 + \mathbf{H}_1 \). Taking into account the dominant contributions (see (1.23)), we thus find

\[
\tilde{\Gamma} \equiv \tilde{\Gamma}(r_1) \propto \begin{cases} (m/r_1^\mu)^2 \nu^{-1} & \text{for } r_1 \lesssim n^{-1/d}, \\ W^{d/\mu}(m/r_1^\mu)^{2-d/\mu} \nu^{-1} & \text{for } r_1 \gtrsim n^{-1/d} \end{cases}
\]

(1.27)

depending on the smallest cluster distance \( r_1 \). The expected exponential decay \( \langle S_z(t) | r_1 \rangle \sim \exp[-\tilde{\Gamma}(r_1) t] \) is demonstrated in the inset of fig. 1.3 for the example \( d = \mu = 3 \) in the regime \( r_1 > n^{-1/d} \).

In fact, since they exhibit rather small \( H \) (and hence also small \( \tilde{\Gamma} \)), those configurations with large \( r_1 > n^{-1/d} \) may be conjectured to dominate the long–time decay of the overall polarization \( \langle S_z(t) \rangle \). This is substantiated in the averaging of \( \langle S_z(t) | r_1 \rangle \) over the distribution \( \phi_1(r_1) \) of \( r_1 \), which may be done in a calculation similar to that leading to (1.25) in the...
1 Anomalous relaxation due to heavy-tailed field distributions

Figure 1.3: Spin polarization $\langle S_z(t) \rangle$ as a function of $W^2 t / \nu$ in the case of rapidly fluctuating cluster moments [$\nu / W = 10$ (□), 50 (+), and 100 (×) for $d=\mu=3$, and $\nu / W = 10$ for the three other combinations of $d$ and $\mu$]. Data points refer to the simulations and the solid lines are fits according to eq. (1.28). The inset shows the exponential decay of $\langle S_z(t) | r_1 \rangle$ and the scaling as discussed in the text for 4 different radii $r_1 \gtrsim n^{-1/d}$, $r_1 = 6.5$ (+), 7.0 (×), 7.5 (⋆), and 8.0 (○) in the case $\mu = d = 3$, and $n = 0.01$ (the solid line is drawn as a guide for the eye).

previous section. In particular, the dominant contribution (due to the second line in (1.27)) may be evaluated asymptotically for large times by means of a (in this case straightforward)
saddle point approximation, which yields

$$\langle S_z(t) \rangle \sim \exp \left[ -\text{const} \left( \nu^{-1} W^2 t \right)^{d/2\mu} \right].$$

(1.28)

In fig. 1.3, both the scaling with $(W^2 t / \nu)$ and the stretched exponential decay with the exponent $d/2\mu$ are confirmed by our computer simulations for the same $\mu$ and $d$ values as in fig. 1.2.

Thereby, it should be kept in mind that our picture of the precession in the fast case $\nu \gg W$ was developed for the prevailing configurations with $r_1 \gg (m/\nu)^{1/\mu}$. For the rare instances of $r_1 \ll (m/\nu)^{1/\mu}$, by contrast, one has $h_1 \gg \nu$, i.e. a situation as in the case of slowly fluctuating clusters moments. While this merely gives an exponential, subdominant contribution to $\langle S_z(t) \rangle$ for multiaxial clusters, the stretched exponential decay (1.28) will, for uniaxial clusters, at long times be masked by the much slower power law decay (1.26) precisely due to those rare configurations with $h_1 = m / r_1^\mu \gg \nu$.

Comparison to other approaches

Looking back at our treatment, let us re-emphasize the importance of properly taking into account the Lévy statistics for the field together with the quenched disorder of spatial cluster configurations. Concerning this, it may be instructive to consider explicitly the failure of a too direct, mean field type application of the ‘strong collision approximation’,
1.4 Case of rapidly fluctuating cluster moments

which was already pointed out by Uemura et al. ([Uem85]; cf. below). Within such a
simplistic description, one might assume that the field $\mathbf{H}$ at the probe site is drawn anew
from the overall distribution $\psi(\mathbf{H})$ in (1.8) with the rate $\nu$. By scaling arguments similar to
those used above, one would then be led to an exponential ansatz $\langle S_z(t) \rangle \sim \exp(-\tilde{\Gamma}_{mf} t)$ for
$\nu \gtrsim 1$ both in the cases of slowly and rapidly fluctuating cluster moments and irrespective
of whether the clusters possess only one easy axis or more.

In the slow case $\nu \ll W$, in particular, one would indiscriminately get $\tilde{\Gamma}_{mf} \propto \nu$ by
essentially the same argument as for the multiaxial clusters in the preceding section (cf.
(1.21)), although, as we have seen, this does not apply to uniaxial clusters. In the fast case $\nu \gg W$, in brief, the distribution of the ‘diffusion length’ $H/\nu$ as introduced above would exhibit the heavy tail (1.9) of the field distribution. Accordingly, a spin’s mean square displacement after time $t$ should be dominated by the largest $H = H_{\max}(\nu t)$ encountered during the corresponding $\nu t$ field renewals, a typical value of which may be estimated by
\[
\int_{H_{\max}} H \, 4\pi H^2 \psi(H) \sim (\nu t)^{-1}\]
cf. sec. 1.2.1 and the references given there. With the thus obtained $H_{\max} \sim (\nu t)^{1/d} W$, the scaling of the characteristic depolarization time $\tilde{\Gamma}_{mf}$ may be
determined by $(\nu/\tilde{\Gamma}_{mf})^{2\mu/d}(W/\nu)^2 \sim 1$, which eventually yields $\tilde{\Gamma}_{mf} \sim \nu(W/\nu)^{d/\mu}$ in obvious
discrepancy to our actual result (1.28).

Hence, we may conclude that, as a minimum requirement within our approach, the ‘most
specific’ element in the envisaged rate $\tilde{\Gamma}$, i.e. $H$ in the fast and $H_t$ in the (uniaxial) slow
case, should be decomposed into the contribution from the next nearest cluster and that
from the still further ones in order to eventually give the correct statistics.

As mentioned above, Uemura et al. [Uem85] tackled the problem (for $\mu = d = 3$) by a
somewhat different approach. Thereby, the field fluctuations in a given spatial configuration
of field sources are simply assumed to be Gaussian distributed, yet with a configuration
dependent standard deviation $\Delta$. The distribution of the latter, $\rho(\Delta)$, is then not derived
directly from the spatial distribution of the field sources, but determined self-consistently
by requiring that the superposition of the Gaussians yields the correct Cauchy distribution
of the Cartesian components $H_\alpha$ of the local field. Referring to our remark on the $H_\alpha$ in
sec. 1.2.1, we may note that this actually is not fully consistent with the three-dimensional
Lorentzian (1.10), which would require a slightly different $\rho(\Delta)$. In any case, averaging over
the standard deviations $\Delta$ eventually yields the root-exponential decay of $\langle S_z(t) \rangle$ implied by (1.28) for $\mu = d = 3$, which the aforementioned authors predicted specifically for a
dynamic regime $\nu \gtrsim 20 W$.

Finally, let us also mention an earlier approach [Hay79] to describe $\mu$SR based on the
strong collision approximation, in which the temporal evolution of the probability distribu-
tion $p(S, H, t)$ to find a certain $S$ and $H$ at time $t$ is considered in a kind of master
equation. Although this is done explicitly for a Gaussian $H(t)$, it may be of interest in
the present context, since it directly involves the precession equation (1.1). Hence, when
combined with the Lévy statistics and proper subensemble concepts, it might serve as a
starting point for an attempt to go beyond our ‘diffusion description’ of the spin dynamics.

\footnote{In fact, one could imagine that the sketched description applies to a mobile spin probe undergoing some
fast hopping process (over distances $\gg n^{-1/d}$) through the sample – which, however, is clearly different
from our problem of spins in definite fixed locations.}
1.5 Applications to experiments

In the following, we shall be concerned with practical applications of the results from the preceding sections. Thereby, the discussion will be split in two: In the present section, we will consider immediate applications of our model to μSR measurements in random assemblies of superparamagnetic particles, which we took as primary experimental reference in sec. 1.1. Then, within the subsequent summary chapter, we shall also address possible generalizations and modifications of our approach, which may allow to make use of our findings in further of the many instances of spin precession in a stochastic environment described in the introduction.

A concrete example

To demonstrate the direct applicability of our results, let us take a specific example. Fig. 1.4 shows the μSR asymmetry (cf. sec. 1.1) in a sample of single-domain cobalt clusters in a copper matrix as measured by Bewley et al. [Bew98]. As obtained at a temperature $T = 2.4 \text{K}$, this asymmetry $a(t)$ is linked to the static Kubo–Toyabe function $\langle S_z(t) \rangle$ of (1.20) (with $\mu = d = 3$) by the relation

$$ a(t) = a_1 \langle S_z(t) \rangle + a_{bg}, $$

where $a_1$ is an experimental constant (‘initial asymmetry’) and $a_{bg}$ accounts for the time- and temperature-independent background signal (associated with muons stopping outside the sample). The background contribution was measured in the absence of the sample to be $a_{bg} = 0.021$. In their ‘best fit’, though, the above authors used $a_1$, $W$ and $a_{bg}$ as fitting parameters, thus getting a value of $W = 6.8 \mu s^{-1}$ in moderate agreement with some expensive numerical modeling carried out, among other purposes, to estimate $W$. Thereby, besides, Bewley et al. also recognized the strongly decreasing influence of 1st, 2nd, 3rd, . . . nearest neighbor clusters in correspondence to what we identified as Lévy hierarchy in sec. 1.2.1.

According to our theory, $W$ for the full dipolar interaction is given by (1.14), which gives $W' \simeq 4.54 \gamma \mu_0 m n / 4 \pi$ in standard SI units (and with the integral and initial prefactor evaluated). Closely following the statements given in the preceding reference, we calculated $m n$ by taking $1.7 \mu_B$ ($\mu_B$ is the Bohr magneton) as magnetic moment per Co atom and a density of $0.02 \cdot (1 - 0.15)$ Co atoms among all atoms (including 15% of the Co atoms in the Co$_2$Cu$_{98}$ sample dissolved in the copper matrix and not forming clusters). For the number density of Cu atoms we took a value of $n_{Cu} = 8.45 \cdot 10^{28} \text{m}^{-3}$ (see e.g. [Kit71]).

With the gyromagnetic ratio of the muon $\gamma \simeq 851.5 \text{MHz/T}^{-1}$ as mentioned in sec. 1.1, we finally get a value of $W \simeq 8.76 \mu s^{-1}$. An unbiased least squares fit to the above $a(t)$ with merely $a_0$ and $W$ as fitting parameters, but $a_{bg} = 0.021$ fixed to the above, independently measured value, yields $W \simeq 8.77 \mu s^{-1}$, which thus is in excellent agreement with our theory. For a graphical comparison to the raw data, both the the original authors’ and our fitted curves are shown in fig. 1.4(a).

At higher temperatures, when the fluctuations of the cluster moments become important, the situation in the considered material is slightly complicated by the Co and Cu nuclear
moments offering an additional independent ‘channel’ of muon spin relaxation. Hence, our results from sec. 1.4, i.e. in particular (1.28), do not apply directly. Still, we may note that, in their specifically adopted analysis, the above authors successfully rely on the assumption of a root–exponential polarization decay due to the cluster moments.

Proposals for new experiments

Besides being of use for the analysis of existing experiments, our work also allows us to make new predictions. As an example, one could test for dimensionality effects in the anomalous relaxation laws by depositing superparamagnetic cluster onto a surface (or by embedding them in separated layers within a bulk material). Both the static and the dynamic situation will be accessible by tuning $W$ via the cluster concentration $n$ and by tuning the typical cluster reorientation rate $\nu$ of (1.3) via the temperature $T$.

As a specific example, for the multiaxial iron nanoclusters studied in [Jac00a], one finds for the attempt frequency $\nu_0 \simeq 10^9 \text{s}^{-1}$ and for the anisotropy energy $E_a \simeq 51$ K. In that experiment, $W \simeq 7 \cdot 10^5 \text{s}^{-1}$ (for 0.1% volume fraction iron in the silver matrix), implying a crossover from the static to the dynamic situation around a temperature $T_B = E_a / \ln(\nu_0/W) \simeq 7$ K (cf. sec. 1.1), which obviously is in a range accessible in experiments.

Even so, one may ask whether the interesting dynamical regimes characterized by anomalous relaxation laws, which are clearly observable in the computer simulations, will also be accessible in practical $\mu$SR measurements. In fact, with respect the range of time scales, this should not be a problem, as there are even not so recent examples of $\mu$SR measurements covering about 3.4 decades in time (see e.g. [Uem85]), whereas according to our simulation
1. Anomalous relaxation due to heavy-tailed field distributions

results in figs. 1.2 and 1.3, much less should suffice to identify the predicted regimes – provided that these are shifted as explained to match the experimental time window.

With respect to the range of \( \langle S_x(t) \rangle \) values, on the other hand, it will undoubtedly be very difficult to resolve the decay down to such low values as displayed – basically for demonstration purposes – e.g. for the case of rapidly fluctuating moments in fig. 1.3. Again, however, the anomalous relaxation may already be identified from a smaller, well accessible portion of the data. To demonstrate this explicitly, we have re-plotted in fig. 1.4(b) one of the data sets of fig. 1.3, this time using \(- \log (\langle S_x(t) \rangle)\) as ordinate in the double logarithmic diagram. Thereby, the asymptotic stretched exponential law (1.28) appears as a straight line with slope \( \mu/2d \), which may well be observed for a remaining polarization \( \langle S_x(t) \rangle \geq 10\% \) (taken as an exemplary value). Accordingly, it should still be possible to identify the anomalous exponent even if in a \( \mu \)SR experiment the statistics of the measured values cannot be improved to resolve very low values of \( \langle S_x(t) \rangle \).

Obviously, much more could be said on such practical issues, the proper choice of materials and their parameters, possible experimental settings etc. In any case, the preceding discussion should have made clear that there are – both already existing and possible new – interesting applications of our theoretical findings. So, as an important general aspect, let us eventually point out the flexibility of our approach based on comprehensible scaling arguments, by which it may be adapted to the experimental situation at hand.

As an example, for a (quasi-)uniform distribution of muon stopping sites in the sample, the abovementioned layered material (containing the magnetic clusters in separate parallel planes) does not correspond exactly to the case \( d = 2 \) as considered in our approach. However, without going into details, we note that our treatment may readily be adjusted to such a special arrangement of field sources by including in the scaling arguments the ratio of thicknesses of ‘magnetic’ and neutral layers, depending on which the relaxation should incorporate some characteristics of both cases \( d = 2 \) and 3. In this context, let us mention some recent experimental progress in the availability of moderated ‘ultra-low’ energy muons [Mor04], which possess a quite precisely tunable penetration depth in the sample. The technique, which has been used e.g. to measure the penetration of a magnetic field into a superconductor [Jac00b], may also open novel possibilities for the study of materials as considered here.

Thereby, clearly, our modeling of the sample material bears some idealizations which, in general, may not be overcome as straightforwardly and may eventually restrict the applicability of our findings, namely the neglect of interactions between the field sources as well as their possible variation in size (or strength). Thus, as announced above, we shall review these assumptions and tentatively survey potential generalizations as well as limitations of the present treatment within the following summary chapter.
2 Summary of Part I

Synopsis
The subject of Part I of the present work is the spin precession in disordered systems of randomly distributed and eventually fluctuating magnetic field sources, i.e. in particular the anomalous spin relaxation due to the heavy-tailed local field distributions shown to emerge quite naturally in that type of system.

At the beginning of chap. 1, we presented the precession of a spin probe in its local field $H$ as central element of various experimental techniques. We noted the classical treatment of the spin dynamics and identified as conceptually simple, yet fundamentally interesting problem the decay of the initial polarization of an ensemble of independent spins in the presence of randomly distributed field sources, whose fields decreasing as $r^{-\mu}$ with distance $r$ give rise to the abovementioned type of local field distributions for exponents $\mu > d/2$ (with $d$ being the system’s dimension). Thereby, we chose muon spin relaxation ($\mu$SR) in random assemblies of superparamagnetic clusters as guiding experimental example, which was portrayed in some detail in the following.

In sec. 1.2, we calculated exactly the broad, Lévy type local field distribution in our model system. In particular, we also derived the width $W$ of the corresponding Lorentzian distribution in $d = 3$ for full dipolar fields, i.e. including both their secular and non-secular parts, and we discussed potential modifications due to finitely sized (as opposed to idealized point-like) clusters as field sources. Further, we deduced the polarization decay $\langle S_z(t) \rangle$ for static local fields, thereby generalizing the well-known Kubo–Toyabe function for variable $\mu$ and $d$.

In the two subsequent sections, the more subtle dynamic case, in which cluster moments reorient with a (thermally activated) rate $\nu$, was tackled based on scaling arguments extracted from specifically adapted ‘effective’ pictures of the precession dynamics. Motivated by the Lévy statistics of the fields, we considered subensembles of spatial cluster configurations defined with respect to the distances of the clusters closest to the spin probe, which, owing to this type of statistics, dominate $H$. This, in fact, was not only demonstrated as a proper way to take into account the quenched disorder in the system, but turned out an essential prerequisite for the applicability of mean field type arguments.

Thus, refining and largely broadening previously known results, we postulated and verified by computer simulations a rich anomalous relaxation scenario with characteristic exponents depending on the ratio $d/\mu$. In particular, for slowly fluctuating cluster moments ($\nu \ll W$), the long-time decay of $\langle S_z(t) \rangle$, which follows an initial behavior as in the static case, exhibits a remarkable dependence on the symmetry characteristics of the field sources, namely a
simple exponential decay for multiaxial clusters vs. a slow power law decay for clusters with only one easy magnetization axis. This power law may also become visible at very long times in the decay of \( \langle S_z(t) \rangle \) due to rapidly fluctuating moments (\( \nu \gg W \)), which is otherwise given by a stretched exponential law irrespective of the type of clusters.

In sec. 1.5, finally, it was demonstrated that our findings may readily be applied to experiments. For a recent \( \mu \)SR measurement in a superparamagnetic cluster system as mentioned above, we found our prediction for the width \( W \) of the local field distribution in very good agreement with the value obtained from the relaxation data (corresponding to the static case \( \nu \approx 0 \)). Moreover, we also suggested new experiments for that class of materials, whereby we gave some detailed arguments as to why and how it should be possible to observe e.g. the predicted dimensionality effects (i.e. the varying exponents) of the anomalous relaxation laws. After all, we pointed out the potential adaptability of our approach, which relies on rather simple and transparent scaling arguments, to specific experimental conditions – such as an arrangement of the clusters in separate parallel layers.

Possible extensions and limitations of the present approach

Taking up the last-mentioned point, we note that our treatment may likewise be generalized to situations where an additional external field \( H_{\text{ext}} \) is present, which generically may be either parallel or perpendicular to the \( z \)-direction (so-called longitudinal or transversal field \( \mu \)SR, respectively; see e.g. [Lee99]). Obviously, complications for our approach arise when the external field is so strong (and applied sufficiently long) that the orientation of the cluster moments may not be taken random any more or, for the dynamic case, when their flip dynamics is altered substantially.\(^1\) Moreover, the analytic evaluation of some of our results, as far as it relied on isotropy arguments in ‘\( H \)-space’, may be ramified.

In fact, in presence of an external field the \( \mu \)SR experiment bears a number of parallels to a measurement of the transverse relaxation in NMR probes. So it may be interesting to note that stretched exponential relaxation laws similar to our result (1.28) were derived for NMR in disordered systems (such as structural glasses) by averaging over a distribution of Bloembergen–Purcell–Pound type relaxation rates [St68;4].

As pointed out before, an idealization in our treatment consists in considering point-like clusters as field sources with a unique strength \( m \). Concerning the clusters’ spatial extent as such, we already argued in sec. 1.2.1 that crossover effects to a Debye-like relaxation behavior typical for Gaussian processes should be of minor importance as long as the cluster sizes \( a \) are much smaller than the mean distance \( n^{-1/d} \). Moreover, any variance in the strength of the magnetic moments \( m \), as genuinely associated with differently sized clusters, neither changes the local field distribution nor – by implication – the relaxation in the static case, except that \( m \) formally has to be replaced by its mean value.

In the dynamic regime, however, a broad distribution of cluster sizes may require a refined analysis, in which subensembles should be defined by both the nearest cluster distances and the relevant cluster sizes. Put differently, the dominant contributions \( h_{1,2} \) and \( H_{1,2} \) to the

\(^1\)While details will depend on the precise reorientation dynamics of the moments (see e.g. [Dor97]), one may, for a first estimate, compare \( \mu_0 H_{\text{ext}} m \) (in SI units) to the anisotropy energy \( E_a \).
local field, which enter our basic scaling arguments, then should be analyzed not only with respect to the distances \( r_{1,2} \), but also regarding possible values of \( m \). Clearly, this may as well include individualized time scales \( \nu^{-1} \), although, for superparamagnetic clusters, it should be noted that the anisotropy energy \( E_a \) determining the flip rates may not in all cases scale simply with the cluster volume and hence \( m \) (cf. [Bew98]).

Another so far neglected aspect, which may become important in particular for the field dynamics, are interactions between the cluster moments. In fact, at high temperatures \( T \), these interactions may be accounted for by an effective, temperature dependent width \( W = W(T) \) of the local field distribution (1.8) (for an approximate calculation in \( \mu = d = 3 \), see [Hel75]). At low temperatures \( T \), by contrast, the cluster dynamics cannot be described any longer by a Poisson process with rate \( \nu \). For dipolar systems in \( d = 2, 3 \), this occurs for \( T \lesssim 2 m^2 n^{d/3} \) [Rin98a, Höh02]; e.g. in the materials studied in [Bew98, Jac00a] this would be below 1K. In this low-temperature regime the problem becomes more difficult and the above relaxation laws may no longer hold true. Prominent examples of materials studied by \( \mu \)SR, whose inner dynamics is generally non-Poissonian, include spin glasses [Ker96, Cam99] and related disordered magnetic systems, which may e.g. exhibit nontrivial sequential ordering transitions of longitudinal and transverse components of their magnetic moments [Lie01].

Clearly, the description of such complex interacting systems is a challenging field of study in its own right. Corresponding attempts for distinctly interacting (i.e. less dilute) assemblies of superparamagnetic particles are mentioned in sec. 3.1.3 in Part II of the present work. Indeed, as explained there, the sophisticated non-equilibrium phenomena in spin glasses at low temperatures may in some sense be viewed as an ‘ultimate’ instance of nontrivial dynamics in a strongly interacting disordered system. We shall be concerned with that type of spin dynamics, albeit in a rather qualitative, abstracted model, in the following Part II.

For the present, let us point out that elements of our preceding approach, namely the field distribution and the subensemble concept, may be of use, e.g. as first approximations, for the treatment of interacting cluster systems or some analogous problems in dielectric materials – such as the stochastic relaxation in an orientational glass [Kog00] or the ‘nearly constant loss’–behavior of (disordered) ionic conductors (cf. [Maa06]; note also that the above temperature estimate was taken from simulation studies [Rin98a, Höh02] concerned with this subject).

**Outlook**

From the preceding discussion, we hope that our findings will stimulate further research on the challenging problem of spin precession in disordered systems. Besides rather direct applications as proposed in sec. 1.5, this includes further development of our approach to describe \( \mu \)SR or related dynamical probes also in other complex systems. Thereby, from a theoretical point of view, it should be particularly interesting to consider local fields \( \mathbf{H}(t) \) exhibiting both a broad spatial distribution and some more sophisticated, eventually long-range temporal correlations, examples of which may be given by the abovementioned
interacting systems. In any case, the suggested scaling methods should potentially give deeper insight into mechanisms of the spin relaxation, thus helping to exploit the information from the spin probes on the inner dynamics of the studied materials.
Part II

A real space renormalization group approach to spin glass dynamics
3 Aging, rejuvenation and memory effects in systems far from equilibrium

3.1 Phenomenology

3.1.1 Spin glasses

A spin glass may be defined as a disordered system of spins (i.e. magnetic moments) with mixed (ferro- and antiferromagnetic) interactions, which below a well-defined temperature $T_c$ undergo a cooperative transition to a highly irreversible, metastable disordered frozen state [Myd93]. We shall elucidate the elements of this concise description in the following.

At this point, let us note that basic facts about spin glasses are discussed comprehensively in earlier reviews of the field [Bin86, vHe87] as well as textbooks [Fis91, Myd93], where the last-mentioned covers mainly experimental (material) aspects. As far as such concepts are presented without further notice in the following, reference to these works should be understood.

Spin glass materials

Typical spin glasses consist of magnetic atoms, i.e. atoms providing a net electronic (spin and orbital) angular momentum and a corresponding magnetic moment, which on random positions replace some fraction $x$ of the atoms of an essentially nonmagnetic host material. Prime examples (so-called ‘canonical’ spin glasses) are binary metallic alloys like Cu$_{1-x}$Mn$_x$ or Au$_{1-x}$Fe$_x$ (with orders of magnitude $x \approx 10^{-2}$ and $T_c \approx 10$ K), cf. below.

‘Good moment’ metals like Mn, Fe, Gd, and Eu are from the group of transition and rare earth elements with partly filled 3$d$ and 4$f$ shells, respectively. Of course, their actual magnetic moment depends crucially on the interaction with the host material into which they are built. In particular, an anisotropic crystalline environment may give them an effective Ising character (see sec. 3.2).

As for interactions among the moments, the magnetic dipolar interaction is usually negligible for single moments on an atomic scale [Ash76]. Instead, ‘exchange interactions’ dominate by effectively linking relative spin orientations to the atoms’ electronic orbital state (i.e. Coulomb interactions) through the symmetry requirements for the overall multiparticle wave functions.

Based on the overlap of wave functions, the range of direct exchange is limited to neighboring atoms. Most important in insulating (or semiconducting) spin glasses is the so-called
superexchange, whereby in a sort of covalent mixing an intervening host atom may effectively pass on the spin polarization of one magnetic atom to another. A typical scenario is the (anti-)ferromagnetic coupling of (next-)nearest neighbor moments, which, when both are combined, may yield the abovementioned mixed interactions.

In metallic spin glasses, there is the stronger and much more long ranged Ruderman–Kittel–Kasuya–Yoshida (RKKY) interaction mediated by the sea of conduction electrons [Rud54]. These get (spin) polarized by the presence of a magnetic moment, which in turn is sensed by other moments. The polarization can be calculated in a linear response formalism [Whi83], whereby the corresponding susceptibility exhibits an oscillatory algebraic decay for large distances $r$ from the magnetic perturbation [Fet71],

$$
\chi_{\text{electr.}}(r) \sim \frac{\cos(2k_F r)}{k_F r^3}, \quad k_F r \gg 1
$$

(\(k_F\): Fermi wave vector).\(^1\) We note that this asymptotic behavior stems from the (virtual) discontinuity of the electrons’ Fermi distribution at the Fermi surface and is in complete analogy with Friedel oscillations of the charge density surrounding a charged perturbation. For spins $\mathbf{S}_{i,j}$ at a distance $r_{ij}$, (3.1) yields an interaction $E_{\text{RKKY}} = -J(r_{ij})\mathbf{S}_i \cdot \mathbf{S}_j$ with $J(r) \propto \chi_{\text{electr.}}(r)$.

An important point is that the mixed interactions together with the moments’ random positions naturally lead to conflicting orientational preferences for some of the moments. This so-called frustration [Tou77] proves essential for some of the ‘glassy’ aspects of spin glasses in the following.

Today, there are some hundred materials known which fit the description given so far. Somewhat special are random-bond systems, where two types of moments fully occupy the sites of an ordered lattice, but interactions are random due to the irregular alternation of both on these sites, e.g. Fe and Mn in Fe$_{1-x}$Mn$_x$TiO$_3$. Notable are also amorphous metallic spin glasses, where the randomness is due to the non-crystalline state of the material rather than to diluted moments. As for the latter, limited miscibility may in fact lead to clustering of the moments, and one may end up e.g. with a system of superparamagnetic particles as treated in Part I of this work rather than with a (classical) spin glass.

Similarly, at too high concentrations $x$, large effective moments formed out of (accidentally) neighboring magnetic atoms may dominate the magnetic behavior in what is then called a micromagnet or cluster glass. Eventually, (anti-)ferromagnetic ordering may percolate through the system. In the so-called re-entrant spin glasses also mentioned as examples in Part I, one longitudinal spin component is supposed to order ferromagnetically before freezing of transverse components yields a disordered state at lower temperature.

Remarkable at the lower end of concentrations $x$ delimiting spin glass behavior is the Kondo effect (in metals), which below a temperature $T_{\text{Kondo}}$ lets effectively disappear magnetic moments through a firm screening by oppositely polarized conduction electrons (see e.g. [Ash76]).

\(^1\)This (nevertheless prototypical) expression is actually for free electrons and $T = 0$. 
3.1 Phenomenology

Figure 3.1: In phase magnetic susceptibility $\chi'(\omega)$ as a function of temperature $T$ measured in the metallic spin glass Cu$_{1-x}$Mn$_x$ with $x = 9.4 \cdot 10^{-3}$ at probing frequencies $\omega = 1.33\,\text{kHz}$ (□), 234Hz (○), 10.4Hz (×), and 2.6Hz (∆). The cusp with the slight $\omega$–dependence displayed in the inset is characteristic of the freezing transition. From [Mul81].

Spin glass transition

At its critical temperature $T_c$, a spin glass undergoes a phase transition from the high-temperature paramagnetic phase to a disordered frozen state in the so-called spin glass phase.

Thereby, similar to the phase transition of a ferro- or antiferromagnet, the spins get locked in fixed directions, which in a spin glass, however, display no regular pattern. Consequently, the average magnetization\footnote{Note that for convenience in the following theoretical treatment, we consider as ‘magnetization’ the dimensionless spin polarization.} $M = 1/N \sum_i S_i$ as well as any ‘staggered’ magnetization

$$M(k) = \frac{1}{N} \sum_i e^{-i k r} S_i \quad (3.2)$$

($N$: number of spins, $k$: arbitrary wave vector) vanish, as demonstrated e.g. by the absence of magnetic Bragg peaks in neutron scattering. We will come back to the question of what might then serve as order parameter of the transition in sec. 3.2.

Experimentally, the first hint of the transition, which marks the nascent of spin glass physics in the early 1970’ses, was a cusp in the magnetic susceptibility as shown in fig. 3.1. Actually, its location turns out frequency dependent, so $T_c$ is defined by the static limit. The heat capacity, by contrast, displays a broad maximum somewhat above $T_c$.

An important feature of spin glasses is the onset of sophisticated remanence effects below $T_c$. As an example, when cooled below $T_c$ with an applied magnetic field (so-called field–cooling or fc), the spin glass displays a nearly temperature independent magnetization $M_c$.
which is constant in time, however decays extremely (and, in fact, more and more) slowly upon removal of the field. When the field is applied only after cooling to the spin glass phase (zero-field-cooling, zfc), the magnetization $M$ is lower and increases to its fc value $M_{fc}$ while the sample is reheated to $T_c$. If the temperature $T < T_c$ is left constant instead, $M$ approaches $M_{fc}$ only (increasingly) slowly.

All of this indicates that the spin glass phase is not a ‘simple’ frozen state with the sole peculiarity of being disordered, but entails complex dynamics encompassing a very wide range of time scales. In some aspects, this is reminiscent of structural glasses and thus qualifies a spin glass as a glassy system, cf. below.

The spin glass transition may be studied by further experimental techniques, including $\mu$SR as described in Part I of this work. In accordance with most of current research, our interest in the following will mainly be in the rich non-equilibrium phenomena deep in the spin glass phase, which are introduced in the next subsection(s), rather than in the transition and critical dynamics around $T_c$.

3.1.2 Non-equilibrium effects in the spin glass phase

Aging

Spin glasses, like almost all glassy systems, display so-called aging [Vin96, Bou98]: Important properties of these materials such as susceptibilities are not constant in time as one might expect, but depend on the age of the system, i.e. the time that has passed since the transition to the glassy state. Typically, what is observed following a quench below the critical temperature is a continuous slowing down of the dynamics on all time scales; in spin glasses this extends from microscopic times $\tau_0 \simeq 10^{-11}$ s up to the largest time scales $\sim 10^6$ s accessible in experiments.

As an example, fig. 3.2 shows results from a recent experiment by Hérisson and Ocio [Hé02], who for the first time could measure directly the spin (magnetization) autocorrelation function

$$C(t, t') = \frac{1}{N} \sum_i \langle S_i(t' + t) S_i(t') \rangle$$

($\langle \ldots \rangle$: thermal average, cf. sec. 3.2) in a spin glass. The sample is cooled rapidly below $T_c$ and subsequently left at rest at fixed temperature for a certain waiting time $t'$ before the actual measurement starts. As may clearly be seen, the behavior of the autocorrelation function depends strongly on the waiting time: $C(t, t')$ decays increasingly slowly for larger $t'$ and, in particular, keeps changing even for large $t'$, all of which reflects the slowing down of the spin dynamics.

More common and convenient in experiments (as opposed to theory or simulations, see below) are measurements of the magnetic susceptibility $\chi(\omega)$ or, with similar information content, of fc or zfc magnetizations. Not merely in spin glasses, the aging quantity may in general be decomposed into a stationary and an aging contribution [Bou98, Rin02],

$$C(t, t') = C_{\text{st}}(t) + C_{\text{ag}}(t, t') ,$$

(3.4)
where, as indicated, the first is independent of the waiting time, whereas the second often obeys a scaling relation

\[ C_{\text{eq}}(t, t') = f \left( \frac{t}{\tau(t')} \right) \quad \text{with e.g. } \tau(t) \sim t^\mu, \mu \leq 1, \]

in which \( f \) is a scaling function and the power law behavior for the 'effective' waiting time \( \tau(t') \) is meant as a common example.

**Rejuvenation**

Fig. 3.3 shows the imaginary part (dissipation) \( \chi''(\omega) \) of the magnetic susceptibility measured for the same spin glass material as in fig. 3.2. After an initial quench to \( T = 12 \text{ K} \), \( \chi'' \) (actually \( \chi''(\omega, t) \)) displays aging as described above.

However, following a slight decrease of temperature to \( T = 10 \text{ K} \) at a time \( t_1 \), surprisingly, \( \chi'' \) does not simply continue to decrease – possibly with a modified velocity – as one might expect. Instead, aging almost starts anew, i.e. the system behaves as if a much shorter time had passed since its transition to the spin glass state. In this sense, it seems 'younger', hence the term rejuvenation for this effect.

In other words, the started (fragmental) thermalization at one temperature proves incompatible to that at another temperature. A possible conjecture would be that in the spin glass phase, the target equilibrium states (or at least the interim states showing up on experimental time scales) at different \( T \) are not simply related, i.e. they 'look' to each other much like the disordered phase from above \( T_c \). This sets spin glasses apart from sim-
Figure 3.3: Out of phase magnetic susceptibility $\chi''(\omega, t)$ vs. time $t$ measured at the indicated temperatures and probing frequency $\omega = 0.01$ Hz in the same type of spin glass as in fig. 3.2. In the inset, the data from the intermediate stage $t_2$ at $T = 10$ K is left out to demonstrate the seamless continuation of aging at $T = 12$ K. From [Vin96].

pler aging systems as e.g. ferromagnets, which at all temperatures below their $T_c$ (besides different densities of thermal excitations) evolve towards a common ground state.

Memory

When in the measurement in fig. 3.3, after an additional time $t_2$, the temperature is brought back to the initial 12 K, there is another surprise. This second temperature shift does not lead to another rejuvenation step similar to the first one. Instead, the system ‘re-ages’: It seems to have kept a kind of memory of its evolution at this temperature during $t_1$ and aging continues as if the period $t_2$ at another temperature had not been there, see the inset of fig. 3.3. In fact, the latter is a bit of an exaggeration (related to time resolution), since in general there may be a short transient.

Even more spectacular are ‘memory dip’ experiments [Jon98, Miy01]: When recording $\chi''$ while cooling the spin glass below $T_c$ at some constant rate $r = dT/dt$, one gets a steadily decreasing curve $\chi''(T)$. If the cooling is interrupted for some time at up to 4-5 intermediate $T_i$, $\chi''$ drops (ages) during each interruption, but upon resumption of the cooling slightly rises again to join the $\chi''(T)$ curve of uninterrupted cooling (rejuvenation). Now, when this sample is continuously reheated at rate $-r$, $\chi''(T)$ nevertheless exhibits dips at the $T_i$ resembling those during the preceding interrupted cooling, which demonstrates even multiple memory (to several temperatures).

However, involving cooling rate effects, the last-mentioned experiments are less simple to analyze theoretically (cf. chap. 7). We note that rejuvenation and memory effects may
also be induced by changes of external control parameters other than the temperature as well as by changes of bonds (interactions) in theoretical models, see below.

Clearly, all three phenomena depicted above, aging, rejuvenation and memory effects, hint to the complex nature and sophisticated dynamics of the spin glass phase and call for a consistent theoretical explanation, a contribution to which is the aim of the present Part II of this work. More details of experimental findings will be given as we compare them to results of our theoretical approach in the following chapters.

### 3.1.3 Examples from other systems

The essential prerequisites of spin glass behavior – disorder and frustration due to competing interactions – are not unique to interacting single magnetic atoms, but may also be found in a wide variety of disordered systems of different interacting entities, e.g. electric dipolar or quadrupolar particles [Myd93, Alb98], or even small superconducting grains (coupled via Josephson junctions) [Joh85].

Spin glass–like freezing is under discussion for disordered ensembles of larger magnetic aggregates (with dominant magnetic dipolar interaction) such as the systems of superparamagnetic clusters described in Part I of this work [Kle01, Ulr03, Che04].

As a tendency, these systems often exhibit some of the characteristics of spin glasses (or glassy systems in general) like a kind of freezing transition and aging, yet the more sophisticated non-equilibrium effects like (strong) rejuvenation and memory are usually less pronounced or unobservable. One argument, detailed in [Jön05] for the case of a frozen ferrofluid, is that the latter effects may involve fairly large time scales as compared to the time scale \( \tau_0 \) of the underlying dynamics. Thus, phenomena observable for fast–flipping atomic moments (see above) may fall out of experimentally accessible times for much slower larger particles.

Some of the above phenomenology is also found in other glassy systems, most prominent are structural glasses [Bar03b]. These are different in that the disorder is self–induced dynamically, i.e. the material could principally form a crystal, whereas disorder and frustration in spin glasses are irrevocable (considering only spin degrees of freedom as such).

Indeed, possibly the earliest example reported of a kind of rejuvenation effect has been the Kovacs effect [Kov63, Ang00]: Upon rapid cooling to a temperature \( T_1 \) below the glass temperature \( T_g \), the specific volume \( v \) of a polymer glass decreases slowly towards a limiting value \( v_\infty(T_1) \), which is a decreasing function of temperature. When the sample is heated to \( T_2 > T_1 \) just in the instant when the specific volume passes the value \( v \approx v_\infty(T_2) \) it does not simply stay there, but rises somewhat before decaying again to \( v_\infty(T_2) \) on a curve similar to the one obtained by direct cooling to \( T_2 \). Note that, contrary to the terminology used here, the Kovacs effect in the glass literature is often referred to as a memory effect (memory to the state above \( T_g \), so to speak).

In recent years, there has been growing interest in aging, rejuvenation and memory effects in such diverse systems as simple liquids [Leh97], polymer glasses [Bel00], soft materials (like pastes [Clo00], colloidal suspensions [Via02], and foams [Höh99]), granular matter [Jöss00] as well as magnetic flux lines in dirty type–II superconductors [Ros89, Pap01]; see [Ber03]
for a recent overview.

While, certainly, not all of these systems share the same mechanisms, their variety stresses the general importance of this type of non-equilibrium phenomena and further motivates theoretical efforts, whereby, hopefully, progress of understanding in one class of system might bring forth new ideas and insight also for others.

3.2 Theoretical approaches

3.2.1 A standard model

In order to clarify the essential mechanisms underlying generic complex phenomena, it is good practice in theoretical statistical physics to attempt to capture these in as simple (and, hopefully, treatable) as possible models. In this vein, Edwards and Anderson – in a paper [Edw75] which marks the onset of spin glass theory – proposed to consider (classical) spins \( S_i \) on a regular lattice with Hamiltonian

\[
\mathcal{H} = -\frac{1}{2} \sum_{i,j} J_{ij} S_i S_j ,
\]

(3.6)

where the couplings \( J_{ij} \) are taken to be independent random variables with a distribution which merely depends on the separation \( r_{ij} \) of spins \( S_{i,j} \).

Specifically, in this work we refer to the Edwards–Anderson (EA) model as Ising spins \( S_i = \pm 1 \) on a (hyper-)cubic lattice with Hamiltonian

\[
\mathcal{H} = -\frac{1}{2} \sum_{\langle ij \rangle} J_{ij} S_i S_j - H \sum_i S_i ,
\]

(3.7)

where \( H \) is an eventual external magnetic field applied in the spins’ ‘+’-direction.\(^3\) The bonds \( J_{ij} (\equiv J_{ji}) \) are restricted to nearest-neighbor pairs of spins on the lattice and drawn randomly from a Gaussian distribution

\[
P(J_{ij}) = \frac{1}{\sqrt{2\pi J^2}} \exp \left[ -\frac{J_{ij}^2}{2J^2} \right]
\]

(3.8)

with zero mean, i.e. no predominance of ferro- or antiferromagnetic couplings, and standard deviation \( J \) setting the energy scale.

Another popular choice, especially in computer simulations (cf. below), is a bimodal distribution \( P(J_{ij}) = 1/2 (\delta(J_{ij} - J) + \delta(J_{ij} + J)) \) giving the so-called \( \pm J \)-model [Tou77]. As in fig. 3.4, it is often taken to illustrate frustration and ambiguity of the system’s ground state. The essential role of frustration is illustrated by a ‘deformed’ variant, the so-called Mattis model [Mat76]: With bonds chosen specifically correlated in order to avoid frustration, \( J_{ij} = J \xi_i \xi_j \), where \( \xi_i = \pm 1 \) denotes a random variable assigned to site \( i \), a simple renaming of spins \( \tilde{S}_i = S_i \xi_i \) maps this model to an ordinary ferromagnetic Ising model, revealing it as a mere ‘disguised ferromagnet’.

\(^3\)Note that in Part II of this work, contrary to Part I, the magnetic field is taken in units of an energy \( \mu_0 g \mu_B H \), where \( g \mu_B \) is the magnetic moment associated with a spin \( S_i \).
3.2 Theoretical approaches

Figure 3.4: Schematically: Frustration in a $\pm J$ spin glass model. In the plaquette on the left, the arrangement of ferro- and antiferromagnetic bonds (straight and jagged lines, respectively) allows for a spin configuration to satisfy all bonds (as would its inverse). In the frustrated plaquette on the right, at least one bond has to be broken.

In any case, once drawn, the bonds are assumed constant for that particular system, which corresponds to so-called frozen-in or quenched disorder (as opposed to mutable annealed disorder). As usual for systems with random disorder, since one is typically interested in generic self-averaging quantities, i.e. quantities with vanishing sample-to-sample variance in the thermodynamic limit, the theoretical treatment may conveniently include an average over the disorder. At this point, let us note that in principle, one could also just rely on the self-averaging, however the thermodynamic limit may be harder to perform for quantities still depending on the disorder. In case a quantity of interest is not self-averaging, one will aim to study its distribution, cf. the next subsection. Within our approach, we won’t come across such subtleties [Fis91, Nis01].

Edwards and Anderson also proposed a dynamically defined order parameter

$$q_{EA} = \lim_{t \to \infty} \lim_{N \to \infty} \langle S_i(t + \tau) S_i(\tau) \rangle,$$

where the bar denotes the discussed disorder average. Corresponding to the long-time limit of the spin autocorrelation (3.3), this order parameter reflects the freezing of spins below for $T < T_c$ and for this purpose substitutes the vanishing magnetization (3.2).

Note the crucial order of limits in (3.9): The thermodynamic limit $N \to \infty$ has to be taken first, since, in a finite system, any correlations eventually vanish. Moreover, as (3.9) is meant in thermal equilibrium, starting from a non-equilibrium situation actually requires another limit $t' \to \infty$ before that of $t$, cf. sec. 5.1.

Today, there is some consensus that the EA model (3.7) above its lower critical dimension, i.e. for $d \geq 3$, in principle bears all characteristic features of spin glasses (see e.g. [Jim05]). An obvious exception are particular effects in the dynamics of freely rotatable Heisenberg spins $S_i$, which are studied in a separate area of research [Kaw01, Ber04].

Despite its conceptual simplicity, the EA model largely resists straightforward analytical or numerical treatment, cf. below. Difficulties of the latter originate from the already mentioned large relevant time (and length) scales. While simulations may typically cover times $t/\tau_0 \in [1, 10^6]$ (with $\tau_0$ e.g. a Monte Carlo step), $t/\tau_0 \in [10^{12}, 10^{18}]$ for experiments on atomic spin glasses [Jön05]; at the same time, finite size effects are unusually strong and, so much the worse, not well understood.

Indeed, already the search for the non-obvious ground state of the EA or $\pm J$-model (in
3 Aging, rejuvenation and memory effects in systems far from equilibrium

d = 3) is an NP-hard combinatorial optimization problem, which, put simply, implies a computational effort growing exponentially with system size [Har02]. In fact, it has become a common test case for heuristic optimization algorithms, some of which, e.g. simulated annealing, are in turn inspired by physical mechanisms [Har02, Har04].

The complications mentioned underline the interest in treatable modifications of the basic EA model as well as different meta-models. Such approaches for spin glasses and glassy systems in general fall into two classes [Ber03], i.e. mean field–type theories and spatial or domain growth pictures, which are outlined in the remainder of this chapter.

3.2.2 Mean field–type theories

(Multi-)trap models

In so-called trap models [Bou92], the evolution of a glassy system is viewed as movement of a point particle in the high-dimensional configuration space. Thereby, the reduction of a many–body to a one-particle problem qualifies this as a mean field–type approach.

Usually, one assumes an effective thermally activated diffusive motion through a rugged random energy landscape. Then, on basis of a generic broad distribution of energies E such as \( \rho(E) \sim \exp(-E/T_g) \) for \( E \leq 0 \), which may be motivated e.g. by extreme value statistics, one readily obtains a heavy-tailed distribution of trapping times

\[
\rho(\tau) \sim \tau^{-(1+T/T_g)}
\]

(3.10)

with diverging mean for \( T < T_g \). For the aging behavior, this yields a \( t/t' \)-scaling, i.e. (3.5) with \( \mu = 1 \). Moreover, subaging (\( \mu < 1 \)) and more sophisticated scaling properties may be obtained depending on the choice of jump rates for the particle [Rin00].

Rejuvenation and memory effects as in spin glasses are found in multi–trap models with hierarchical energy landscapes [Bou95, Sas00]: Upon lowering the temperature, new (deeper) levels of the hierarchy are explored (rejuvenation), while upon reheating, the system still finds itself in the higher level ‘valley’ explored before.

A problem of these approaches is the substantiation and interpretation of assumptions and results in phase space in terms of less abstract conceptions in real space.

Infinite–range models

Mean field approximations (in a stricter sense), known from elementary statistical mechanics as a first step of understanding of collective phenomena, typically become exact in the limit of infinite–range interactions, hence the interest in models including the latter. For structural glasses, a prominent example is the p–spin spherical model [Cug93].

For spin glasses, the Sherrington–Kirkpatrick (SK) model [She75] is most important. Basically, it is the EA model (3.7), however with Gaussian distributed random bonds (3.8) between all spins irrespective of their distance, whereby the standard deviation of the Gaussian is rescaled, \( J \rightarrow J/\sqrt{N} \), in order to keep thermodynamic quantities such as the energy extensive. A similar model due to Viana and Bray [Via85], where each spin interacts
with a finite number of randomly chosen other spins, implies finite connectivity, but also
lacks any lattice topology; it has not been solved exactly though.

In the SK model, as often in the statistical mechanics of disordered systems, the perfor-

mance of the disorder average (cf. above) of the free energy $F = -T \log(Z)$ is facilitated by
the ‘replica trick’,

$$\log(Z) = \lim_{n \to 0} \frac{Z^n - 1}{n},$$

since averaging a power $Z^n$ of the partition function $Z$ (for integer $n$, which is then continued
analytically) is easier mathematically than averaging its logarithm [Dot01]. However, a
naive procedure leads to the same unphysical instable results (e.g. negative entropy) as
does an elementary mean field ansatz. Note that a refined approach (including an Onsager
reaction field term) by Thouless, Anderson, and Palmer (TAP) gives a better hint to the
behavior to be expected [Fis91, Nis01].

The correct analytical solution of the SK model due to Parisi (see e.g. [Méz87]) involves
so-called replica symmetry breaking, where, in simple words, replica of the system (cor-
responding to factors of $Z$ in $Z^n$) have to be allowed different behavior. The main outcome
is a complicated energy landscape with a distinct hierarchy of states, which may be best il-
\n\llustrated in comparison to a conventional ferromagnet (e.g. the ferromagnetic Ising model).
In the latter, ergodicity is broken below $T_c$ in that there is an infinite energy barrier (in
the thermodynamic limit) separating the regions of phase space with positive and negative
magnetization $M$.

In the SK model, the splitting up repeats continually at any temperature below $T_c$ yielding
an infinity of states, each in its own separate rugged ‘valley’ of the energy landscape, which
will be broken up further by barriers growing infinite upon lowering of the temperature.
Thus, the states characterized by their spin configuration may be thought of as organized
in a tree-like hierarchical structure. Indeed, with a metric given by the overlap

$$q_{\alpha\beta} = \frac{1}{2} \sum_i \langle S_i \rangle_{\alpha} \langle S_i \rangle_{\beta},$$

where \{\langle S_i \rangle_{\alpha}\} is the thermal equilibrium spin configuration of a state $\alpha$, the states form
elements of an ultrametric space.$^4$ An often cited consequence is a continuous distribution
$P(q)$ of possible overlaps among replica of the system.

Traditionally, studies building on the infinite-range SK model constitute the mainstream
of theoretical spin glass research, which is also reflected in available textbooks on the sub-
ject [Fis91, Dot94, Nis01]. Interestingly, the corresponding novel, highly elaborate analytical
theory has found interdisciplinary applications in the field of neural networks and informa-
tion processing (as a starting point, see [Méz87] or the aforementioned books).

As for the non-equilibrium dynamics in the spin glass phase, which is our main concern,
the aforementioned structure of the solution translates into ‘dynamic ultrametricity’, which
includes the presence of a continuous hierarchy of diverging time scales and leads to aging as

\footnote{Mathematically, this is defined as a space where $d(\alpha, \beta) \leq \max \{d(\alpha, \gamma), d(\beta, \gamma)\}$ holds for the distance
d(\ldots) of arbitrary elements $\alpha, \beta, \gamma$ in addition to the usual, less restrictive triangle inequality (without
the ‘max’). The resulting hierarchical structure may be imagined as described.}
well as, asymptotically, rejuvenation and memory effects [Cug99]. As a drawback (also noted in the preceding reference), the hierarchy of time scales, which is central to the approach, seems incompatible with the scaling behavior found in experiments, thus questioning the general validity of the scenario.

### 3.2.3 Spatial (domain growth) theories

In domain growth theories, as implied by their name, a glassy system’s aging dynamics is viewed in analogy — or better: generalization — of the domain coarsening known from conventional ordered systems, e.g. a pure ferromagnetic Ising model quenched below its critical temperature. Typically, the description is in terms of a growing characteristic coherence length or domain size \( L(t) \), whereby scaling arguments may be adopted from (real space) renormalization calculations yielding effective interactions on large length scales, see sec. 4.1.2.

Since our approach clearly fits into this category, many details will be given in the following chapters. Here we emphasize some basics of a corresponding model for spin glasses, whereby, in view of its later use, the next passage will be more detailed than the other parts of the present introductory chapter.

**Droplet model**

As established mainly by Fisher and Huse [Fis88], the so-called droplet model aims at a comprehensive description of spin glass behavior in terms of droplets, i.e. low lying thermal excitations. An assumption, which, though formally not indispensable, is of practical importance (to keep scaling arguments manageable), is that there are merely two equilibrium states at a given \( T \), i.e. a state \( \Gamma \) and its ‘inverse’ \( \bar{\Gamma} \) with all spins in the opposite direction. A droplet on length scale \( L \) then may be defined as the excitation (patch of e.g. \( \bar{\Gamma} \) in a \( \Gamma \) region) of lowest energy with linear extent between \( L \) and \( 2L \) including a given site. Trivially, the energy cost stems from the droplet’s boundary, since the interior is unchanged upon spin reversal.

The free energy necessary for its creation for large \( L \) scales as

\[
F(L) \sim T(T) \left( \frac{L}{L_0} \right)^\theta ,
\]

where \( L_0 \) is an elementary length (e.g. the lattice constant) and the stiffness modulus \( T(T) \sim J = \text{const.} \), not too close to \( T_c \), cf. below. From numerical studies (with [Pal00] as a recent example), \( \theta \simeq 0.21 \) in the 3D EA model, which is consistent with the argument \( 0 < \theta \leq (d - 1)/2 \). Put simply, \( d - 1 \) would correspond to a (flat) domain wall in a uniform ferromagnet, the factor 1/2 comes from the addition of random contributions (broken bonds) of both signs, and the ‘\( \leq \)’ comes from laying the droplet boundary preferably through weak or anyway frustrated bonds. The case \( \theta < 0 \) indeed occurs in dimensions below \( d = 3 \) and, making excitations at large enough \( L \) thermodynamically favorable, impedes long-range...
order.\(^5\)

A natural scaling ansatz for the distribution of droplet free energies in the system is

\[
\rho (F_L) dF_L = \tilde{\rho} \left( \frac{F_L}{J(L/L_0)^\theta} \right) \frac{dF_L}{J(L/L_0)^\theta},
\]

where, due to the disorder and frustration, the distribution will be broad and have weight down to zero energy, \(\tilde{\rho}(0) > 0\), implying at all \(T\) a population of active droplets which determine (quasi-)equilibrium behavior of the spin glass.

Due to the possible energy minimization just depicted and in sharp contrast to excitations in uniform systems, a droplet will naturally be a very ‘ragged’ object. In fact, the number of sites at a its surface can be argued to scale as \((L/L_0)^{d_k}\) with a (fractal) exponent \(d - 1 < d_s < d\). According to a recent numerical study [Pal99], \(d_s \simeq 2.68\) in the 3D EA model.

The droplet model assumes thermally activated dynamics with energy barriers scaling with droplet size \(L\) as

\[
B(L) \sim \Psi(T) \left( \frac{L}{L_0} \right)^\psi,
\]

where \(\psi \geq \theta\) is another exponent and the modulus \(\Psi(T)\) behaves similarly as \(\Upsilon(T)\).\(^6\) Aging then proceeds by equilibration of droplets on a length scale \(L(t)\) increasing with relaxation time \(t\),

\[
t = \tau_0 \exp \left[ \frac{J(L/L_0)^\psi}{T} \right] \quad \text{or} \quad L(t) = L_0 \left[ \frac{T}{J} \log \left( \frac{t}{\tau_0} \right) \right]^{1/\psi}.
\]

As mentioned in sec. 3.1.2, (strong) rejuvenation effects additionally demand a mechanism making aging (thermalization) at one temperature incompatible to that at another. Within the droplet model, this follows from a somewhat subtle, though revealing argument [Bra87, Fis88].

According to (3.14), two spins at a distance \(L\) typically experience an effective coupling of order \(J(L) \sim F(L) \sim (L/L_0)^\theta\), since the reversal of one of them implies the creation of a corresponding droplet or, in other words, domain wall surrounding it. In fact, this free energy, \(F(L) = U - TS(T)\), is a delicate balance between an inner energy contribution \(U\) due to the breaking or satisfaction of bonds at the droplet’s surface and the change of entropy \(S(T)\) of the \(\sim (L/L_0)^{d_k}\) spins affected by just that. Their individual contributions being effectively random in sign, \(S(T)\) grows as \(\sim (L/L_0)^{d_s/2}\) with \(L\).

For a given configuration of spins, a change of temperature \(\Delta T\) unhangs the above balance by an amount \(\Delta\{TS(T)\} \sim (L/L_0)^{d_s/2} \Delta T\) to leading order in \(\Delta T\) (cf. [Asp02]). The decisive point is that, for \(\zeta \equiv d_s/2 - \theta > 0\), the perturbation induced by a temperature shift typically exceeds the original coupling \(J(L)\) beyond the so-called overlap length

\[
L^* = L_0 \left( \frac{J}{\Delta T} \right)^{1/\zeta}.
\]

\(^5\) For a recent comprehensive study of \(\theta\) in various dimensions \(d\), see [Boc05c].

\(^6\) In detail, both vanish near \(T_c\) as \(\sim (1 - T/T_c)^{\nu_0}\) with \(\nu\), as usual, denoting the critical exponent of the correlation length.
Thus, the equilibrium spin configuration at some temperature $T$ may be expected to be destabilized beyond a length scale of order $L'(\Delta T)$ due to a temperature shift to $T + \Delta T$.

In other words, equilibrium spin configurations at two different temperatures, which are similar on small length scales $L \ll L'$, are completely decorrelated at large length scales $L \gtrsim L'$. Depending on the difference of temperatures $\Delta T$ and the length scales explored during the simulation or experiment, one thus expects either simple accumulative aging, rejuvenation, or, in general, a crossover from the first to the second.

These considerations will be made more explicit within our approach in sec. 5.2. The ultimate aftereffects of temperature changes $\Delta T$, how small whatsoever, beyond a length $L'$ have been termed temperature chaos in (vague) analogy to chaotic dynamical systems, where a small modification of initial conditions may lead to drastically different trajectories on large enough time scales. In this vein, the ‘chaos exponent’ $\zeta$ resembles a Lyapunov exponent.

Before returning to our general overview of theoretical approaches to spin glasses, let us add two remarks. First, a condition $\zeta = d_u / 2 - \theta \geq 0$ (with equality to zero as improbable extreme case) follows from the inequalities for $d_u$ and $\theta$ argued for above, whereas in a uniform ferromagnet, our common example of the opposite, $\zeta = d - 1 - (d - 1) \equiv 0$. Second, ‘chaotic’ alteration of equilibrium states may also (theoretically even more simply) be induced by bond perturbation, i.e. a small random modification of the ensemble of couplings $\{J_{ij}\}$.

Finally, as for memory effects, an obvious possible mechanism within a domain growth scenario is that the domain structure obtained at some temperature remains on large length scales, which are not explored during a short (or low-temperature) transient phase at another temperature (see e.g. [Ber03]). Again, more on that will be said in the following chapters.

Similar to the other approaches, an open problem of the droplet model is the justification of its assumptions and the substantiation of its concepts, i.e. droplets, length scales etc., within microscopically more concrete models such as, prototypically, the EA model.

Alternative domain growth approach

There is another approach in real space [Ber02a], which is interesting theoretically because it demonstrates that disorder and frustration are not indispensable prerequisites for aging, rejuvenation and memory effects as discussed in sec. 3.1. In fact, in most general terms, the minimum requirement for the latter two is that a system’s actual state carries some ‘lasting’ $T$–specific structure. The failure of a uniform ferromagnet to do so has two (not strictly separated) aspects: First, as already mentioned, the evolution at all $T < T_c$ is towards similar states (of essentially uniform magnetization on a large scale). Second, away from $T_c$ the typical spatial extent $\xi_{eq}(T)$ of thermal equilibrium fluctuations about these states is usually much smaller than the growing coherence length, $\xi_{eq}(T) \ll L(t)$; as a consequence, the adaption of their spectrum after a temperature shift occurs practically instantaneously (as viewed in the present context).

Thus, for glassy behavior, an idea is to postulate the opposite $\xi_{eq}(T) \gg L(t)$ in usual
3.2 Theoretical approaches

situations [Ber02a]. An example is the XY model in $d = 2$, where spins on a two-dimensional lattice are free to rotate in the plane and are coupled via ferromagnetic nearest neighbor interactions. Below a Kosterlitz–Thouless transition, the system exhibits quasi-long-range order, in which correlations decay algebraically with $T$–dependent exponents, interpretable as a line of critical points in the phase diagram (see e.g. [Cha95]). Then, with (long-range) thermal fluctuations playing a role similar to droplets and $L(t)$ identified as dynamic correlation length, one may think of a scenario for aging, rejuvenation and memory effects qualitatively similar to that of the droplet model. A difference is that critical fluctuations are reshuffled on all length scales after a temperature shift $\Delta T$, so there is no overlap length.

As a drawback, the clear occurrence of the abovementioned effects in the XY model is connected with the additional assumption of a temperature dependent spin wave stiffness $\rho(T)$ to ensure separation of length scales. More generally, the requirement of continual criticality may be relieved somewhat for glassy systems; as an example, $L(t)$ might be effectively bounded above by a reasonably large, though finite $\xi_{eq}(T)$, beyond which excitations are thermally activated and thus require exponentially increasing times [Ber02a, Ber02b]. Thereby, however, disorder re-enters as important factor.

The approach then complies with an explanation for rejuvenation and memory effects which does not involve ‘chaotic’ incompatibilities of equilibrium states at different temperatures [Bou00]. Instead, the coherence length $L(t)$, whose slow growth is responsible for (isothermal) aging, is assumed to increase continuously also after a temperature shift $\Delta T$. The latter, however, lets fast thermal equilibrium fluctuations (on shorter length scales) suddenly fall out of equilibrium; and this reappearance of short-scale out-of-equilibrium structures is what is noticed as rejuvenation. Memory, on the other hand, is again associated with the large-scale domain structure.

Clearly, this is interesting in view of rejuvenation and memory effects in systems where there is no conceivable mechanism analogous to temperature chaos, e.g. some of the systems mentioned in sec. 3.1.3 or, as an exemplary theoretical model, a site-diluted ferromagnetic Ising model [Jim05]. The distinction between both domain growth scenarios might also serve as a starting point for the classification of various aging phenomena.

Comparison of approaches

The two classical families of spin glass theories, i.e. those based on the mean-field SK model and the droplet/scaling picture, respectively, differ quite radically in their assumptions and predictions for the spin glass phase. Distinctions include (i) the structure of the ground state (many states/continuous $P(q)$ vs. merely two – spin flipped – states); (ii) the behavior in a magnetic field (stability below the de Almeida–Thouless line in the phase diagram vs. dynamic instability at any $H > 0$, cf. sec. 6.1); (iii) the question of replica symmetry breaking; (iv) the nature of excitations (ultrametric hierarchy vs. fractal droplets); ... and many more properties derived from these.

While, in the past, there have been strong debates between advocates of both views as to which is the more appropriate description of real spin glasses, by now concensus seems to have grown that neither of these descriptions alone is fully satisfactory and more insight in
the different facets of the problem is desirable. Thereby, the fact that these and many other issues could not be ultimately settled even by the most recent, extensive numerical efforts sheds some light on the complexity of the problem. Examples of current simulations of the dynamics include the references further below; for studies of the ground state structure and nature of excitations, we refer to the overview in [Har02, Har04].

As should have become clear from all the preceding descriptions, to date, there is no unique solvable model for spin glasses capturing all possible features of these complex materials, and, at the same time, being unambiguously derivable from a microscopic standpoint. By contrast, the ‘true’ nature of spin glasses still remains a challenge to theoretical description. Thus, we consider inappropriate any kind of dogmatism, which all too easily may become a hindrance to scientific progress.

As for the domain growth theories, the relative role of (critical) fluctuations as compared to temperature chaos (if present) for rejuvenation and memory effects is a subject of great interest in current research [Ber02b, Yos02, Jön04, Jim05]. While fluctuations in a fixed energy landscape may account for weak rejuvenation phenomena such as in the Kovacs effect, they might not be sufficient for the sharp rejuvenation and memory effects observed in spin glasses. Temperature chaos, on the other hand, involves eventually very large length scales \( L(t) \gtrsim L^* \) [Asp02], which, besides being hard to reach in conventional simulations, may in some cases be unrealistic also in experiments.

This, once again, illustrates the interest and difficulties of studying the non-equilibrium dynamics of spin glasses on large length and time scales, which is the main motivation for the development of our approach in the following chapters. By the end of Part II of this work, we will come to its contribution for a refined comprehensive domain growth scenario for spin glasses. More generally, the underlying original ideas may be of value also for similar phenomena in other glassy systems as exposed in the present chapter.
4 Spin glass model on a hierarchical lattice

4.1 Model

4.1.1 Hierarchical lattice

In theoretical physics, models are occasionally studied in ‘unnatural’ topologies. A well-known example, besides the already mentioned infinite connectivity of mean field models, is the Bethe lattice (also known as Cayley tree) [Bet35] for the Ising model or related problems. As a main motivation, these topologies allow for a simplified, often exact theoretical treatment, which in many cases is analogous or identical to approximate theories for systems on ordinary Bravais lattices. Thus, one may not only hope for transferable results, but also get some insight into the nature and implications of the approximations.

In this work, the ‘standard’ EA spin glass model presented in the previous chapter is studied on a hierarchical lattice, whose structure is explained in the following. As established by Berker and Ostlund [Ber79] (and discussed systematically in [Kau81]), it belongs to a class of systems where certain approximate real-space renormalization group (RSRG) schemes become exact. In our case, this is the Migdal–Kadanoff (MK) approximation, which will be detailed in the next subsection.

Similar to a fractal, a hierarchical lattice may be constructed iteratively. In the example depicted in fig. 4.1, one starts with two sites and a bond connecting them. Then, in every step of the iteration, each bond is replaced by a set of new bonds and sites. In part (a) of the figure, which corresponds to the lattice as we will employ it, these are \( p' = 4 \) pairs of bonds, each embracing a new site.

We note that the construction may readily be generalized to other starting points, different replacements, some non-iterated bonds or even stochastic elements in the iteration. Some of the former possibilities have been applied in an early work [McK82] linking qualitatively spin glass behavior and chaotic RSRG trajectories, though in way different from more recent approaches. Here, we adhere to the scheme of fig. 4.1.

The assignment of a dimension \( d \) to such a hierarchical lattice is not completely obvious. If, as suggested by fig. 4.1(b), a bond is replaced by lines of \( b \) bonds in step \( k \) of the iteration, a natural interpretation is the multiplication of the system’s linear extent \( L \) by a factor \( b \), i.e. \( L = L_0 b^k \), where \( L_0 \) is the length unit (given by the length of a single bond). The total
number of bonds after step $k$ is $N_b = (p' b)^k$, and that of the sites

$$N = 2 + \sum_{j=1}^{k} (b - 1)p' (p' b)^{j-1} = 2 + (b - 1)p' \frac{1 - (p' b)^k}{1 - p' b}, \quad (4.1)$$

hence both scale as $(p' b)^k$ for large $k$. Matching this to the system’s ‘volume’ $L^d$, one obtains $d = \log(p') / \log(b) + 1$.

Since the MK approximation of the RSRG is best for the analogue of $b$ being as small as possible (cf. below), one usually chooses $b = 2$ also for the hierarchical lattices. Then, with the abbreviation $p = 2p'$ for the number of new bonds per replaced old bond, we have $d = \log(p) / \log(2)$ justifying our choice $p = 8$ ($p' = 4$) for a 3D spin glass model. For that purpose, we assign an Ising spin $S_i = \pm 1$ to each site and a random coupling drawn from a Gaussian distribution (3.8) to each bond in a hierarchical lattice of the desired size $L/L_0 = 2^k$.

Unlike sites on an ordinary cubic lattice, those on the hierarchical lattice are not all topologically equivalent, but there is a hierarchy of connectivities $z$, cf. fig. 4.1: The spins \{ $S^{(0)}$ \} of level 0, by which we mean those created in the last step of the lattice’s iterative construction, have merely two nearest neighbors; the \{ $S^{(1)}$ \} created in the second last step have $z = 2p' = 8$; etc. In general, $z = 2(p')^\alpha = 2 \times 4^\alpha$ on level $\alpha$ except for the two outermost spins which were there first, which naturally is a finite size effect. We will explore consequences of this hierarchy for the dynamics of our model in sec. 4.2.

A related particularity, which will play a role in what follows, is that the hierarchical topology restricts the possible structure of domain walls. As an example, let us consider laying a domain wall between the two outer spins (black dots) in the system on the right of fig. 4.1(a). One of the outer spins is of level $\alpha = 2$, the other of a higher level $\beta > \alpha$ provided the whole object of linear extent $L/L_0 = 2^\alpha = 4$, which we will later refer to as a ‘cluster’ of level $\alpha$ (see sec. 4.2.2), forms part of a larger hierarchical lattice.

In any case, while there is some freedom through which of the bonds to lay the (single) domain wall, it turns out that their number is always exactly $(p')^\alpha$ ($4^2 = 16$ in the example), i.e. half the connectivity of the lower level boundary spin. A simple way to see this is...
to start with the wall through the $(p')^a$ bonds reaching from one of the boundary spins into the cluster and then to observe effects of moving it. Accordingly, there are always 
\[ (p')^a = (L/L_0)^{d-1} \] sites at the surface of the domain wall, i.e. the corresponding ‘fractal’ exponent introduced in sec. 3.2.3 here has the trivial value $d_s = d - 1$.

Note that the argument is more involved for a domain wall between arbitrary two spins in the lattice. However, as we will see, in most approaches excitations are genuinely treated on the level of clusters. In fact, as shown in the next subsection, the topological restrictions allow for an exact renormalization scheme, in which a cluster may be replaced by single ‘effective’ bond.

### 4.1.2 Real space renormalization group

**Renormalization**

The concept of renormalization (see e.g. [McC04]), or more exactly perturbative renormalization, originally stems from quantum field theory, where divergences in perturbative calculations of physical processes may be resolved by adapting or ‘renormalizing’ the original ‘bare’ coupling constants of the theory. First rather considered a formal nuisance, this was later – in fact inspired by results from statistical physics – understood as a hint to the limits of applicability of known theories on very small length scales.

In statistical physics, one usually starts with what seems to be an appropriate description on a microscopic scale and is interested in the resulting large-scale behavior.\footnote{Actually, the fact that simple theoretical models eventually yield a macroscopic behavior similar to much more complicated real materials may already be viewed as an instance of universality, cf. the following text.} Renormalization, in this context, involves coarse graining, i.e. the thinning of degrees of freedom coupled to an increase in the considered length scale. In particular, in the renormalization group one tries to preserve the form of the system’s Hamiltonian during gradual coarse graining in order to study the ‘flow’ of coupling constants, which here actually means the ratio of these and temperature $T$.

In the classical approach, interest is in nontrivial fixed points of the corresponding map, i.e. those not at zero or infinity (associated with $T \to \infty$ and $T = 0$, respectively), which pertain to phase transitions. Thereby, the critical behavior and especially critical exponents are determined by properties of the map in the vicinity of the fixed points irrespective of fine details of the underlying model, which is an important result known as universality.

In fact, renormalization methods rest on profound mechanisms of mathematical physics, which is reflected by their application to a large variety of problems as well as their deep relation to scaling and asymptotic analysis [Che96, Bar03a]. Here, we will utilize the coarse graining aspect of renormalization in real space (as opposed to essentially field-theoretic methods for continuous systems operating in Fourier space).
Figure 4.2: (a) Elementary example of coarse graining by decimation: The coupling of the two outer spins $S_{1,2}$ via the middle spin $S_0$ is emulated by a direct effective coupling $J'$ between the former. (b) The bond-moving scheme of the MK approximation illustrated for a square lattice.

Migdal–Kadanoff scheme

Ideally, a coarse graining procedure should not alter thermodynamic properties, which means, in formal terms, it should leave invariant the partition function $Z$. For the elementary example of a chain of three Ising spins depicted in fig. 4.2(a),

$$Z = \sum_{S_0,1,2=\pm 1} \exp \left[ J_1 S_1 S_0 + J_2 S_2 S_0 \right],$$

where the ‘vee’ (or háček) accent stands for division by temperature, $\tilde{J} \equiv J/T$. By performing the sum over $S_0$ and recombining terms, (4.2) can be shown to be identical to

$$Z = f(J_{1,2}) \sum_{S_1,2=\pm 1} \exp \left[ J' S_1 S_2 \right]$$

with an ‘effective’ coupling

$$J' = \frac{1}{2} \log \left[ \frac{\cosh(J_1 + J_2)}{\cosh(J_1 - J_2)} \right] = \tanh^{-1} \left[ \tanh(J_1) \tanh(J_2) \right]$$

between the remaining spins $S_{1,2}$, and a prefactor $f(J_{1,2}) = 2 [\cosh(J_1 + J_2)(J_1 - J_2)]^{1/2}$, which will be less interesting for our purpose.\footnote{While our approach to spin glass dynamics will employ ‘effective’ couplings (see sec. 4.2), we will not actually calculate the partition function (or any derived thermodynamic quantities) as in a conventional (equilibrium) statistical mechanics treatment. Hence, we need not consider the prefactor $f(J_{1,2})$ in (4.3).}

This procedure is the central element of an exact RSRG scheme for 1D systems with nearest neighbor interactions: ‘Decimation’ of e.g. every second site yields a system with effective couplings $J'$ between nearest neighbors of the remaining spins; thus bonds change, but the Hamiltonian is form–invariant. Unfortunately, for hypercubic lattices in $d > 1$, a similar proceeding inevitably introduces some novel ‘diagonal’ (farther reaching) bonds. So it requires an approximation at some point to keep the transform manageable upon iteration.

An obvious possibility is to neglect some of the new types of couplings, see e.g. [Bin92, Kad00]. Proposed by Migdal and Kadanoff [Mig76], another option is to first create 1D
segments as in fig. 4.2(a) within original lattice. As illustrated in fig. 4.2(b) for a square lattice, this is done by moving bonds between some pairs of spins to neighboring pairs, which are then coupled by (the sum of) several bonds. Thereby, the movement of ‘unwanted’ bonds (as opposed to a simple neglect) may be argued for in a kind of variational calculation, i.e. the resulting approximate partition function is an upper bound to the exact one.

In $d$ dimensions, a new bond is given by

$$J'_{\ldots} = \tanh^{-1} \left[ \frac{\tanh \left( \sum_{k=1}^{d} J_{k} \right)}{\tanh \left( \sum_{k=d+1}^{2d} J_{k} \right)} \right], \quad (4.5)$$

where we abstain from elaborating on the indexing of the couplings (which doesn’t matter anyway for either uniform or a statistical treatment of uncorrelated random bonds). The bond-moving scheme may readily be generalized to create linear segments of length $b > 2$; however, involving more displacements, this rather aggravates the approximation. For systems with uniform bonds, by contrast, an analytical continuation to $b \to 0$ gives good results [Cre92].

The problem is much simpler on the hierarchical lattice of fig. 4.1, which consists of nested 1D segments from the outset, so that a suitable RSRG transform may be performed exactly. Indeed, in view of the lattice, the decimation of the spins with lowest connectivity $z = 2$ corresponds to going back one step in the its iterative construction. Like that, stepwise course graining proceeds from the right to the left in fig. 4.1(a), whereby in each step the old bonds are replaced by effective renormalized bonds between the remaining spins.

In detail, in the $n$-th step of the course graining ($n \geq 1$), i.e. when the spins $\{S^{(\alpha)}\}$ of level $\alpha = n - 1$ are those of lowest connectivity, a group of $p'$ of them located between two higher-level spins $S^{(\beta)}_{i,j}$ ($\beta > \alpha$) is replaced by an effective bond

$$J'^{(n)}_{ij} = \sum_{k=1}^{p'} \tanh^{-1} \left[ \frac{\tanh \left( J^{(n-1)}_{k,i} \right)}{\tanh \left( J^{(n-1)}_{k,j} \right)} \right], \quad (4.6)$$

where $J^{(n-1)}_{k,i}$ denotes the old bond between spin $S^{(\alpha)}_{k}$ and one of the remaining spins $S^{(\beta)}_{i,j}$. Note that, in this notation, the bonds ‘of level 0’ are the initial Gaussian distributed couplings, $J^{(0)}_{i,j} \equiv J_{i,j}$. Similar to (4.5), the expression (4.6) is an immediate generalization of the elementary case (4.4), since contributions to the effective bond from parallel paths simply add (and prefactors $f(\ldots)$ multiply).

Just as in the hypercubic lattice, a course graining step corresponds to a doubling of the considered length scale (for $b \equiv 2$), i.e. bonds of level $n$ are associated with a length $L_{n} / L_{0} = 2^{n}$.

Clearly, with summation and hyperbolic tangent operations interchanged in (4.5) as compared to (4.6), the MK renormalization in a hypercubic and hierarchical lattice are not algebraically identical (for $d > 1$). Still, in their overall predictions and especially statistically, i.e. in view of their action on a distribution of bonds, both are very similar, see e.g. [Dro00] for the case of the EA as well as a three-spin model in $d = 3, 4$. While important differences such as in the effects of an additional applied field (see sec. 6.1) should be born in mind,
models on the hierarchical lattice are commonly regarded as ‘exact realizations’ of the MK approximation in the sense of sec. 4.1.1. As far as the distinction is not made explicitly, we will refer to the hierarchical lattice and (4.6) as the MK approach in the following.

### 4.1.3 Previous applications to spin glasses

The MK approach has successfully been employed in the study of spin glass models for many years. As most attractive feature, it constitutes one of the rare instances where large length scales can be explored conveniently by means of the explicit, simple RSRG transform discussed above. In a number of aspects, predictions from the MK approach comply well with experimental findings and numerical results from models on ordinary lattices. In particular, it confirms behavior as devised in the droplet model. Methodologically, however, this should rather be considered an important consistency check than an independent argument, since, as mentioned in sec. 3.2.3, the droplet model is largely based on scaling assumptions motivated by RSRG results. In the following, we will briefly review some of the results of the MK approach in comparison to other methods, which, in our view, also recommend it as starting point for further studies.

As an early result, the MK renormalization was found to predict correctly (as is known by now) the lower critical dimension for the spin glass transition of the EA model [Sou77]. As shown there, this numerical outcome may be reproduced by an approximate analytical calculation: Assuming that the distribution of bonds stays Gaussian upon iteration of the renormalization transform and using the low-temperature limit of (4.4) (and its derivatives), $J' \simeq \text{sgn}(J_i J_j) \min \{|J_i|, |J_j|\}$ for $T \to 0$, one obtains for the standard deviation of bonds of level $n$

$$J^{(n)} = \left[ \left( \frac{J^{(n)}_{ij}}{J_{ij}} \right)^2 \right]^{1/2} \simeq \left[ \left( 1 - \frac{2}{\pi} \right) n' \right]^{n/2} \cdot J \simeq (0.363 \, n')^{n/2} \cdot J.$$  

(4.7)

This displays the vanishing of large scale couplings (corresponding to $n \gg 1$) in $d = 2$ ($n' = 2^{d-1} = 2$) and their eventual increase for low $T$ in $d = 3$ ($n' = 4$). Indeed, for the latter, one finds a critical temperature $T_c \simeq 0.896$ (value from [Asp02]).

Further, the MK renormalization gives the scaling behavior (3.13) and (3.14) of $F_L$, i.e. the free energy cost of excitations or, equivalently, the effective couplings on length scale $L$ [Asp02]. The corresponding ‘stiffness’ exponent is found to be $\theta \simeq -0.27$ for $d = 2$ [Bra84] and $\theta \simeq 0.26$ for $d = 3$ [Asp02], which is some percent below what predicts the simple approximation (4.7).

More refined analytical estimates of $\theta$ are discussed in [Bou03] in the context of the scaling of average ground state energies, $E(L) = e_0 L^d + e_1 L^{d_{\Theta_s}}$, with constants $e_{0,1}$ and $\Theta_s \equiv \theta$ for the hierarchical lattice. Thereby, the latter is also found to obey the generic requirement $\Theta_f \equiv d/2$ for finite dimensional spin glasses [Weh90] concerning the scaling of fluctuations due to different realizations of the disorder $[E^2(L) - E(L)^2] \sim L^{\Theta_s}$.

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\(^3\)As usual, the signum function is defined by $\text{sgn}(x) = -1$ for $x < 0$, $0$ for $x = 0$, and $1$ for $x > 0$. For the derivation of the expression for $J'$, cf. sec. 6.1.
The above values for $\theta$ compare reasonably well with results of numerical calculations independent of renormalization on hypercubic lattices, in which typically the energy is monitored while a domain wall is forced to pass through the system by e.g. changing boundary conditions from periodic to antiperiodic (in one direction). According to recent studies, $\theta \approx -0.29$ for $d = 2$ [Har03] and, as already mentioned, $\theta \approx 0.21$ for $d = 3$ [Pal00] (cf. sec. 3.2.3).

Concerning the scaling (3.15) of typical energy barriers with $L$, the relevant exponent has recently been proved to be $\psi = d - 1$ in any dimension on the hierarchical lattice [Dro04]. This value, which actually corresponds to a flat domain wall in a uniform system, is at the upper limit of the range $\theta \leq \psi \leq d - 1$ predicted by the droplet model. To date, there are no unequivocal results from models on conventional lattices or measurements on real spin glasses; however, at least some of them point to values quite close to $d - 1$, see the discussion in the preceding reference.

As a central feature, the MK approach exhibits temperature chaos [Ban87, Nif92] and indeed, due to the access to large length scales, has become a favorite object of study regarding this effect. With $d_\phi$ and $\theta$ from above, the chaos exponent becomes $\zeta = d_\phi/2 - \theta \approx 0.74$. The energy--vs.--entropy argument of sec. 3.2.3 may be reproduced and the overlap length $L^*(\Delta T)$ of (3.17) can be identified with a length $\sim L_0 \cdot 2^n$, where $n$ is the number of iterations of the MK renormalization at two different temperatures $T, T + \Delta T$, after which the respective effective bonds obtained from an initially identical set $\{J_{ij}\}$ become decorrelated [Ban87, Asp02]. Thus, the transform (4.6) acts as a chaotic map with temperature $T$ or, as already mentioned, a small random perturbation added to each bond as sensitive parameters.

The important aspect that $L^*(\Delta T)$ or, more precisely, $L^*(T, \Delta T)$ may become very large for small $\Delta T$, has been studied systematically in the MK approach and, as far as allowed by restrictions in system sizes, found similarly by transfer matrix calculations on a cubic lattice [Asp02]. For the latter, the chaos exponent may be estimated $\zeta \approx 1.15$.

In compliance with the explanation in sec. 3.2.3 (and the formula for $\zeta$), a recent investigation indicates that the chaos effect is weakened by energy ‘stiffness’ and amplified by entropic fluctuations in the system [Krz04]. This is revealed by a comparison of Ising spin glasses with $\pm J$, Gaussian, and a heavy-tailed distribution of bonds $P(J_{ij})$ as well as a spin glass model with continuous (planar) XY spins, which are listed here in ascending order of ‘chaos’ as observed by the MK renormalization and an approximate generalization thereof for XY spins, respectively. A shorter overlap length $L^*$ might eventually be a clue as to why rejuvenation effects are stronger in experiments on materials with continuous rather than Ising-like spins.

In some studies, the MK approach has been utilized to examine the overlap distribution $P(q)$ discussed in sec. 3.2.2, see [Moo08, Dro00] and references therein. Interestingly, results reminiscent of replica symmetry breaking and traces of an apparent deAlmeida–Thouless line in the presence of a magnetic field (cf. sec. 6.1) are found for small system sizes and close to $T_c$. So it is argued that critical fluctuations together with finite size effects may also account for indications of mean field–like behavior in some simulations on conventional lattices. However, we desist from delving further into this controversial issue.
4 Spin glass model on a hierarchical lattice

An enlarged set of reduction rules may be applied to a bond–diluted system on the hierarchical lattice [Boe03]. The bond–dilution generally enables the numerical treatment of systems of larger size or higher dimension, and the combination of data at various bond densities in a scaling ansatz may offer an accurate method to determine (eventually) universal properties such as characteristic exponents. For conventional lattices, the reduction scheme has to be supplemented e.g. by an optimization heuristic [Boe05a] or an exact graph–theoretical matching algorithm (for \( d = 2 \) [Boe05b], whereby still profit is taken from insight into the dilution method gained on the hierarchical lattice.

While all the mentioned applications of the MK scheme are concerned with ‘static’ quantities, which are extracted mainly from the bond distributions, in our approach we will attempt to utilize it for dynamical simulations. Since there is no simple mapping to the dynamics on hypercubic lattices, it is not clear a priori whether or in which way this may lead to any meaningful outcome. Yet, as we shall demonstrate in the following, the MK scheme may indeed serve as a basis of a viable and insightful approach to spin glass dynamics.

Parallel to our approach, a similar – and in a way complementary – scheme has been proposed by Sasaki and Martin [Sas03]. Since their approach to the dynamics relies on Monte Carlo simulations, we will discuss it together with other possible adaptations of this technique in sec. 6.2. There we will also comment on a previous study [Ric00], in which an attempt was made to explore the dynamics on the hierarchical lattice in the presence of a magnetic field by a straightforward application of Monte Carlo simulations.

4.2 Effective dynamics

4.2.1 Choice of a dynamics

The conception of dynamics generally associated with the EA model – or, in this respect, any ‘magnetic’ Ising model – consists of single spin flips with non–conserved order parameter (Glauber dynamics) and thermally activated rates of e.g. Metropolis type. So this is also what we have in mind as basic dynamical mechanism on the hierarchical lattice, whereby, as in many aspects, the peculiarities of the topology leave some room for additional options; as an example, one might argue for attempt frequencies, i.e. prefactors of the rates, depending to some degree on the level of the envisaged spin, cf. sec. 6.2.

As stated previously, our main interest is in the effective dynamics of the EA spin glass model on the hierarchical lattice on large time and length scales. An effect outshining any (reasonable) choice of dynamical details is the hierarchy of time scales in the system’s relaxation caused by its hierarchical construction.

For an explanation, let us consider a spin configuration out of equilibrium such as after an abrupt change of temperature \( T \). Then, the (re–)thermalization of the spins \( \{ S^{(0)} \} \) of level 0 with only two nearest neighbors merely requires single spin flips, i.e. elementary steps of the dynamics. For the thermalization to proceed onto a length scale twice as large, i.e. for a spin \( S^{(1)}_i \) and (e.g.) \( S^{(2)}_j \) to assume their preferred relative orientation, their state has to be ‘transmitted’ through a set of \( \ell \) spins of level 0. Very likely, \( S^{(1)}_i \) and \( S^{(2)}_j \) may be trapped in a metastable state due to the thermalization of those of their nearest neighbors which are
coupled more strongly to one of them than to the other.\textsuperscript{4} Thus, it is easy to imagine that the thermalization on increasing length scales, i.e. of spins of ever higher levels $n$, becomes increasingly complicated, or, as measured by the number of single spin flips required in (suitable) sequence, increasingly slow. Also, the discussion applies similarly to the creation or annihilation of thermal equilibrium fluctuations on different length scales.

Certainly, this is a general mechanism which, however, is particularly tangible (and pronounced) on the hierarchical lattice because the latter allows for a mapping of abstract length scales to specific classes (i.e. levels) of spins. It underlies the assumed scaling (3.15) of typical energy barriers with length scale $L$, $B(L) \sim J(L/L_0)^\psi$. Thereby, as noted in sec. 4.1.1, $\psi = d - 1$ has lately been proofed exactly for the hierarchical lattice by a more systematic and quantitative version of the preceding considerations, in which thermalization of lower level spins was assumed [Dro04].

Hence one may write for the typical relaxation time $t_n$ at level $n$

$$t_n = \tau_0 \exp \left[ J(L_n/L_0)^\psi T \right] = \tau_0 \exp \left[ \frac{J}{T} \cdot 2^{n(d-1)} \right], \tag{4.8}$$

which up to the specification of levels $n$ and length scales $L_n/L_0 = 2^n$ is identical to (3.16) postulated in the droplet model; again, $\tau_0$ is a microscopic time unit. In order to obtain a plain, concise mechanism for our effective dynamics on very large time and length scales, we will confine to a time resolution corresponding to these ‘epochs’ $t_n$.

Thereby, reliance on the specific form of (3.16) or (4.8) may be avoided by studying the essential characteristics of spin glass behavior as a function of the growing thermalization length $L_n = L(t_n)$, of which it suffices to bear in mind that it grows (doubles) on some logarithmic or even coarser time scale. On this abstract level, results from the (obviously) qualitative spin glass model on the hierarchical lattice may be compared to experimental findings by inserting the appropriate growth law of $L(t,T)$ as observed in that specific system, cf. sec. 5.2.2.

As main approximation, which may be justified by the large separation of time scales (4.8) together with the confinement in time resolution, in the $n$-th epoch we categorically consider spins of levels $n + 1$ and higher \{$S^{(n+1,n+2,...)}\} to be frozen in, while the spins \{$S^{(n-1,...,0)}\} fluctuate with strongly decreasing relaxation times $t_n \gg t_{n-1} \gg \cdots \gg t_0$.

To obtain a representative spin configuration of the $n$-th epoch, one may conveniently employ the exact MK renormalization as illustrated in fig. 4.3: First, the slowest, yet mobile spins $S_i^{(n)}$ are thermalized, i.e. aligned with Boltzmann weights $\propto \exp[-S_i^{(n)} h_i^{(n)}/T]$ in their effective (time-averaged) local fields

$$h_i^{(n)} = J_i^{(n)} S_i^{(n+1)} + J_i^{(n)} S_k^{(n+2,n+3,...)}, \tag{4.9}$$

Here $J_i^{(n)}$ are the effective couplings (of level $n$) given by the RSRG transformation (4.6), which take into account the thermalization of the faster spins at lower levels $n' < n$ lying

\textsuperscript{4}This effect is genuine: If $S_i^{(1,2)}$ flip first, they won’t ‘know’ about their relative orientation. If, by contrast, any of the interjacent $S_k^{(0)}$, $k = 1, \ldots, n'$, flips first, it will likely strengthen either $S_i^{(1)}$ or $S_j^{(2)}$ (depending on which of them the coupling is tighter) in their current orientation.
4 Spin glass model on a hierarchical lattice

Figure 4.3: Sketch of the hierarchical lattice, drawn here for $d = 2$ ($p' = 2$) for simplicity. As explained before, repetitive application of the MK renormalization yields a series of increasingly coarse grained ‘copies’ of the original system (on the left). As illustrated on the right, in the effective dynamics, spins up to level $n$ are considered mobile in the $n$-th epoch. To get a representative spin configuration of the corresponding time scale $t_n$, spins of levels $n$ down to 0 are thermalized in sequence, each in that ‘copy’ of the system where it has attained minimal connectivity $z = 2$ (and would be decimated in the next renormalization step); cf. text.

originally (before coarse graining) between $S_i^{(n)}$ and $S_i^{(n+1,n+2,...)}$. In the second step, the spins $S_i^{(n-1)}$ are thermalized in their effective local fields $h_i^{(n-1)}$, which depend on the spins $S_i^{(n)}$ updated in the first step (and the effective couplings $\{J^{(n-1)}\}$). By repeating this procedure, which corresponds to going from the right to the left in the series of systems depicted in fig. 4.3, the spins $\{S^{(n)}\}, \{S^{(n-1)}\}, \ldots, \{S^{(0)}\}$ are updated one after the other in the $n$-th epoch.

We note that, insofar as one does not aim at a resolution of the dynamics within an epoch $t_n$, the described ‘downward’ update of spin levels may be understood more simply as a convenient means to obtain a configuration of the mobile spins $\{S^{(n,n-1,...,0)}\}$ with correct thermodynamical weight under the boundary conditions given by the still frozen spins $\{S^{(n+1,n+2,...)}\}$.

In the way described, our scheme, which we will refer to as the effective dynamics in the following, efficiently yields a representative spin configuration of successive epochs $t_n$, by which the system’s evolution may be monitored on large time and length scales out of reach of conventional Monte Carlo simulations. Furthermore, its clear mechanism is amenable to (approximate) analytical approaches combined with scaling arguments, see below. We shall demonstrate in the following chapter that all of this constitutes an interesting dynamical spin glass model with a rich and realistic behavior.

As suggested above, our system together with the effective dynamics may also be considered as a standalone model independent of conventional dynamical schemes. Clearly, though, the objective will usually be an interpretation in as close as possible analogy to the microscopically more realistic (concrete) EA model. An obvious shortcoming of the effective dynamics is the drastically limited time resolution, which impedes the direct observation of
4.2 Effective dynamics

quantities on fine time scales, e.g. frequency dependent susceptibilities. In chap. 6, possible extensions and alternative dynamical schemes for the EA model on the hierarchical lattice will be discussed.

In the remainder of the present chapter, we will further illustrate the effective dynamics and its possible analysis with the spin autocorrelation function, which subsequently will be the main object of study within our approach.

4.2.2 Impact on the spin autocorrelation function

Spin autocorrelation

As noted in sec. 3.1.2, a preferred observable in theoretical treatments of spin glasses is the spin autocorrelation function $C(t, t')$ with basic definition (3.3), since this gauge–invariant (i.e. invariant under reversal of all spins) two–time quantity is a quite direct measure of the dynamics of individual spins and does not require the inclusion of an external magnetic field. The last point is particularly relevant on the hierarchical lattice where, as mentioned earlier and to be discussed thoroughly in sec. 6.1, an external field acts differently than on conventional lattices.

In our approach, following the standard protocol, we will generically consider a quench from $T = \infty$, i.e. a random spin configuration, to a temperature $T < T_c$ at time zero and subsequently apply the effective dynamics to get representative spin configurations of successive epochs $t_n$. In this way, we may study the evolution of the spin autocorrelation across these growing time scales.

Specifically, we may first apply the $n$–th step of the effective dynamics to obtain a reference state corresponding to the system after a waiting time $t' = t_n$. Then, the effective dynamics is restarted (i.e. beginning with $\{S^{(0)}\}$, ascending levels of spins are treated as mobile) to yield configurations at times $t_n + t_0, t_n + t_1, \ldots$ as they develop from the reference state.\footnote{In other words, the end $t = t_n$ of the waiting time, \textit{not} the instant $t = 0$ of the quench, is taken as ‘time zero’ for subsequent epochs of the effective dynamics.} On this basis, we obtain the spin autocorrelation function

$$C(t_m, t_n) = \sum_\alpha \sum_{k_{\alpha}} \langle S^{(\alpha)}_{k_{\alpha}}(t_n + t_m) S^{(\alpha)}_{k_{\alpha}}(t_n) \rangle,$$  \hspace{1cm} (4.10)

where $\langle \ldots \rangle$ denotes a thermal average and an average over the random initial spin orientations for a fixed realization of the disorder, while the bar denotes the disorder average over the random bonds. Moreover, in this adaption of (3.3) for the hierarchical lattice, the inner sum is over the $N_{\alpha}$ spins $\{S^{(\alpha)}\}$ on level $\alpha$, and the outer sum is over all levels.

The quantities $w_{\alpha} > 0$, $\sum_\alpha w_\alpha N_\alpha = 1$, are weighting factors, which allow one to take into account that spins at different levels are not (topologically) equivalent. So, their appearance issues from the profound question of how exactly to interpret the hierarchical topology in terms of a conventional lattice – as far as this is possible anyway, cf. the discussion below. For the spin autocorrelation (4.10), there seem to be two natural choices: \textit{(i)} per site weighting $w_\alpha = \text{const.} (= 1/N)$, where every spin counts the same; vs. \textit{(ii)} per bond
weighting \( w_\alpha \propto 4^\alpha \), where a spin counts according to its connectivity \( z \), cf. sec. 4.1.1. In any case, we find that both possibilities yield analogous results and thus, primarily as a matter of convenience, mainly use per bond weighting (ii), since it turns out to let \( C(t_m, t_n) \) display more directly its eventual power law decay, see the next chapter.

Note that, after averaging over the disorder, initial conditions, and realizations of the effective dynamics, the contribution to \( C(t_m, t_n) \) of every spin of a certain level \( \alpha \) is the same, so that the inner summation in (4.10) may be replaced by a factor \( N_\alpha \). The weighting factor for level \( \alpha \) (as a whole) obeys \( w_\alpha N_\alpha \propto \gamma^{-\alpha} \) for per site weighting (i) and \( \propto 2^{-\alpha} \) for per bond weighting (ii), because (in general dimension \( d = \log(p)/\log(2); p = 2 p' \)) \( N_\alpha \propto \gamma^{-\alpha} \) and \( \gamma \propto (p')^\alpha \).

### Cluster analysis

The time evolution of the system by means of the effective dynamics is best analyzed in terms of clusters, which are realizations of domains or droplets postulated in the scaling arguments of sec. 3.2.3. As mentioned earlier, a cluster is defined for each effective coupling \( J_{ij}^{(\alpha)} \) and consists of all faster spins at levels below \( \alpha \), which are traced out in the RSRG to give \( J_{ij}^{(\alpha)} \). An example for a cluster (of level 2) is the region marked gray in the system on the left of fig. 4.3. Each cluster has two boundary spins which are the spins \( S_i^{(\alpha)}, S_j^{(\beta)} \) connected by \( J_{ij}^{(\alpha)} \) after renormalization. As indicated, one is a spin \( S_i^{(\alpha)} \) at level \( \alpha \), which we call the ‘master spin’ of the cluster, while the other is a still slower spin \( S_j^{(\beta)} \) of level \( \beta > \alpha \).

An important remark is that \( 2 J_{ij}^{(\alpha)} \) is the difference of the cluster’s free energies of the cases that the boundary spins are parallel and anti-parallel. If the master spin flips but the slower boundary spin keeps unchanged, the spins in the interior of the cluster are exposed to a ‘twisted–boundary condition’, which triggers flips of an \( O(1) \) fraction of these spins. It is by this mechanism that fluctuations occurring at high levels propagate down and erase correlations of the low-level spins.

In fact, these de–correlations caused by twisted boundaries can be quantified in a precise manner. Let us consider a certain spin \( S^{(\alpha)} \) at level \( \alpha \). Since the clusters are hierarchically nested, this spin \( S^{(\alpha)} \) is part of unique clusters with master spins \( S^{(\gamma)} \), \( \gamma > \alpha \). By using symmetry considerations we can write for the correlator appearing in (4.10)

\[
\langle S^{(\alpha)}(t_n + t_m) S^{(\alpha)}(t_n) \rangle = \frac{1}{1 - 2 \alpha(t_m, t_n)} \prod_{\gamma = \alpha + 1}^{m} \frac{1 - r_\gamma(t_m, t_n)}{1}, \tag{4.11}
\]

where \( r_\gamma(t_m, t_n) \) is the probability that the spin \( S^{(\gamma)} \) flips between \( t_n \) and \( t_n + t_m \) under the condition that the master spins \( S^{(\gamma')} \), \( \gamma < \gamma' \leq m \), remained unchanged (for a given realization of the disorder). The product over \( \gamma \) in (4.11) is the consequence of the twisted

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6Theoretically, one could think of very specific dynamical schemes where even the form of (4.10), i.e. the averaging inside the summation over levels and spins, would be inept. However, for the usual cases considered here and, especially, the effective dynamics, this clearly is not an issue.
boundary effect: None of the boundary spins of the ‘super’−clusters containing the spin $S^{(\alpha)}$ is allowed to flip between $t_n$ and $t_n + t_m$, if $S^{(\alpha)}(t_n + t_m)$ should give (on average) a non−zero overlap with $S^{(\alpha)}(t_n)$; see app. A.2 for an explicit proof of this rule.

In situations where typical flip probabilities are small, $r_\gamma \ll 1$, (4.11) may be linearized to yield

$$\langle S^{(\alpha)}(t_n + t_m) S^{(\alpha)}(t_n) \rangle \simeq 1 - 2 \bar{r}_\alpha(t_m, t_n) - \sum_{\gamma = \alpha + 1}^m \bar{r}_\gamma(t_m, t_n).$$

(4.12)

In the following, this expression will prove useful since, together with scaling assumptions for the flip rates $\bar{r}_\gamma(t_m, t_n)$, it allows for an (approximate) analytical description of the decay of the spin autocorrelation.

Let us close this preparatory discussion by pointing out a particular implication of the linearization (4.12): Referring to a specific spin $S^{(\alpha)}$ and its master spins (in a given realization of the random bonds), the rates $r_{\alpha, \gamma}$ in (4.11) in principle are subtly correlated. Clearly, this is because the renormalized couplings at higher levels are calculated from those (of the same cluster) at lower levels and because some spins $S^{(\alpha)}$ share common higher−level neighbors $S^{(\gamma)}$, $\gamma > \alpha$. Although these correlations might be estimated weak in many cases, the use of (4.12) allows to neglect them altogether.
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5.1 Aging

In the present chapter, we will exhibit and discuss core findings of our RSRG approach to spin glass dynamics. We start with the first of the elemental non-equilibrium effects introduced sec. 3.1.2, i.e. isothermal aging. For its study, we proceed as explained in sec. 4.2: We take our system on the hierarchical lattice with a random spin configuration mimicking an ideal quench from $T = \infty$ to a temperature $T < T_c$, let it then evolve up to the $n$-th epoch of the effective dynamics to simulate a waiting time $t_n$, and begin monitoring the spin autocorrelation function $C(t_m, t_n)$ of (4.10) on successive epochs $t_m$.

As motivated in sec. 4.2, results will generically be presented in terms of the growing thermalization length $L(t_m)$, cf. (4.8). The minimum simulated system size is $2^{15}L_0$, i.e. 15 steps of the lattice’s iterative construction. For further details of the simulations, we refer to app. A.1. As far as not indicated otherwise, the data below are for per bond weighting.

Fig. (5.1) shows the typical behavior of $C(t_m, t_n)$ as a function of $L(t_m)$ at two exemplary temperatures after various waiting times $t_n$ (corresponding to pre-thermalized length scales $L(t_n)$). Clearly, there are two different regimes, a quasi-equilibrium regime with a slow decay for $L(t_m) \leq L(t_n)$ and the aging regime with a fast decay for $L(t_m) > L(t_n)$. Qualitatively, it is easy to understand that the system’s evolution changes upon progressing to not yet thermalized length scales. Indeed, we will show in the following how the two regimes may be analyzed quantitatively on basis of the effective dynamics.

Quasiequilibrium regime

As may be seen in fig. 5.1, the relaxation of $C(t_m, t_n)$ in the quasiequilibrium regime $L(t_m) \leq L(t_n)$ is very close to that of the equilibrium limit $C_{eq}(t_m) = \lim_{n \to \infty} C(t_m, t_n)$. The latter is shown in fig. 5.2 for various temperatures and, for comparison, also for per site weighting of the spins. Since this alternative puts more relative weight on faster (lower) levels of spins, the decay is faster, yet the overall behavior is similar to the per bond weighting.

The equilibrium autocorrelation $C_{eq}(t_m)$ measures the average overlap between an initial spin configuration of a fully (on a levels) thermalized system and a later spin configuration, in which spins up to level $m$ had a chance to flip (with respect to their initial state) due to equilibrium fluctuations. To begin, let us consider low temperatures with small typical flip
5 Results and discussion for the spin autocorrelation

Figure 5.1: Spin autocorrelation \( C(t_m, t_n) \) as a function of \( L(t_m) \) at \( T/T_c = 0.3 \) (open symbols) and \( T/T_c = 0.65 \) (filled symbols) for \( L(t_n)/L_0 = 2^0, 2^2, \ldots, 2^{14} \) (from left to right). Note the change in behavior upon surpassing \( L(t_n) \) in each case – as marked for the example \( L(t_n)/L_0 = 2^8 \) (\( \triangledown, \blacktriangle \)).

Figure 5.2: Equilibrium limit \( C_{eq}(t_m) \) of the spin autocorrelation as a function of \( L(t_m) \) at \( T/T_c = 0.1, 0.3, \ldots, 0.9 \) (from above). Open symbols are for our standard per bond weighting, the filled (gray) ones are for per site weighting.
rates, so that the linearization of (4.12) may be employed. We then have

\[ C_{eq}(t_m) \simeq \sum_{\alpha=0}^{m} w'_\alpha \left( 1 - 2 \bar{r}_\alpha(t_m) - \sum_{\gamma=\alpha+1}^{m} \bar{r}_\gamma(t_m) \right) + \sum_{\alpha=m+1}^{\infty} w'_\alpha \tag{5.1} \]

with the abbreviation \( w'_\alpha \equiv w_\alpha N_\alpha \). Here, the first sum over levels \( \alpha \leq m \) collects the contributions (4.12) of eventually flipped spins, whereas the sum with \( \alpha > m \) counts the unchanged spins of higher levels (which trivially have autocorrelation equal to unity). Due to the normalization of weighting factors \( w'_\alpha \), (5.1) may be written

\[ C_{eq}(t_m) \simeq 1 - 2 \sum_{\alpha=0}^{m} w'_\alpha \bar{r}_\alpha(t_m) - \sum_{\alpha=0}^{m} w'_\alpha \sum_{\gamma=\alpha+1}^{m} \bar{r}_\gamma(t_m) . \tag{5.2} \]

According to (3.9) (and the discussion there), the long-time limit of (5.2) gives the Edwards–Anderson order parameter, \( q_{EA} = \lim_{t_m \to \infty} C_{eq}(t_m) \). Its re-insertion in (5.2) yields a provisional result

\[ C_{eq}(t_m) \simeq q_{EA} + \sum_{\alpha=0}^{m} w'_\alpha \sum_{\gamma=m+1}^{\infty} \bar{r}_\gamma(t_m) + \sum_{\alpha=m+1}^{\infty} w'_\alpha \left( 2 w'_\alpha \bar{r}_\alpha(t_m) + \sum_{\gamma=\alpha+1}^{\infty} \bar{r}_\gamma(t_m) \right) \tag{5.3} \]

describing the approach of \( q_{EA} \) by \( C_{eq}(t_m) \). Clearly, any further progress requires information about the flip rates \( \bar{r}_\alpha \).

**Thermal equilibrium fluctuations**

As defined in sec. 4.2.2, \( r_\alpha(t_m, t_n) \) denotes the probability that a spin of level \( \alpha \) is flipped in epoch \( t_m \) (after a waiting time \( t_n \)) under the condition all the master spins are unchanged, i.e. that it experiences the same effective local field \( h^{(\alpha)} \) of (4.9).\(^1\) Thus, with the Boltzmann probability \( p_\pm = \exp[\pm h^{(\alpha)}/T] / (2 \cosh[h^{(\alpha)}/T]) \) for the spin to point up or down, respectively,

\[ r_\alpha = 2p_+p_- = \frac{1}{1 + \cosh[\Delta_\alpha/T]} , \tag{5.4} \]

where \( \Delta_\alpha \equiv 2 |h^{(\alpha)}| \) is the (free) energy difference associated with the flip (corresponding to an excitation energy \( F(L_\alpha) \) in the language of the droplet model).

As documented in fig. 5.3, we have investigated numerically the distribution \( \rho(\Delta_\alpha) \) of energy gaps in the fully thermalized system and found that it follows the same scaling form as the distribution of effective bonds in the MK renormalization,

\[ \rho(\Delta_\alpha) d\Delta_\alpha = \hat{\rho} \left( \frac{\Delta_\alpha}{\Delta_\alpha^0} \right) \frac{d\Delta_\alpha}{\Delta_\alpha^0} \tag{5.5} \]

\(^1\)In general, this is true only up to the variations of \( h^{(\alpha)} \) caused by an eventual change of temperature or perturbation of bonds after the waiting time \( t_n \), which will be considered in sec. 5.2.
Figure 5.3: (a) Equilibrium distribution of (free) energy gaps $\Delta_\alpha$ for excitations on level $\alpha$ in a scaling plot according to (5.5). As indicated, $T = 0.3T_c$ in the example. (b) Growth with length scale $L_\alpha$ of the characteristic value $\Delta_\alpha^*$ of energy gaps (normalized by $\Delta_0^*$; open symbols) in comparison to that of the standard deviation $J^{(\alpha)}$ of effective bonds in the MK renormalization (filled gray symbols). Note the virtually perfect match for each of the temperatures $T/T_c = 0$, 0.3, 0.7, and 0.9 (from above).

with the characteristic value (root mean square) $\Delta_\alpha^* = \sqrt{\langle \Delta_\alpha^2 \rangle} \sim J(L_\alpha/L_0)^\theta$, see part (b) of the figure. We find $\Delta_\alpha^* = cJ^{(\alpha)}$ with a constant $c \approx 3.22$ independent of temperature $T$. A naive estimate of this factor is $2 \cdot \sqrt{2} \approx 2.83$, where the square root comes from the addition of two random bonds, and the deviation may be ascribed to the system’s thermalization. We note that to our knowledge this is the first direct determination of the distribution $\rho$ and its postulated scaling (3.14) on the hierarchical lattice.

From (5.4), we may now calculate the average flip rate $\bar{r}_\alpha$ entering the expressions for the spin autocorrelation,

$$\bar{r}_\alpha = \int_0^\infty d\Delta_\alpha \frac{\rho(\Delta_\alpha)}{1 + \cosh(\Delta_\alpha/T)} = \frac{T}{\Delta_\alpha^*} \cdot \hat{\rho}(0) \int_0^\infty dx \frac{1}{1 + \cosh(x)} + O(T^2),$$

(5.6)

where the second step is an expansion for low temperatures with the remaining dimensionless integral equal to unity. In other words, excitations are considered only up to an energy of order $T$.

Consequences for the equilibrium autocorrelation

Insertion of (5.6) together with the scaling of $\Delta_\alpha^*$ into the expression (5.3) for the equilibrium spin autocorrelation yields

$$C_{eq}(t_m) \simeq q_{EA} + c' \frac{T}{J} \left( \frac{L(t_m)}{L_0} \right)^{-\theta} + O(T^2),$$

(5.7)

---

Footnote: For per site weighting of spins, there is an additional contribution $O((L(t_m)/L_0)^{-\theta+3})$ which, however, proves clearly negligible for (our) large $L(t_m)/L_0$, cf. the following text.
Figure 5.4: (a) Equilibrium spin autocorrelation $C_{eq}(t_m)$ at $T/T_c = 0.1, 0.2, \ldots, 0.9$ (from above; per bond weighting) plotted to verify the asymptotic behavior predicted in (5.7). The Edwards–Anderson order parameter $q_{EA}$ may be obtained as intercept with the abscissa of a linear extrapolation for small arguments $(L(t_m)/L_0)^{-\theta}$. (b) Results for $q_{EA}$ as a function of temperature for the case of per bond weighting ($\Delta$) and per site weighting ($\blacktriangle$) of spins. The (light gray) line $y = 1 - T/T_c$ is a guide to the eye.

where $c' = \hat{\rho}(0)/\{c \cdot 2^{\theta}(2^\theta - 1)\}$. For $q_{EA}$ itself, inserting (5.6) in the long–time limit of (5.2) gives

$$q_{EA} \simeq 1 - c'' T / J$$

(5.8)

where $c''$ is another constant.\footnote{Explicitly, $c'' = \hat{\rho}(0)/(2 - 2^{-\theta})(1 - 2^{-k}) \cdot \{c(1 - 2^{-\theta})(1 - 2^{-k+\theta})\}$, in which $k = 1$ for per bond weighting and $k = 3$ for per site weighting.}

Indeed, as shown in fig. 5.4, the expression (5.7) aptly describes the numerical results for $C_{eq}(t_m)$ on large length scales and may thus be used to extract $q_{EA}$. The latter then clearly exhibits the linear decay with temperature $T$ claimed in (5.8), see fig. 5.4(b). As for quantitative agreement, we find the constant $c'$ to be around 10% off numerically determined slopes, whereas $c''$, though correctly predicting a steeper decay of $q_{EA}$ for per site weighting, is $\sim 15\%$ (per site weighting: 30\%) above the actual slope close to unity in fig. 5.4(b). Concerning this graph, we note that the numerical uncertainty of the values for $q_{EA}$, whose determination becomes more difficult for increasing temperatures, is roughly demarked by the symbol size for $T$ close to $T_c$.

At this point, a natural question is why the above description of the (quasi–)equilibrium behavior, which was introduced as a low–temperature approximation, works reasonably well even rather close to $T_c$. For an answer, one should keep in mind that it applies on large length scales where, through the renormalization, the system departs from criticality for any $T < T_c$. Moreover, the sustaining correlations at large $t_m$ obviously stem from clusters (and realizations of the dynamics) with exceptionally few fluctuations, so that $C_{eq}(t_m)$ is dominated by specific ‘sub–ensembles’ of these – in a way similar to the mechanisms
discussed in Part I of this work. Certainly, if one was interested in a still more precise analytic treatment of the qualitative spin glass model, this would be a possible starting point.

**Aging regime**

Returning to the two–time (non–equilibrium) spin autocorrelation $C(t_m, t_n)$ of fig. 5.1, we now come to the second regime $L(t_m) > L(t_n)$, where the spin autocorrelation decays quickly as the system evolves on length scales not yet reached during the preceding waiting time $t_n$.

Specifically, in epoch $t_m$, spins at levels $n < \gamma \leq m$ are newly thermalized and thus will be flipped with probability $r_\gamma = 1/2$ (with respect to their still random orientation at the end of the waiting time). By the twisted boundary effect, any such a flip will erase the autocorrelation of the underlying cluster. On the other hand, with probability $(1/2)^{m-n}$ that none of its master spins has flipped, a cluster of level $n$ will retain its quasiequilibrium autocorrelation. Hence, we have

$$C(t_m, t_n) = \left(\frac{1}{2}\right)^{m-n} \left[C(t_n, t_n) - \sum_{\alpha=n+1}^{\infty} u'_\alpha \right] + \sum_{\alpha=m+1}^{\infty} u'_\alpha ,$$  

(5.9)

where, similarly as in (5.1), the last sum accounts for spins still immobile in epoch $t_m$, while, inside the square brackets, the analogous contribution is subtracted from the remaining $C(t_n, t_n)$ of (the end of) the quasiequilibrium regime.

Note that (5.9) is an exact relation, which may be derived purely formally also by inserting the trivial $r_\gamma = 1/2$ into (4.11). Setting in the respective $u'_\alpha$, we get$^4$

$$C(t_m, t_n) = C(t_n, t_n) \left(\frac{L(t_m)}{L(t_n)}\right)^{-\lambda}$$  

(5.10)

with $\lambda = 1$. Hence, the spin autocorrelation may be calculated exactly in the aging regime $L(t_m) > L(t_n)$ once $C(t_m, t_n)$ is known. Also, the aging regimes of arbitrary $C(t_m, t_n)$ naturally lie on a common master curve in a scaling plot of $C(t_m, t_n)/C(t_n, t_n)$ vs. $L(t_m)/L(t_n)$ (except for $t_n = 0$ in the case of per site weighting, see the preceding footnote).

To conclude the discussion of isothermal aging, we note that the central results (5.7), (5.10), and (5.8) agree with the scaling forms suggested by the droplet scaling theory of spin glasses [Fis88]. In fact, the latter may be derived by closely analogous, though (in absence of an underlying microscopic model) necessarily more abstract considerations, e.g. in what concerns the distribution of excitation energies $\rho(F_L)$ with the important property $\rho(0) > 0$, cf. sec. 3.2.3.

For the aging behavior (5.10), in particular, one should recognize that it follows quite directly from the construction of the model. Yet, while it may not offer fundamentally new insights, it may well augment confidence in the effective dynamics as a reasonable dynamical

$^4$ Again, for per site weighting there is another additional contribution $(L(t_m)/L_0)^{-1}(L(t_m)/L_0)^{-2} - (L(t_n)/L_0)^{-2}$, which is clearly subdominant except for $t_n = 0$, where it yields $C(t_m, 0) = (L(t_m)/L_0)^{-3}$. 

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scheme. In the following section(s), we shall build on this experience and, besides, also mention how the findings may be compared to experimental results.

5.2 Rejuvenation

5.2.1 Results from the effective dynamics

As one of our central issues, we hereafter investigate the implication of temperature chaos on aging and rejuvenation effects in temperature shift protocols \((T_1, T_2)\). As before, we monitor the autocorrelation function \(C(t_m, t_n)\) of spin configurations at the end of a waiting time \(t_n\) and in subsequent epochs \(t_m\), whereby, however, the temperature switched from \(T_1\) to \(T_2 = T_1 + \Delta T\) after the waiting time.

Figure 5.5 shows the typical behavior of \(C(t_m, t_n)\) as a function of \(L(t_m)\) after such a temperature shift. For early epochs \(t_m\), it resembles (qualitatively) \(C(t_m, t_n)\) in the isothermal case in fig. 5.1. Specifically, we can identify an aging regime for \(L(t_m) > L(t_n)\), for which, as will be shown shortly, the scaling (5.10) still holds exactly with the same underlying mechanism. So, as before, most interesting will be the autocorrelation in the limit of infinite waiting time, i.e. the analogue of \(C_{eq}(t_m)\), which we now denote by \(C_{\infty}(t_m) = \lim_{n \to \infty} C(t_m, t_n)\).

As is obvious from fig. 5.5, the latter decays to zero for \(t_m \to \infty\), while the isothermal \(C_{eq}(t_m)\) converges to the plateau \(q_{EA}\) following (5.7). Hence the aging effect, i.e. the increasing persistence of correlations for increasing waiting time \(t_n\), becomes progressively irrelevant and the system clearly exhibits rejuvenation, suggesting that temperature chaos effect is coming into play.

Of special interest are ‘twin’ studies, in which the autocorrelation \(C(t_m, t_n)\) observed after a \(T\)-shift \((T_1, T_2)\) is compared to that after a shift \((T_2, T_1)\), i.e. a perturbation of the same strength \(\delta \equiv |\Delta T|/T_c\) including the same temperatures \(T_{1,2}\) however with their roles interchanged. As also shown for the example in fig. 5.5, we generally find \(C(t_m, t_n)\) as a function of \(L(t_m)\) identical for both cases, a remarkable result with parallels to experimental findings to be discussed further below.

As mentioned earlier, rejuvenation may also be induced by adding a random perturbation to each bond (on level 0),

\[
J_{ij}^{(0)} \mapsto J_{ij}^{(0)} + \frac{\delta \tilde{J}_{ij}}{\sqrt{1 + \delta^2}},
\]

(5.11)

where the \(\tilde{J}_{ij}\) are independent Gaussian random numbers obeying the same statistics as the initial \(J_{ij}^{(0)}\), and the denominator is to keep the distribution \(P(J_{ij})\) unchanged. As an advantage, (5.11) readily yields short overlap lengths \(L^*(\delta) \sim \delta^{-1/\zeta}\) (cf. sec. 3.2.3) for large \(\delta\), which would be unattainable or at least require one of the temperatures \(T_{1,2}\) close to \(T_c\) in a temperature shift. Thus, bond perturbation is often employed in conventional simulations suffering from restrictions in system size (and simulation time). Since the very advantage of our approach is the practical absence of such restrictions, we use bond perturbation protocols merely for comparison in the \(T = 0\) limit of the dynamics.
Figure 5.5: Spin autocorrelation $C(t_m, t_n)$ as a function of $L(t_m)$ in twin temperature shift protocols $(0.3T_c, 0.7T_c)$ (open symbols) and $(0.7T_c, 0.3T_c)$ (filled symbols) for $L(t_m)/L_0 = 2^0, 2^2, \ldots, 2^{14}$ (from left to right). Note the virtually perfect match of open and filled symbols. In accordance with [Asp02], we find $L^*/L_0 = 2^2-2^5$ for the overlap length of bonds in the MK renormalization at these two temperatures, cf. sec. 4.1.3.

Corresponding results for $C_{\infty}(t_m)$ together with some additional data from $T$-shifts are displayed in fig. 5.6. There, for completeness, we also document the analogous results in the case of per site weighting of spins which, once again, prove closely similar.

Cluster analysis and masking of obvious effects

For the analysis, let us first concentrate on a temperature shift protocol $(T_1, T_2)$ for definiteness. As in the isothermal case, it is built on the flip probability $r_\alpha(t_m, t_n)$ of a spin $S(\alpha)$ under the condition of unchanged master spins. In compact form, we have

$$r_\alpha(t_m, t_n) = \begin{cases} 
0 & \text{for } \alpha > m, \\
1/2 & \text{for } n < \alpha \leq m, \\
p_+(T_1)p_-(T_2) + p_-(T_1)p_+(T_2) & \text{for } \alpha \leq m, n.
\end{cases} \quad (5.12)$$

Therein, the first two lines apply to spins not yet mobile or newly thermalized in epoch $t_m$, respectively. They hold identically in both isothermal and shift protocols and thus, being at the origin of (5.10), again yield this exact relation for the aging regime $L(t_m) > L(t_n)$.

By contrast, the third line in (5.12), which (together with the first) applies in particular in the limit of infinite $t_n$, is a generalization of (5.4) taking into account the shift $(T_1, T_2)$.
Explicitly, it reads

\[ r_\alpha = \frac{\exp\left[ h^{(\alpha)}/T_1 \right] \exp\left[ -\tilde{h}^{(\alpha)}/T_2 \right] + \exp\left[ -h^{(\alpha)}/T_1 \right] \exp\left[ \tilde{h}^{(\alpha)}/T_2 \right]}{4 \cosh\left[ h^{(\alpha)}/T_1 \right] \cosh\left[ \tilde{h}^{(\alpha)}/T_2 \right]} \tag{5.13} \]

\[ = \left[ 1 + \frac{\cosh\left[ h^{(\alpha)}/T_1 + \tilde{h}^{(\alpha)}/T_2 \right]}{\cosh\left[ h^{(\alpha)}/T_1 - \tilde{h}^{(\alpha)}/T_2 \right]} \right]^{-1}, \tag{5.14} \]

where \( h^{(\alpha)} \) and \( \tilde{h}^{(\alpha)} \) denote the effective local fields experienced by \( S^{(\alpha)} \) at \( T_1 \) and \( T_2 \), respectively.

Indeed, the distinction between \( h^{(\alpha)} \) and \( \tilde{h}^{(\alpha)} \) points out an important aspect, i.e. that a temperature shift affects the spin-flip probabilities in two ways: (i) it changes the effective couplings (and thus the effective local fields) due to the \( T \)-dependence of the RSRG transformations, and (ii) it changes the weighting of these fields in the Boltzmann probabilities. Hence, a natural interest is to isolate the subtle effect (i) from the obvious effect (ii), which is present in any system with thermally activated dynamics.

For this purpose, an intuitive ansatz is to consider a correlation function

\[ \hat{C}_{\alpha}(t_m) \equiv \frac{C_{\alpha}(t_m)}{\sqrt{C_{\alpha}(t_m)_{T_1} C_{\alpha}(t_m)_{T_2}}}, \tag{5.15} \]

which is 'normalized' by the isothermal autocorrelation functions at temperatures \( T_{1,2} \).
5 Results and discussion for the spin autocorrelation

To somewhat substantiate this ansatz, let us assume for a moment that effect (i) is absent, i.e. \( h^{(\alpha)} = \tilde{h}^{(\alpha)} \). Then, inserting \( T_2 = T_1 + \delta T_c \) in (5.14), we get to first order in the perturbation \( \delta \)

\[
r_{\alpha} = \frac{1}{1 + \cosh[2h^{(\alpha)}/T_1]} \left( 1 - c^\dagger \delta \right) + O(\delta^2),
\]

(5.16)

whereby the precise form of \( c^\dagger = c^\dagger(h^{(\alpha)}, T_1) \) doesn’t matter here. Applying the same expansion to the isothermal flip rate (5.4) at \( T = T_2 \) clearly yields a correction \( 2c^\dagger \delta \), i.e. twice the above value. For the spin autocorrelation, this leads to

\[
C_{\infty}(t_m) = C_{eq}(t_m)T_1 + c^\dagger \delta + O(\delta^2), \quad C_{eq}(t_m)T_2 = C_{eq}(t_m)T_1 + 2c^\dagger \delta + O(\delta^2)
\]

(5.17)

with a common function \( c^\dagger = c^\dagger(T_1) \) of not further specified form. In any case, (5.17) implies \( \dot{C}_{\infty}(t_m) = 1 + O(\delta^2) \) in the absence of effect (i). Thus, although this is a hypothetical consideration, it may give some hint of how the ‘normalization’ (5.15) works (as shall be demonstrated below).

**Temperature chaos**

Now we may focus on effect (i) which, as introduced above, captures the temperature dependence of the effective (renormalized) couplings. From the discussions in sec. 3.2.3 and sec. 4.1.3, it is clear that therefor the temperature chaos effect enters the effective dynamics. Thus, although the following treatment will be deliberately elementary, it will essentially amount to a collection and adaptation of the previous, more general arguments.

Here, we are interested in the induced flip probability \( \tilde{r}_\alpha(\delta) \) of a spin \( S^{(\alpha)} \). As a rough estimate, such a flip becomes likely if the typical change of effective couplings \( \Delta J^{(\alpha)} = J^{(\alpha)} - J^{(\alpha)}_{T_1} \) induced by the temperature shift exceeds the original energy gap \( \Delta_\alpha \) before the shift, i.e.

\[
\tilde{r}_\alpha(\delta) \sim \int_0^{2|\Delta J^{(\alpha)}|} d\Delta_\alpha \rho(\Delta_\alpha)
\]

(5.18)

with the distribution \( \rho(\Delta_\alpha) \) of (5.5).

Thereby, the physical mechanism leading to \( \Delta J^{(\alpha)} \) in the renormalization is as discussed before: \( 2J^{(\alpha)} \) is the difference of free energies of the cluster associated with \( S^{(\alpha)} \) between the two states with parallel and anti-parallel boundary spins, which below \( T_c \) have a ‘relative domain wall’ passing passing through \( L^{d-1}_\alpha \) links. Accordingly, one can write for the free energy change due to the \( T \)-shift \( 2\Delta J^{(\alpha)} \sim \Delta \{TS_\alpha\} \), where \( S_\alpha \) is the difference of the entropies associated with the two states.

As argued in sec. 3.2.3, \( S_\alpha \) may be considered the sum of random local entropy fluctuations associated with the \( L^{d-1}_\alpha \) links along the relative domain wall, yielding \( S_\alpha \sim L^{(d-1)/2} \). Concerning its temperature dependence, \( S_\alpha(T) \sim \sqrt{T} \) was pointed out in [Asp80] in discrepancy to the previously common assumption of a linear relation [Fis88]. Here, however,

\footnote{Explicitly, \( c^\dagger = \sinh[2h^{(\alpha)}/T_1]/(1 + \cosh[2h^{(\alpha)}/T_1]) \cdot T_c/T_1 \). Note also that we actually used a (potentially) signed \( \delta = \Delta T/T_c \) and an expansion in \( \delta T_c/T_1 \) for this illustrative calculation.}
we linearize in $\Delta T = T_2 - T_1$ anyway; explicitly

$$\Delta \{TS_{\alpha}\} \sim -T_2 L_\alpha^{(d-1)/2} \sqrt{T_2} + T_1 L_\alpha^{(d-1)/2} \sqrt{T_1} \sim -\frac{3}{2} L_\alpha^{(d-1)/2} \sqrt{T_1 \Delta T},$$

(5.19)

or, expressed with the dimensionless $\delta = |\Delta T|/T_c$ from above, essentially $\Delta J^{(\alpha)} \sim \delta L_\alpha^{(d-1)/2}$.

Insertion in (5.18) finally yields

$$\bar{r}_\gamma(\delta) \sim \int_{[\gamma \gamma]} \frac{L_\alpha^{(d-1)/2}}{L^*} dy \tilde{\rho}(y),$$

(5.20)

where we used the scaling of energy gaps (5.5) and $L^*(\delta)/L_0 \sim \delta^{-1/\zeta}$ is the overlap length (3.17). Based on these considerations, we may now distinguish two regimes in the behavior of the correlation function $\hat{C}_\infty(t_m)$.

**Weakly and strongly perturbed regime**

In the *weakly perturbed regime* $L(t_m) \ll L^*(\delta)$, the flip rates are generally small, and the last equation (5.20) gives $\bar{r}_\gamma(\delta) \sim \tilde{\rho}(0)[L_\gamma/L^*(\delta)]^{\zeta}$. Accordingly, we may adopt (5.2), which is linearized in the rates $r_\alpha$, to obtain

$$\hat{C}_\infty(t_m; \delta) = 1 - c^i \tilde{\rho}(0) \left( \frac{L(t_m)}{L^*(\delta)} \right)^{\zeta},$$

(5.21)

where $c^i$ is a constant (see also below). Quite importantly, this implies that the rejuvenation effect does not appear suddenly at the overlap length $L^*(\delta)$, but rather gradually emerges.

In the late stage $L(t_m) \gg L^*(\delta)$, which we call *strongly perturbed regime*, the flip rates become $1/2$, which corresponds to the equilibrium states at $T_{1,2}$ being completely decorrelated on these large length scales. Then similarly to (5.10) we expect

$$\hat{C}_\infty(t_m; \delta) \sim \left( \frac{L(t_m)}{L^*(\delta)} \right)^{-\lambda}$$

(5.22)

with $\lambda = 1$.

Equivalent effects are expected for the bond perturbation protocols. In fact, since we take these in the $T = 0$ limit of the dynamics, the situation is even simplified: In the limit of infinite waiting time $t_m$, the system is fully thermalized, i.e., in its ground state, and there are no thermal fluctuations. Thus, $C_{eq}(t_m) \equiv 1$, and the normalization (5.15) is ineffective—and also unnecessary, because the decay of $C_{\infty}(t_m)$ is exclusively due to effect (i).

As demonstrated in fig. 5.7, we successfully tested the predicted scaling (5.21) and (5.22) on our numerical results. As for the short-time scaling in part (a) of the figure, one may recognize that it works reasonably well, though not perfectly. Indeed, this can be traced back to an approximation made to obtain (5.21). Since it is somewhat technical without offering much additional insight, we defer its discussion to app. A.3. The long-time scaling in part (b), on the other hand, works very well. Quite remarkably, the form of the master curve is apparently universal for both temperature and bond shifts.\(^6\)

\(^6\)This is notwithstanding the fact that, as might be expected, a particular $\delta$ acts more strongly as a direct bond perturbation in (5.11) than indirectly as a $T$-shift $\delta = |\Delta T|/T_c$, cf. the conversion factor in fig. 5.7.
5 Results and discussion for the spin autocorrelation

![Graphs showing the decay of the normalized correlation function $C_{\infty}(t_m)$ as a function of $(L(t_m)/L^*(\delta))^\delta$ in a scaling plot according to (5.21). The data are for various $T$-shifts $(0.3T_c, (0.3+\delta)T_c)$ and bond perturbations (in the limit $T = 0$) as indicated. The straight line is a guide to the eye. (b) Analogous scaling plot of the long-time decay of $C_{\infty}(t_m)$ according to (5.22) with the same symbols as in (a). In both parts of the figure, a prefactor 25 is chosen for $L^*(\delta)$ in the $T$-shifts to merge the master curves with those from the bond perturbations.]

**Temperature shift symmetry**

A final point to consider is the remarkable twin $T$-shift symmetry, i.e. the finding mentioned above that the spin autocorrelation $C(t_m, t_n)$ as a function of the explored length scale $L(t_m)$ does not depend on the order of temperatures $T_{1,2}$ in a temperature shift protocol.

In fact, this is obvious for $L(t_m) = L(t_n)$, since then all the spins thermalized during the waiting time are re-thermalized at the new temperature (in an unchanged environment of not yet updated master spins). Thus, clearly, the roles of $T_{1,2}$ are interchangeable. From there, the $T$-shift symmetry carries over to all $L(t_m) > L(t_n)$ due to the exact scaling (5.10) in the aging regime.

Also, it is easy to see that the decay mechanism in the strongly perturbed regime, i.e. the decorrelation of equilibrium states for $T_{1,2}$ beyond the overlap length $L^*$, does not depend on which of both is the actual temperature during $t_m$. So the interesting point is the $T$-shift symmetry in the early, weakly perturbed regime, which we find virtually exactly also for simulated data of high statistical accuracy.\(^7\)

To elucidate this behavior, one may note that the flip rate $r_\alpha$ of (5.14) – and hence also (5.12) – indeed is invariant under a formal exchange of temperatures $T_{1,2}$ together with the corresponding effective fields $\hat{h}^{(\alpha)}$, $\tilde{h}^{(\alpha)}$. However, these fields depend on master spins which, for $L(t_m) = L(t_n)$, are thermalized at the first temperature of the $T$-shift protocol, i.e. either $T_1$ or $T_2$, up to length scale $L(t_m)$. In other words, going from a protocol $(T_1, T_2)$ to $(T_2, T_1)$ does not simply correspond to the mentioned exchange of temperatures and fields in (5.14). In fact, in the weakly perturbed regime, i.e. on length scales far below the overlap

\(^7\)For the data shown in fig. 5.5, for example, a conservative estimate of the standard deviation is $\sim 2 \cdot 10^{-4}$.
length, spin configurations at $T_{1,2}$ (of the master spins in question) will be similar. Thus, whereas not being an exact proof, the preceding argument may still give some hint as to why deviations from an exact $T$-shift symmetry, if occurring at all, should be rather small.

### 5.2 Rejuvenation

#### 5.2.2 Comparison to experimental findings

At this point, let us pause for a moment in the exploration of our model in order to point out some important parallels to experimental observations, i.e. the behavior of real spin glasses.

Specifically, we would like to comment on possibilities to identify in experiments the growing coherence length $L(t)$. While being inherent to the effective dynamics, such a length scale may be established in conventional (Monte Carlo) simulations of spin glasses by comparing two replica of the same system, see e.g. [Kom99, Ber02a]. In experiments, by contrast, it obviously has to be determined indirectly.

One possibility relies on the out of phase susceptibility $\chi''(\omega, t)$ (cf. sec. 3.1.2), for which the droplet model predicts [Fis88, Jön02a]

\[
\frac{\chi''(\omega, t) - \chi''(\omega)}{\chi''(\omega)} \sim \left[ \frac{L(1/\omega)}{L(t)} \right]^{d-q}.
\]

(5.23)

Therein, $L(1/\omega)$ is the typical size of droplet excitations induced by the alternating magnetic field and $\chi''(\omega)$ denotes the susceptibility in thermal equilibrium, for which there also exist scaling predictions (involving e.g. the order parameter), but which practically may be taken as fitting parameter (see the preceding references).\(^8\)

Together with the logarithmic growth law for $L(t)$ in (3.16), the last relation yields an explicit scaling prediction for the susceptibility which, indeed, has been tested successfully in [Jön02a]. As pointed out there, however, a decisive point is to take as unit time in (3.16) a critical correlation time $\tau_0(T) \sim \tau_0[1 - T/T_c]^{-\nu}$, where $\nu$ is the dynamical critical exponent and $\nu$ that for the correlation length, rather than simply the microscopic time scale $\tau_0 \sim h/J$. Interestingly, this may lead to effective scaling forms $\sim (\omega t)^{-k(T)}$ for the susceptibility with a temperature dependent exponent $k(T)$ as found in some simulations and experiments, see e.g. [Vin96, Kom99].

Another possibility to determine $L(t)$ is via the zfc magnetization $M_{\text{zfc}}(t, t')$, whose logarithmic derivative $S(t) \equiv H^{-1}dM_{\text{zfc}}/d\log(t)$ exhibits a maximum which may be identified as 'effective age' $t_{\text{eff}}$ of the spin glass [Lun83]. In isothermal protocols, $t_{\text{eff}}$ turns out to be of the order of the waiting time $t'$, whereas after a temperature shift $(T_1, T_2)$, the difference of $t_{\text{eff}}$ to the nominal $t'$ may be used to examine to what extent aging at both temperatures is accumulative [Gra88]. Most elegantly, this may be done in the already mentioned twin temperature shift protocols $(T_1, T_2)$, $(T_2, T_1)$, see [Jön02b].

Note that $t_{\text{eff}}$ is a monotonously increasing function of the waiting time $t'$, which in a $T$-shift protocol $(T_1, T_2)$ is influenced by two (potentially) competing effects: On one hand,

\(^8\)We note that recently there were suggestions to use also nonlinear susceptibilities for probing dynamical length scales in glassy systems, see e.g. [Bon05].
the higher $T_1$, the faster the thermalization during the waiting time\(^9\), hence, as a tendency, the higher is $t_{\text{eff}}$. On the other hand, the larger the shift $|\Delta T| = |T_2 - T_1|$, the stronger is the rejuvenation effect, i.e. the lower is the effective age $t_{\text{eff}}$ at the new temperature $T_2$.

In the present context, the important point is that, as shown in the preceding reference, all of this may be translated consistently in length scales $L(t)$ obtained from the logarithmic growth law (3.16) with the abovementioned refinements as well as the overlap length $L^*(\Delta T)$ as derived within the droplet model. Thereby, an obvious advantage of the analysis in terms of length scales is that these incorporate temperature and time dependencies in a unified way. In particular, a scaling very similar to (5.22) is found for the emergence of the rejuvenation effect and, most importantly, twin $T$-shift protocols yield symmetric effects in terms of length scales in analogy to our results in fig. 5.5.

### 5.3 Memory effects

Returning to our model with the effective dynamics, we finally investigate also memory effects. The most straightforward protocol to study these is a one step temperature cycle, in which the system evolves at a temperature $T_1$ for a time $t_n$ and subsequently at another temperature $T_2$ for a time $t_{n'}$. Then the temperature is put back to $T_1$ and we measure the spin autocorrelation.

In fact, as indicated in the following scheme,

$$\{t_n; T_1\} \rightarrow \{t_{n'}; T_2\} \rightarrow \{t_m; T_1\},$$

there are two natural generalizations of the previous two-time correlation function: One, which we keep calling the spin autocorrelation $C(t_m, t_{n'}, t_n)$, measures the overlap of spin configurations at time $t_n + t_{n'}$, i.e. immediately after the second $T$-shift, and after some additional time $t_m$. The other variation, which we call memory correlation $C_{\text{mem}}(t_m, t_{n'}, t_n)$, is calculated from spin states after the first waiting time $t_n$ and at $t_n + t_{n'} + t_m$; hence it measures directly to what extent the system ‘looks’ like after the first stage at $T_1$.

**Spin autocorrelation**

As demonstrated in fig. 5.8, there are (up to) three regimes discernable in the behavior of the ‘conventional’ spin autocorrelation $C(t_m, t_{n'}, t_n)$: Initially, i.e. for $L(t_m) < L(t_{n'})$, there is ‘pure’ rejuvenation. The system evolves on short length scales thermalized at $T_2$ during the second waiting time $t_{n'}$, so that $C(t_m, t_{n'}, t_n)$ follows the autocorrelation function $C(t_m, t_{n'}; \delta = |T_1 - T_2|/T_c)$ as after a simple $T$-shift ($T_2, T_1$), cf. fig. 5.5. In the case $L(t_{n'}) \geq L(t_n)$, this is the only regime, i.e. there is no memory to the first stage and $C(t_m, t_{n'}, t_n) = C(t_m, t_{n'}; \delta)$ for all $t_m$.

\(^9\)This is because the dynamics is thermally activated.
**Figure 5.8:** Spin autocorrelation $C(t_m, t_{n'}, t_n)$ as a function of $L(t_m)$ after one-step temperature cycling $T_1 \rightarrow T_2 \rightarrow T_1$ with $T_1/T_c = 0.3$ and $T_2/T_c = 0.7$. The duration of the first stage is varied as $L(t_n)/L_0 = 2^1, 2^2, \ldots, 2^{14}$ from left to right, while that of the second is $L(t_{n'})/L_0 = 2^4$ (open symbols) and $2^8$ (filled symbols). Note that the curves for $L(t_{n'}) \geq L(t_n)$ lie on top of each other (cf. text). The (gray) vertical lines indicate the recovery times $L(t_{n'})$ according to (5.25).

More interesting is the opposite case $L(t_{n'}) < L(t_n)$. There, for $L(t_{n'}) < L(t_m) \leq L(t_n)$, a plateau region shows up indicating that the system’s evolution recovers length scales already thermalized at temperature $T_1$ during the first stage $t_n$. Hence $C(t_m, t_{n'}, t_n)$ exhibits merely a weak decay similarly as the isothermal equilibrium correlation $C_{eq}(t_m)$, although it lies below the latter due to the initial decay. In a generic domain growth scenario, the time of the transition to this second regime characterized by a decelerated decay is referred to as recovery time $t_{n'}$, [Yos01]. It is determined by the condition

$$L_{T_1}(t_{n'}) = L_{T_2}(t_{n'})$$  \hspace{1cm} (5.25)

where the indices are to emphasize the temperature dependence of the growth of the coherence length.

Eventually, for $t_m > t_n$ (or $L(t_m) > L(t_n)$, since both are at $T_1$), our system evolves beyond the limits of the first thermalization at $T_1$ and $C(t_m, t_{n'}, t_n)$ enters a steep decay analogous to (5.10) – as always when $L(t_m)$ exceeds all previously thermalized length scales.

**Memory correlation**

In a way complementary information on the system’s evolution is contained in the memory correlation $C_{mem}(t_m, t_{n'}, t_n)$ displayed in fig. 5.9. It starts from a value $C_{mem}(0, t_{n'}, t_n) = C(t_{n'}, t_n; \delta = |T_1 - T_2|/T_c)$. Then, for $L(t_m) < L(t_{n'})$, length scales thermalized at $T_2$ during the second waiting time are (re-)thermalized at temperature $T_1$, so that the spin

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configuration becomes more and more similar to that after the first waiting time at \( T_1 \). Correspondingly, the memory effect manifests itself in a rise of \( C_{\text{mem}}(t_m, t_{n'}, t_n) \).

After recovery from the second stage, i.e. for \( L(t_m) \geq L(t_{n'}) \), the memory correlation becomes identical to the isothermal spin autocorrelation at \( T_1 \), \( C_{\text{mem}}(t_m, t_{n'}, t_n) = C(t_m, t_n)_{T_1} \), which is given by the rightmost curves (for the two exemplary values of \( t_n \), i.e. open and filled symbols) in fig. 5.9. Eventually, just as in fig. 5.1, there is the fast decaying aging regime for \( L(t_m) > L(t_n) \).

So far, the description applies to all \( L(t_n) \), \( L(t_{n'}) \). However, in the case \( L(t_{n'}) > L(t_n) \), which is rather trivial for \( C(t_m, t_{n'}, t_n) \), the memory correlation \( C_{\text{mem}}(t_m, t_{n'}, t_n) \) exhibits a marked plateau region in the range \( L(t_n) \leq L(t_m) \leq L(t_{n'}) \), i.e. between the initial rise and the eventual coalescence with the isothermal \( C(t_m, t_n)_{T_1} \).

For an explanation, let us first consider the value of the memory correlation for \( t_m = t_n \). It quantifies the overlap between two spin configurations which are both thermalized at temperature \( T_1 \) up to length scale \( L(t_m) \), however in different environments: Immediately after the first waiting time \( t_n \), all the (still immobile) master spins are random corresponding to infinite temperature. After the the second waiting time, by contrast, some of them (i.e. those pertaining to length scales \( \leq L(t_{n'}) \)) are thermalized at \( T_2 \), i.e. uncorrelated to their initial state. Thus, the situation is analogous to the aging regime in an isothermal protocol at \( T_1 \), cf. sec. 5.1, and we expect \( C_{\text{mem}}(t_m = t_n, t_{n'}, t_n) = C(t_m = t_{n'}, t_n)_{T_1} \) as confirmed by the data in fig. 5.9.\(^{10}\)

\(^{10}\)Note that we use the recovery time (5.25) in the preceding expression since, for \( T = T_2 \) during the second waiting time, the length scale in question is \( L(t_{n'}) \equiv L_{T_2}(t_{n'}) \neq L_{T_1}(t_{n'}) \).
5.3 Memory effects

In subsequent epochs, the master spins thermalized at \( T_2 \) are – level by level – updated at temperature \( T_1 \). Thereby, those of them which remain constant clearly do not alter the ‘isothermal’ overlap of the underlying cluster. Any flip of a master spin, on the other hand, is equally likely to make the environment more or less similar to its random state after the first waiting time, i.e. it equally likely increases or decreases the overlap. Consequently, \( C_{\text{mem}}(t_m, t_n', t_n) \) is constant until, upon the recovery time, the decay mechanism of the aging regime as discussed in sec. 5.1 applies.

To conclude the immediate discussion of the observed memory phenomena, we note that the data both in fig. 5.8 and fig. 5.9 are for per bond weighting of spins. We have confirmed that, as might be expected, per site weighting as well as bond cycling, i.e. going from the original to a perturbed set bonds and back at constant temperature, yields completely analogous results.

Interpretation in terms of ghost domains

The discussed relaxation in temperature cycling protocols is reminiscent of experimentally observed rejuvenation–memory effects [Vin96, Nor98], especially in what concerns the characteristic change of behavior beyond a time scale as proposed by the recovery time \( t_m' \) of (5.25). So our approach yields ‘realistic’ results and may be taken as expository model for a variety of memory phenomena.

However, it should be noted that our hierarchical model together with the employed very stringent dynamical scheme does not – or, at least, not clearly – exhibit all the mechanisms proposed recently in a comprehensive domain growth scenario [Yos03, Jön04], for which some indications in experimental data and results of Monte Carlo simulations on conventional lattices have been identified. To prepare the discussion of this point, which will be given by the end of the present Part II of this work, let us rephrase our above findings in terms of the so-called ‘ghost domain’ scenario.

The latter approach was introduced in [Yos01] using a spherical and the Ising version of the Mattis model (cf. sec. 3.2.1) for illustration. Its basic idea may be best captured by means of a simple caricature of a spin glasses’ evolution in a temperature cycle as in fig. 5.10: During the initial stage after the quench below \( T_c \), domains of the equilibrium state \( \Gamma_1 \) at temperature \( T_1 \) (and its spin-flipped counterpart \( \bar{\Gamma}_1 \)) form and grow; see the upper left picture. If we concentrate, for simplicity, on length scales \( L(t) \) beyond the overlap length \( L'(\Delta T) \), the arising domain structure will essentially look like uncorrelated noise as measured by the equilibrium state \( \hat{\Gamma}_2 \) at \( T_2 \), which is sustained by the lower left picture.

Accordingly, following the temperature shift to \( T_2 \) (middle column), the beginning growth of \( \Gamma_2 \) (or \( \bar{\Gamma}_2 \)) domains displays rejuvenation. Now the important point is that the previous domain pattern will not disappear immediately, but may rather persist for some time in the form of fading correlated noise imprinted on the new state; cf. the upper middle picture. In fact, the remanent ‘ghostly’ noise pattern is central to memory effects in the third stage (right column), where it acts as a bias on the initial conditions for the recommencing domain formation at \( T_i \); for example, it may bias small domains to nucleate with the ‘right’ sign

\footnote{For a more rigorous discussion of this scenario, we refer to the preceding references; see also chap. 7.}
5 Results and discussion for the spin autocorrelation

\[ \text{Figure 5.10: Sketch of a spin glass in a temperature cycling protocol } T_1 \rightarrow T_2 \rightarrow T_1 \text{ according to the ghost domain scenario. From left to right, the system's state by the end of each of the three stages (at } T_1, T_2, \text{ and again } T_1 \text{) is depicted. Thereby, the upper row contains imagined projections to the system's equilibrium state } \Gamma_1 \text{ at temperature } T_1 - \text{ let e.g. black regions stand for spins pointing in the direction prescribed by } \Gamma_1, \text{ while those in the white area point in the opposite direction (corresponding to } \Gamma_1). \text{ The lower row contains the same for the equilibrium state } \Gamma_2 \text{ at } T_2; \text{ cf. text.} \]

(either } \Gamma_1 \text{ or } \tilde{\Gamma}_1) \text{ so that they match upon coalescence into greater domains.}

Trying to adopt this intuitive scenario, we note that in our model slow (high level) spins play the role of ghost domains. After strong perturbations, configurations of faster spins which occupy a dominant portion of the volume of a cluster are changed - which manifests in the strong decay of the spin autocorrelation. Yet slower spins in the same volume retain their original configuration. Thus, similar to the preceding description, these slower spins act as remnant symmetry breaking fields by which both the original \textit{amplitude} and \textit{sign} of the overlap with respect to the equilibrium state can be restored, which again may be monitored by the spin autocorrelation as discussed above.

It is apparent from the comparison, though, that there are limits to the modeling of memory phenomena within our approach including the effective dynamics - apart from its naturally incomplete time resolution. To be specific, let us point out two aspects: First, consider the case } L(t_H) \geq L(t_n), \text{ i.e. a length scale explored during the second stage (at } T_2) \text{ exceeding that of the first waiting time (at } T_1). \text{ In our model, as argued above, there will be no memory. In a generic 'ghost' scenario, by contrast, it is not excluded that remanent correlations - although strongly extenuated - may in principle persist also within larger } \Gamma_2 / \tilde{\Gamma}_2 \text{ domains.}

This becomes relevant in more complicated (e.g. multi step) temperature protocols leading to corresponding multiple memory phenomena; cf. the 'memory dip' experiments described in sec. 3.1.2. While our model in principle allows for multiple memory effects, their occurrence hinges on a specific hierarchy of length scales, i.e. that the length scale explored during the stage 'to be remembered' shall surmount all those of subsequent stages.

As a second aspect, note that the dynamics of domain growth in the third stage (of a simple } T^-\text{cycle), expressed by } L_{T_1}(t) \text{ as from (4.8) in our approach, will presumably be altered by the presence of ghost domains. While this might possibly be incorporated to some degree into the effective dynamics, e.g. by assuming a modified growth law } \tilde{L}_{T_1}(t) \text{ for}
the third stage (cf. sec. 4.2.1), such an assumption must be justified itself, i.e. our approach as discussed so far may not offer genuine insight in this respect. As noted above, we shall come back to this issue.

Finally, we briefly mention a detail which is indeed described aptly by our approach. It is about the dependence of memory effects on the sign of $\Delta T = T_2 - T_1$, i.e. whether the spin glass is heated or cooled for the second stage in a $T$-cycle. Note that in some of the theoretical descriptions of spin glasses listed in sec. 3.2, namely those employing the idea of a fixed energy landscape in configuration space, memory effects for $T_2 > T_1$ may not easily be explained.

In the effective dynamics, as in most domain growth scenarios, this is merely a question of length scales. In fact, the data presented in fig. 5.8 and fig. 5.9 are in a case $T_2 > T_1$. Yet, since the domain growth is assumed thermally activated, it is obvious that a higher $T_2$ requires a much shorter waiting time $t_n'$ to reach a length scale $L_{T_1}(t_n)$, or, in terms of the ghost domain scenario, to exterminate remanent correlations. In accordance to this, memory phenomena in experiments are found more clearly for $T_2 < T_1$ (see e.g. [Vin96]).
5 Results and discussion for the spin autocorrelation
6 Extensions of the model

6.1 Effects of a magnetic field

6.1.1 General aspects and renormalization

As worked out in detail in the preceeding chapter, the effective dynamics on the hierarchical lattice exhibits a rich variety of aging, rejuvenation, and memory effects with parallels to experimental findings. Hence, it is natural to ask whether this approach may be extended to yield additional insight or to be further substantiated. This chapter is concerned with such extensions, which however have not been fully explored yet. Partly, this is because they go beyond the scope of the present work, but in part also because they point at more profound limitations of this type of approach to the complex dynamics of spin glasses.

A first, obvious 'candidate' is the inclusion of an external magnetic field $H$ into the effective dynamics, which is interesting for at least two reasons already mentioned in chap. 3: First, most experimental probes imply the application of an external field in order to measure the magnetic susceptibility $\chi$, (z)fc magnetizations etc.; hence the interest in what models predict specifically for such measurements. Second, the two main classes of theoretical approaches, i.e. the mean-field and droplet pictures, differ substantially in the effects ascribed to an applied external field.

Spin glass in an external field

In the infinite-range SK model (cf. sec. 3.2.2 and the references given there), the spin glass (frozen) state is found stable for fields below a critical value $H_c(T)$, which is zero for $T = T_c$ and diverges for $T \to 0$. So the curve given by $H_c(T)$ in an $H$-vs.-$T$ phase diagram, which is referred to as de Almeida-Thouless (AT) line, separates the spin glass phase from a simple paramagnetic phase. More precisely, the infinite number of states for zero field (and $T < T_c$) due to replica symmetry breaking decreases for increasing $H$ to a single state (replica symmetric solution) at $H_c(T)$.

In the droplet picture, by contrast, the spin glass state in principle is unstable against any non-zero field, as is revealed by a simple argument: The magnetic moment of a droplet of size $L$, i.e. the sum of all spins contained, scales as $\sim (L/L_0)^{d/2}$, whence its magnetic energy in an external field scales as $\sim H(L/L_0)^{d/2}$. From (3.13), the typical free energy

\footnote{This is sometimes associated with the names of Imry and Ma, who invoked it for the effect of a random field on a ferromagnet [Imr75].}
necessary to create such a droplet is $F_L \sim J(L/L_o)\theta$ with $\theta \leq (d - 1)/2$. It follows that in any dimension $d$ the (potential) spin glass state becomes thermodynamically unstable beyond a length scale

$$L_H \sim L_o \left( \frac{J}{H} \right)^{1/(d/2 - \theta)},$$

i.e. one expects a paramagnetic state with correlation length $L_H$. Note that the preceding exponent is larger than the chaos exponent $\zeta = d/2 - \theta$ for the overlap length $L^*$ of (3.17), in which sense the field $H$ constitutes a more severe perturbation to the ordered state than a temperature shift $\Delta T$.

For the magnetization (per site), this yields $M \sim (L_H/L_o)^{-d/2} \sim (H/J)^{d/(d - 2\theta)}$, i.e. a non-analytic dependence on the field. However, it should be born in mind that the above relations are assumed to hold asymptotically for large length scales; and there will certainly be a contribution to $M$ linear in $H$ from single spins or small-scale excitations.

Moreover, we have merely considered thermodynamic equilibrium so far. For the non-equilibrium dynamics, one may readily postulate a dynamic change of behavior when the explored $L(t)$ becomes of the order of $L_H$, which for a given experimental protocol might resemble an ‘effective’ AT line in the phase diagram; see e.g. [Fis88, Fis91]. Along these lines, the droplet model allows for a number of scaling predictions for the behavior of a spin glass behavior in a magnetic field (cf. also sec. 5.2.2), similar to those for zero field which we retrieved in the context of our approach in the preceding chapter. However, as shall become clear below, the inclusion of an external field raises specific issues on the hierarchical lattice.

### Renormalization including the field

As noted in sec. 4.1.2 (and discussed comprehensively in [Dro00]), the MK approximation for the RSRG on a (hyper-)cubic lattice and the exact renormalization transform on the hierarchical lattice exhibit distinct differences in the presence of a magnetic field $H$.

In the bond–moving scheme of the former, which is illustrated in fig. 4.2(b), there is some freedom in how to treat the field terms in the Hamiltonian pertaining to the spins deprived of any bonds. Specifically, the question is whether or how the field acting on these solitary spins shall be ‘moved’ together with the bonds, i.e. be taken away and added instead to the local field of still connected neighboring spins.

This question arises because, in order to capture correctly the system’s thermodynamics at low temperatures ($J = J/T \to \infty$, $H$ finite), it is undesirable to leave field terms with disconnected spins. Nor should these fields be moved to the spins remaining after the renormalization step, where they would accumulate and yield ever growing local fields $H_i$ even at high temperatures ($J \to 0$), whereas the local fields may be expected to saturate for vanishing bonds. The solution is to move the fields to the connected spins traced over in the renormalization step. This still may be done in different ways; yet, as noted in the preceding reference, results of the MK approximation are found largely independent of the precise implementation.
6.1 Effects of a magnetic field

On the hierarchical lattice, by contrast, the RSRG scheme is exact, thus leaving no freedom. For the derivation, let us again first consider the elementary chain of three Ising spins \( S_{0,1,2} \) as in fig. 4.2(a), now however with (potentially different) fields \( H_{0,1,2} \) additionally acting on them. Generalizing (4.2) ff., we have the partition function

\[
Z = \sum_{S_{0,1,2} = \pm 1} \exp \left[ J_1 S_1 S_0 + J_2 S_2 S_0 + \sum_{i=1}^{3} \hat{H}_i S_i \right]
\]

(6.2)

to be cast in the form

\[
Z = f(J_{1,2}, \hat{H}_{1,2}) \sum_{S_{1,2} = \pm 1} \exp \left[ J' S_1 S_2 + \hat{H}'_1 S_1 + \hat{H}'_2 S_2 \right],
\]

(6.3)

where, analogously to the bonds, \( \hat{H} \equiv H/T \). A straightforward calculation yields

\[
J' = \frac{1}{4} \log \left[ \frac{\cosh(J_1 + J_2 + H_0) \cosh(J_1 + J_2 - H_0)}{\cosh(J_1 - J_2 + H_0) \cosh(J_1 - J_2 - H_0)} \right],
\]

(6.4)

\[
\hat{H}'_1 = \hat{H}_1 + \frac{1}{4} \log \left[ \frac{\cosh(J_1 + J_2 + H_0) \cosh(J_1 - J_2 - H_0)}{\cosh(J_1 - J_2 + H_0) \cosh(J_1 - J_2 - H_0)} \right],
\]

(6.5)

\[
f(\ldots) = 2 \left[ \cosh(J_1 + J_2 + H_0) \cosh(J_1 + J_2 - H_0) \ldots \right.
\]

\[
\left. \times \cosh(J_1 - J_2 + H_0) \cosh(J_1 - J_2 - H_0) \right]^{1/4};
\]

(6.6)

the expression for \( \hat{H}'_2 \) is that for \( \hat{H}'_1 \) with \( \hat{H}_1 \) replaced by \( \hat{H}_2 \) and with \( J_1 \) and \( J_2 \) interchanged.

In dimension \( d > 1 \), similar to the field-free case, contributions to \( J' \) and \( \hat{H}'_{1,2} \) from parallel paths are summed and those to \( f(\ldots) \) multiplied, cf. the sum in (4.6).\(^2\) The explicit formulas together with an approximation useful for the numerics are given in app. A.4. In each case, as should be, the expressions of sec. 4.1.2 are recovered by taking the limit \( H \rightarrow 0 \).

**Properties of the RSRG transform**

The ‘flow’ of coupling constants and the associated fields for the renormalization on the hierarchical lattice is illustrated in fig. 6.1. In conformance with the droplet picture as described above, the standard deviation of bonds \( J^{(\alpha)} \) for \( T < T_c \) first grows as in the case without a field, cf. fig. 5.3(b). Then, upon a certain number of iterations \( \alpha \) of the transform, the typical strength of the bonds vanishes rapidly reflecting the disappearance of the spin glass state as long range ordered phase.

Thereby, it should be possible to identify the length scale \( L_\alpha \) corresponding to the maximum of \( J^{(\alpha)} \) with the length \( L_H \) of (6.1). For the bond-moving scheme on a cubic lattice, as noted in [Dro00], the thus obtained \( L_H \) is indeed found to grow as a function of \( (J/H) \) with an exponent \( \sim 0.80 \), which (at this level of precision) matches perfectly the one in (6.1),

\(^2\)As before, we will not actually need the prefactor \( f(\ldots) \) for our approach; see the footnote on p. 48.
1/(d/2 - θ) ≈ 0.80. As for the hierarchical lattice, our results for the spin autocorrelation and the growth of the fields are also compatible with this value (see below), whereas the aforementioned study inferred a greater exponent ~ 0.97 from the behavior of the bonds. Since, from our experience, such a deviation could be related to numerical subtleties and the finite sample size in the RSRG calculations (cf. app. A.1), let us remark that it might eventually be interesting to extend the analysis in [Dro00] within a deepened, more specialized study. In any case, the detailed growth of L_H may be an instance where the model on the hierarchical lattice deviates from ‘ideal’ droplet behavior.

Note that the occurrence of a maximum of J^(χ) in itself, i.e., that it does not just decrease monotonically, offers an intuitive possible explanation for an apparent AT line as suggested above. In fact, questions of this type are the main topic in the preceding reference; cf. the first mention of this study in the overview given in sec. 4.1.3.

Here, we also consider the evolution of the (additional) fields which come along with the renormalized bonds. Plotted in fig. 6.1 are the mean μ_H^(χ) and standard deviation σ_H^(χ) of the fields associated with the bonds in the α-th step of the renormalization. Note that these quantities refer to the extra contribution to the field at a site S_i^(χ) due to a single such bond J_ij^(χ), not to the total field H_i^(χ) acting on S_i^(χ) (which consists of two such contributions plus the field H applied initially to all spins, cf. below).

The behavior of μ_H^(χ) and σ_H^(χ) illustrated in the figure may be explained by simple scaling arguments, which we have validated also by varying the number p' = 2^{d-1} of parallel paths in the lattice; cf. sec. 4.1.1. The growth of the mean μ_H^(χ) may be understood as...
simple accumulation of a small bias received from initially \( z_0 / 2 = (p')^\alpha \) neighbors, hence
\[
\mu_H^{(\alpha)} \sim (L_\alpha / L_0)^{\log(p')/\log(2)} = (L_\alpha / L_0)^2 \text{ in } d = 3.
\]

However, let us emphasize that the mean \( \mu_H^{(\alpha)} \), though eventually illustrative, bears a peculiarity: From symmetry arguments, the bias just mentioned should average to zero. Accordingly, the value of \( \mu_H^{(\alpha)} \) and, in particular, the sign it takes are in fact sample–specific. So, the standard deviation considered in the following or, with similar behavior, the mean of the fields’ absolute values clearly are more meaningful physical quantities.

Indeed, in the first iterations \( \alpha \) of the renormalization, where \( J^{(\alpha)} \) still increases (and the spin glass state is ‘intact’), the standard deviation \( \sigma_H^{(\alpha)} \) scales as \( \sim H (L_\alpha / L_0)^{d/2} \), which is reasonable since it corresponds to the possible magnetization (or, more precisely, magnetic energy) of a region of \( \sim (L_\alpha / L_0)^d \) decimated spins. Once the bonds start to vanish, \( \sigma_H^{(\alpha)} \) is dominated by a plain accumulative process, namely the random addition of \( p' \) fields in each step of the renormalization, whence \( \sigma_H^{(\alpha)} \sim c (L_\alpha / L_0)^{\log(p')/2\log(2)} \) (\( = c (L_\alpha / L_0)^3 \) in \( d = 3 \)) with a prefactor \( c = c(H)^3 \).

Finally, let us briefly outline the role of the initial field \( H \), which so far has been understood to be applied uniformly to all spins, \( H_i^{(0)} \equiv H \). So, specifically, one may ask whether the properties of the renormalization transform are altered if the initial field is taken to depend on the level \( \beta \) of a spin \( S_i^{(\beta)} \); for example, it could be weighted by the connectivity, \( H_i^{(0)} \sim \gamma H \), or in an opposite, extreme case be applied merely to the fastest spins, \( H_i^{(0)} \equiv 0 \) for \( \beta > 0 \). Aside from the need to motivate such assumptions, e.g. by a corresponding interpretation of what higher level spins stand for in terms of sites on an ordinary lattice, we find the main features of the renormalization qualitatively unchanged for the examples discussed (with the field strength \( H \) in the same range as in fig. 6.1). In particular, the discussed exponents are the same, which is consistent with \( H \) not being involved explicitly in their derivation.

### 6.1.2 Results from the effective dynamics

With the discussed renormalization at hand, it is in principle straightforward to generalize the effective dynamics to protocols involving an external field \( H \). Thereby, generic interest is in small fields \( H / J \ll 1 \) (at low \( T \)), since our approach has been developed to allow access to large time and length scales such as a large \( L_H \). Moreover, when the spin glass state is not affected strongly before such large scales, there is arguably more reason to maintain the conceptual basis and interpretation of our dynamical scheme as treated in sec. 4.2.1.

In the following, we will employ a zero–field–cooling (zfc) protocol (cf. sec. 3.1.1), in which the system, following the initial quench from \( T = \infty \) to a temperature \( T < T_c \), is first evolved for a time \( t_n \) without the field and, subsequently, during time \( t_m \), in presence of the field. Other possibilities, in particular the ‘opposite’ field–cooling (fc), i.e. application of the field only during the first stage \( t_m \), will be mentioned shortly by the end of this section.

---

\(^{3}\)The scaling with \( H \) (not shown in the figure) may be estimated by matching the two expressions for \( \sigma_H^{(\alpha)} \) at \( L_H \) of (6.1), which yields \( c(H) = H (J/H)^{\xi/2} \) with \( \xi = 1/(d/2 - \theta) \approx 0.80 \); numerically, we find \( \xi \approx 0.76 \) (which is not identical to the deviation for \( L_H \) mentioned above).
Figure 6.2: (a) Spin autocorrelation $C_{zfc}(t_m, t_n)$ as a function of $L(t_m)$ in a zero-field–cooling protocol with $H = 0.01$ ($T = 0.3 T_c$, per bond weighting of spins); the first stages (without field) are $L(t_m)/L_0 = 2^0, 2^2, \ldots, 2^{14}$ (from left to right). (b) Similarly, zfc spin autocorrelation in the limit of infinite waiting time $t_n$, $C_{zfc}^{(\infty)}(t_m)$, for three different fields $H$ as indicated. Open symbols are for per bond weighting, filled ones for per site weighting of spins. The inset is a scaling plot of the ‘normalized’ $\tilde{C}_{zfc}^{(\infty)}(t_m)$ as a function of $L(t_m)/L_H$ in analogy to fig. 5.7(b), cf. text.

Spin autocorrelation

An overview of the behavior of the spin autocorrelation $C_{zfc}(t_m, t_n)$ in the zfc protocol is given in fig. 6.2. Obviously, results are very similar to those from a temperature shift protocol $(T_1, T_2)$ as analyzed comprehensively in sec. 5.2 – cf. in particular fig. 5.5 and fig. 5.6. Thus, regarding the spin autocorrelation, the application of the field $H$ appears to act in much the same way as a temperature shift $\Delta T$ or bond perturbation $\delta J$.

As a particular detail, note the scaling of the autocorrelation in the limit of infinite waiting time, $C_{zfc}^{(\infty)}(t_m) \equiv \lim_{t_n \to \infty} C_{zfc}(t_m, t_n)$, which is displayed in the inset of fig. 6.2(b). To see this, the data for different $H$ are ‘normalized’ by the equilibrium (isothermal, field–free) autocorrelation similarly as in (5.15), $\tilde{C}_{zfc}^{(\infty)}(t_m) \equiv C_{zfc}^{(\infty)}(t_m)/C_{eq}(t_m)$, and plotted as a function $L(t_m)/L_H$. Interestingly, we find the scaling, which proves $L_H$ to be the relevant length scale of the perturbation induced by the field, to work very well with the ‘original’ exponent $1/(d/2 - \theta) \approx 0.8$ in (6.1), i.e. we do not recover its deviation as reported from RSRG calculations (see the preceding section, incl. the footnote on p. 83).

From here, one could evidently go further and adopt much of the analysis of the preceding chapter. For conciseness, however, let us concentrate in the following on the most palpable effect of the field, i.e. the system’s magnetization.

Magnetization

To calculate the magnetization on the hierarchical lattice, it seems natural to apply the same weighting factors $w_{\alpha}$ for the contributions from spins of different levels $\alpha$ as for the
spin autocorrelation of (4.10) (cf. also below). So we define

$$M(t_m) = \sum_{\alpha} w_{\alpha} \sum_{i_{\alpha}} S_{i_{\alpha}}(t_m),$$

(6.7)

with sums and averages as in the aforementioned equation, and we again consider specifically per bond and per site weighting of spins. Also, it should be understood that the magnetization will in general depend on the waiting time(s) as well, $M = M(t_m, t_n \ldots)$.

In fact, the most pertinent question concerning this quantity has been raised by a study [Ric00], in which the authors failed to detect any waiting time dependence of the zfc magnetization in Monte Carlo simulations on the hierarchical lattice. Clearly, this is in sharp contrast to the behavior found in experiments – just recall sec. 5.2.2, where we mentioned the observation of $M_{zfc}(t', t')$ as a possibility to gauge the (effective) age of the sample. While we will comment further on the preceding study in the next section, let us point out here that the effective dynamics indeed exhibits a more ‘realistic’ behavior of our model.

Results for the zfc magnetization $M_{zfc}(t_m, t_n)$ following different waiting times $t_n$ are displayed in fig. 6.3. For the discussion, we note that obtaining such data in fact requires a relatively high computational effort even in the principally quite efficient effective dynamics. This is because the non-equilibrium effects in question truly turn out rather weak, whereas, in the small fields argued for above, the ‘signal-to-noise’ ratio of the magnetization is poor.

As for the field’s strength, coincidence of the ratio $M_{zfc}(t_m, t_n)/H$ for the two different $H$ in part (a) of the figure indicates that we are in a regime where the system’s response is linear in the field. Further, the magnetization for per site weighting is found to grow faster and higher altogether than that for per bond weighting of the spins, which may be expected since the former puts higher weight on the more mobile low-level spins.

In particular, however, $M_{zfc}(t_m, t_n)$ in both cases displays a dependence on the waiting time $t_n$ as genuinely found for spin glasses (see e.g. [Nor98]), i.e. that it grows more slowly for larger waiting time. Thereby, for our data, the limit of infinite waiting time, $M_{zfc}^{(\infty)}(t_m) \equiv \lim_{t_n \to \infty} M_{zfc}(t_m, t_n)$, is marked by the lowest of each of the two sets of curves in fig. 6.3(a).

To better visualize this finding, we consider in part (b) of the figure the ‘excess’ magnetization

$$M_{zfc}^+(t_m, t_n) = M_{zfc}(t_m, t_n) - M_{zfc}^{(\infty)}(t_m),$$

(6.8)

i.e. the amount by which $M_{zfc}(t_m, t_n)$ for a certain finite $t_n$ surpasses its limit for infinite $t_n$. Remarkably, $M_{zfc}^+(t_m, t_n)$ shows a maximum for $L(t_m) = L(t_n)$, thus reflecting the preceding waiting time. In this respect, it resembles the logarithmic derivative $S(t) = H^{-1} dM_{zfc}/d(\log(t))$ mentioned in sec. 5.2.2, whose (direct) application here is hampered by the limited time (or length) resolution of the effective dynamics.

**Fluctuation dissipation relation**

As final point in our discussion of the zfc results, let us report some observations concerning the interrelation of the spin autocorrelation (without field) and the observed zfc magnetization. In thermal equilibrium, these should be connected by the usual fluctuation dissipation
Figure 6.3: (a) Zfc magnetization $M_{zfc}(t_m, t_n)$ (per applied field $H$) as a function of $L(t_m)$ for per site weighting (upper set of curves) and per bond weighting of spins (lower set): $T = 0.3 T_c$. In both cases, the first stages are $L(t_m)/L_0 = 2^0, 2^1, \ldots, 2^5$ (from left to right; see also part (b)). Note the virtually perfect match of the data for $H/J = 0.1$ (open symbols) and 0.05 (filled symbols). (b) Similarly, the ‘excess’ magnetization $M_{zfc}^e(t_m, t_n)$ as defined in (6.8). For clarity, only the data for $H/J = 0.1$ ($T = 0.3 T_c$) are shown. Here, the dark (black) symbols stand for per bond weighting, gray ones for per site weighting of spins.

In the present context, as shown in app. A.5, the latter takes the compact form

$$\chi_{zfc}(t_m, t_n) \equiv \frac{M_{zfc}(t_m, t_n)}{H} = \frac{1}{T} \left(1 - C(t_m, t_n)\right),$$

(6.9)

provided one chooses the same weighting factors $w_\alpha$ for the autocorrelation (4.10) and the magnetization (6.7). Thereby, as indicated, the ‘integrated response’ $\chi_{zfc}(t_m, t_n)$ is simply the zfc magnetization per field.

Out of equilibrium, potential relations of correlations to the response in an external field are generally much less obvious. In fact, the FDT is not only a functional means to deduce linear susceptibilities from the usually simpler treatment without external field; conversely, deviations from its original form may also be taken as a gauge of the respective quantities’ falling out of equilibrium. Hence the study of the FDT and possible generalizations to non-equilibrium settings have attracted eager theoretical interest especially in all kinds of glassy systems (for an overview, see e.g. [Bou98, Cug03]).

Within analytically treatable mean field approaches, in particular, some specific results concerning the form of off-equilibrium deviations from the FDT have been derived [Cug93, Cug94]. Explicitly, violations of the FDT are captured by a factor $X(t, t')$ added to the

---

4 Recent developments concerning this aspect are discussed e.g. in [Lip05] (and references therein). It should be interesting to test some of those novel methods also in our system.
response function\textsuperscript{5}
\begin{equation}
R(t, t') = \frac{X(t, t')}{T} \frac{\partial C(t, t')}{\partial t'},
\end{equation}
which, by definition, yields $\chi_{\text{zfc}}(t, t')$ through the integral
\begin{equation}
M(t, t') = \int_{t'}^{t} ds \, R(t, s) H(s)
\end{equation}
with $H(t) = H_0(t - t')$ for the zfc protocol. For $t \ll t'$ (or $C(t, t') \geq q_{\text{EA}}$), one finds
$X(t, t') = 1$, in which case (6.10) is merely another manifestation of the equilibrium FDT immediately yielding (6.9).

For $t \gg t'$ ($C(t, t') < q_{\text{EA}}$), on the other hand, $X(t, t')$ deviates from unity in a way depending on the type of system considered (cf. below), yet, in the limit of large $t'$, is found to be a function of its arguments only through $C(t, t')$, i.e. $X = X(C)$. Hence the response altogether may be considered a function of $C(t, t')$,
\begin{equation}
T_{\chi_{\text{zfc}}}(t, t') = \int_{C(t, t')}^{1} dC \, X(C),
\end{equation}
which motivates to study the FDT and its violations in a $\chi_{\text{zfc}}$-vs.-$C$ plot (with $t$ as the plot parameter).

Note that, irrespective of whether all of this applies exactly to our model, such a parametric plot offers the advantage that the times $t_m, t_n$ do not appear explicitly, which allows to directly compare our findings to those of other approaches, e.g. the Monte Carlo simulations on the hierarchical lattice in [Ric00], without the need to (fully) specify the growth law for $L(t)$ (cf. sec. 4.2.1).

Our results from the effective dynamics are thus displayed in fig. 6.4. The data shown are for per bond weighting of spins; the corresponding plot for per site weighting looks very similar. For any waiting time $t_n > 0$, we find the equilibrium FDT of (6.9) fulfilled in the first epoch, i.e. when only the spins of level 0 are updated. However, starting from the second epoch, i.e. as soon as the renormalization comes into play through the effective dynamics, the $\chi_{\text{zfc}}$-vs.-$C$ curves (for fixed $t_n$) bend away from the straight FDT line. Finally, all of them approach the horizontal line marked by the ‘saturation’ value
$\chi_{\text{zfc}}(\infty) \equiv \lim_{t_m, t_n \to \infty} \chi_{\text{zfc}}(t_m, t_n)$.

Thereby, in contrast to the observations reported in the last-mentioned reference, we do not find $\chi_{\text{zfc}}$ to stay closer to the FDT line for increasing waiting time – rather the contrary applies. Accordingly, even in the limit of infinite $t_n$, the region where $\chi_{\text{zfc}}$ departs from the FDT line does not move towards the intersection of the latter with the horizontal $\chi_{\text{zfc}}(\infty)$ line. Moreover, as illustrated in the figure, neither the latter region, where the FDT ceases to be valid, nor the $\chi_{\text{zfc}}(\infty)$–FDT intersection correspond to the value of the order parameter $q_{\text{EA}}$ on the abscissa (cf. the next section).

According to the position of the factor $X(t, t')$ (or $X(C)$) in (6.10), the inverse slope of $\chi_{\text{zfc}}$ as a function of the correlation $C$ is occasionally interpreted as an ‘effective temperature’,
$T_{\text{eff}} \equiv -\left(\frac{d\chi_{\text{zfc}}(C)}{dC}\right)^{-1}$ [Cug97]. Adopting this interpretation, one might say that the effective dynamics actually increases the effective temperature. More generally, from the
\textsuperscript{5}Note our use of variables $t_m, t_n$ (alluding to epochs) and $t, t'$ (in a more general context) for basically the same quantities.
Figure 6.4: Parametric plot of the isothermal spin autocorrelation $C(t_m, t_n)$ (in zero field) vs. the zfc response $\chi_{zfc}(t_m, t_n)$ at $T = 0.3T_c$. The plot parameter is $t_m$, i.e. points along the curves correspond to consecutive epochs starting from $t_m = 0$ at $(0, 1)$. Waiting times are $L(t_n)/L_0 = 2^0, 2^1, \ldots, 2^7$ from below (open symbols as in fig. 6.3(a)). Also displayed is the limit of infinite $t_n$ ($\bullet$), the value of $q_{EA}$ for this temperature (from fig. 5.4(b)), and the line $y = 1 - x$ (dash-dotted) pertaining to the equilibrium FDT in (6.9). All data points are for per bond weighting of spins.

point of view of the mentioned mean field approaches, the whole plot in fig. 6.4 appears like a curious mixture of what is expected for different kinds of systems (see e.g. [Bar98, Cug03]): While the plateau marked by $\chi_{zfc}(\infty)$ (implying $X(C) = 0$ in that region) is like in simple coarsening models, the bent part of the $\chi_{zfc}$ vs. $-C$ curves (implying a nontrivial $X(C)$) is slightly reminiscent of mean field spin glass models or also numerical results for the EA model on an ordinary lattice. Let us emphasize, though, that on the present basis such comparisons are definitely speculative, cf. below.

Other protocols

Obviously, the investigation of the effective dynamics in presence of a magnetic field could be pursued further. Aside from a more detailed ‘cluster analysis’ in the spirit of the preceding chapter, this includes the collection of more data, e.g. for different temperatures, but also for other protocols.

As for the abovementioned field–cooling, however, let us note a potential subtlety (see e.g. [Fis91]): When the field is applied immediately after the quench below $T_c$ (till the end of the waiting time $t'$), which would be straightforward to implement in the effective dynamics, one obtains in the measurement time $t$ the so-called isothermal remanent magnetization (IRM). Yet, what is actually referred to as field–cooled magnetization $M_{fc}$ or, as measured during $t$, the thermoremanent magnetization (TRM), is obtained by applying the field already before the quench. Thus, if the quench is not ideal, i.e. infinitely fast from $T = \infty$, the TRM may
contain cooling rate effects; experimentally, one finds $\chi_{\text{TRM}} > \chi_{\text{IRM}}$ for not too high fields $H$ (cf. sec. 3.1.1).

On the theoretical side (i.e., in particular, considering ideal quenches), this is believed to be related to the nontrivial result for mean field spin glass models (see [Yos02])

$$\lim_{t \to \infty} \chi_{\text{eff}}(t, 0) > \lim_{t \to \infty} \lim_{t' \to \infty} \chi_{\text{eff}}(t, t'),$$

which, as may be seen in fig. 6.3 or fig. 6.4, comes as an equality in our approach so far. In fact, in the last-mentioned reference, the inequality is argued to hold also in an extended version of the droplet theory, which takes into account anomalously ‘soft’ droplet excitations in the presence of frozen-in domain walls. Based on additional data for our model, it should be very interesting to see to what extent these scaling concepts may be recovered in the present model and might help to explain our above findings.

Presumably, however, this will at some point reveal the same type of limitations of the spin glass model on the hierarchical lattice as described by the end of the preceding chapter. The same may be expected ultimately for more elaborate protocols, in which the application of the field is combined with temperature shifts or circles (see e.g. [Jön04]).

### 6.2 Monte Carlo dynamics

In the remainder of this chapter, we will discuss Monte Carlo (MC) simulations on the hierarchical lattice, i.e. the ‘direct’ simulation of the system’s dynamics based on single spin flips with thermally activated flip rates and, a priori, without reference to renormalization concepts (cf. sec. 4.2.1).

In fact, such simulations may serve as an extension to our approach in two ways: First, in their conventional form, they may be used to substantiate the assumptions made to motivate the effective dynamics, in particular the separation of time scales (4.8). Second, in combination with the effective dynamics, MC simulations might supplement a finer time and length resolution to our RSRG approach to the dynamics. We will outline these possibilities in order.

**Conventional MC simulations**

Since we have developed our novel approach to spin glass dynamics precisely to go beyond the limitations of classical MC simulations, we have not used them extensively. Still, we report here some of our results, as they may yield some insight into the ‘genuine’ dynamics of the EA model on the hierarchical lattice.

On the technical side, the MC simulations are straightforward to implement, the only exceptional part being the numbering and linkage of the spins, i.e. the initial coding of the hierarchical topology. Our system size is $L/L_0 = 2^5$ and we use standard Metropolis flip rates, $r = \tau_0^{-1} \min(1, \exp[-\Delta E/T])$, where $\Delta E$ is the difference in energy associated with the envisaged spin flip and $\tau_0$ sets the time scale (cf. also below). Thereby, we have tested – besides the most common ‘discrete time’ MC algorithm – the rejectionless ‘waiting time
method’ [Dal01], which previously has been applied to spin glass models on ordinary lattices [Sib03]. The method and experiences for our model are briefly described in app. 6.6.

Some illustrative results for the isothermal spin autocorrelation \( C(t', t') \) are displayed in fig. 6.5(a). Note that the ‘waves’ in the plotted data are reproducible (for other realizations of the random bonds and stochastic dynamics), i.e. they are not due to insufficient statistics.\(^6\) In the case of zero waiting time, an initially nearly exponential decay of \( C(t, t' = 0) \) for \( t' / \tau_0 \lesssim 1 \) is followed by a much slower decrease for \( t' / \tau_0 \gtrsim 1 \). As may be expected for thermally activated rates, the long–time decay is faster at higher temperatures \( T \), yet even above \( T_c \) does not proceed below \( \sim 0.1 \) within a (still conveniently accessible) simulation time \( t' \tau_0 \sim 10^5 \). So, apparently, one does not retrieve the much stronger decay of \( C(t_m, t_m = 0) \) obtained in the effective dynamics (as a function of \( L(t_m) / L_0 \)); see (5.10) or e.g. fig. 5.1.\(^7\)

Similarly, as may be seen in fig. 6.5(a), the isothermal autocorrelation for finite waiting times \( t' \) qualitatively exhibits the expected aging behavior, i.e. a slower decrease after a longer waiting time, yet the overall decay remains modest. The inset of the figure is a tentative scaling plot of \( C(t, t') \) vs. \( \log(t') / \log(t) \), whereby the darkest (and longest) curve corresponds to the shortest of the considered waiting times \( t' \). By contrast to what is suggested in [Ric00], we do not find this type of scaling to work convincingly for our data (cf. below). In fact, the data may be superimposed to a similar degree using \( [\log(t) / \log(t')]^{1/2} \) as scaling variable which, according to the assumed growth law for \( L(t) \) with the barrier

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\(^6\) The standard deviation of the data shown is below the symbol size.

\(^7\) In fact, an attempt to match the results of the two methods leads to the conclusion that the thermalization in the MC simulations may proceed only to small length scales \( L(t) \), which is corroborated further below.
exponent $\psi = d - 1$ (see sec. 4.2.1), should come close to the $L(t_m)/L(t_n)$ scaling found for the effective dynamics results in the aging regime.

Obviously, most of the interesting spin glass behavior as observed by means of the effective dynamics is not recovered in the MC results. Arguably, the reason for this may be seen in fig. 6.5(b). There, we display the level-specific cumulative flip rate $R_\alpha(t)$, i.e. the average number of flips performed by a spin of level $\alpha$ divided by the simulated time $t$ (since the initial quench from $T = \infty$). Correspondingly, a behavior $R_\alpha(t) \sim t^{-1}$ means that the spins of level $\alpha$ are frozen during that stage of the simulation, while the attainment of a constant, $R_\alpha(t) = \text{const.}$, may be taken as a (somewhat crude) indication of the respective level being thermalized.

The figure yields a distinct picture of the system’s dynamics: Shortly after the quench, i.e. up to a time $t/\tau_0 \simeq 1$, some of the randomly oriented spins flip to accommodate to the equally random local field created by their immediate neighbors. As described in sec. 4.2.1, this process tends to trap the system in a metastable state. Subsequently, frequently flipping spins of level 0, which are the most numerous, keep the overall flip rate high, whereas spins of higher levels turn out mobile only on increasingly large time scales. According to fig. 6.5(b), spins of level 3 and higher even at $T = 0.8 T_c$ virtually do not flip at all after $t/\tau_0 \simeq 1$ within an accessible time window (cf. app. A.6).

A simple trial to quantify the separation of time scales is presented in the inset of the figure. It shows $-T \log[R_\alpha(\infty)/\tau_0]$, where $R_\alpha(\infty)$ is the long-time limit of the flip rate as far as it may be read off the main plot, as a function of the length scale $L_\alpha/L_0 = 2^\alpha$. It should be clear, however, that the insinuated comparison to the asymptotic scaling of energy barriers $B(L)$ from (3.15) is somewhat questionable on the basis of the few available data.

**Connectivity weighted dynamics**

As already noted in sec. 4.2.1, the peculiar topology of the hierarchical lattice allows for different assumptions about its ‘natural’ dynamics – which is similar to the freedom in choosing weighting factors $w_\alpha$ for the measured quantities. In the aforementioned MC study [Ric00], for example, the authors used Metropolis flip rates $r$ with an attempt frequency weighted by the connectivity of the respective spin, i.e. a prefactor $1/\tau_0(\alpha) \propto 4^\alpha/\tau_0$ for a spin of level $\alpha$.

Despite low chances that it serves to mobilize appreciably higher level spins, we have also tested this possibility.\(^8\) Thereby, we find the cumulative flip rate $R_0(t)$ of the fastest spins nearly unchanged. More generally, the data for $R_\alpha(t)$ may be superimposed quite well onto those in fig. 6.5(b) by dividing the rates by $4^\alpha$ and, at the same time, multiplying times by $4^{\alpha/2}$. While the divisor for $R_\alpha(t)$ is obvious, the rescaling factor for $t$ may be attributed to the characteristic time scale on which the local field (created by $\sim 4^\alpha$ level 0 neighbors) changes significantly – i.e. an argument similar to those employed frequently in Part I of the present work.

\(^8\)Note that, while both the proposed *prefactors* and the energy barriers $B(L)$ grow exponentially with level $\alpha$, the latter appear in the *exponent* of the flip rates.
In any case, the important point here is that the system’s dynamics does not change qualitatively. In compliance to this, we find the overall behavior of the spin autocorrelation $C(t, t')$ - including the scaling behavior - also quite similar to that in fig. 6.5(a). The same is true for the per site weighted autocorrelation; hence we refrain from explicitly presenting results for all the possible combinations of weighting factors $w_i$ and flip rates.

After all, the whole scenario should not come as much of a surprise, since it confirms the arguments given in sec. 4.2.1 to motivate the effective dynamics. Yet, the extent to which the separation of time scales dominates the model’s dynamics is remarkable, as it effectively precludes the exploration of large length scales by conventional MC simulations.

Our observations thus offer a plain explanation for the difficulties in studying the sought-for sophisticated non-equilibrium effects such as aging by straightforward MC simulations (cf. above). That is, eventually, with an exception: As may be seen in fig. 6.3, in the effective dynamics the waiting time dependence of the zfc response $\chi_{zfc}(t, t')$ to a magnetic field is most pronounced precisely for small length scales $L(t)/L_0$ and $L(t')/L_0$. That no such aging at all was detected in [Ric00] then may be due to a combination of reasons, e.g. the elevated temperature ($T/T_c = 0.8$ as compared to 0.3 in the previous section) and the weighting per spin in the magnetization, which puts more weight on fast fluctuating lower level spins.\footnote{Unfortunately, there is no hint to the statistical accuracy of the results (as e.g. by means of error bars) given in that reference.} Note that the effect is relatively weak and demands elaborate statistics also in the effective dynamics. Within a deepened treatment of the model in presence of an external field, it should be possible to fully clarify this aspect.

At this point, one may of course ask whether the extreme separation of time scales as conspicuously revealed by MC simulations may be more than a technical problem – i.e. what is the appropriate dynamical scheme for the present system and how may it be interpreted in terms of more conventional models. In a more general context, i.e. not tied to the question of nominal time scales, we shall come back to these principal issues in the following summary chapter. Here, we remark that the necessity to reinterpret – or rescale – length and time scales should not be too surprising for this abstract and qualitative model, whose full properties, as outlined above, may not be unveiled by standard MC simulations. In view of some far reaching conclusions drawn previously from such an approach [Ric00], let us hence point out that the MC results at hand arguably should not be mistaken for the dynamics of the EA model on the hierarchical lattice, just as findings from the latter should not be mistaken for the droplet model of spin glasses.

**MC simulations with renormalized bonds**

From the preceding discussion, it seems natural to consider dynamical schemes which might combine the respective advantages of MC simulations, i.e. a detailed dynamics with fine time resolution, and of renormalization approaches, which allow easy access to large scales. Indeed, as mentioned before, a possible such scheme has been developed by Sasaki and Martin [Sas03] at about the same time as our effective dynamics.

Basically, they perform ordinary MC simulations on the hierarchical lattice, yet with
6.2 Monte Carlo dynamics

renormalized bonds of level \( n \) (called ‘NRG’ in the above reference); in other words, they take as their system one of the coarse grained lattices depicted to the right of the original one in fig. 4.3. For a change of temperature in the course of the simulation, in particular, any bond \( J_{ij}^{[n]} \) is replaced by another such renormalized coupling, which is calculated from the same initial Gaussian bonds, however at the new temperature.

The observed quantity, which is supposed to mimic the ac magnetic susceptibility (cf. the FDT in (6.9)), is given by

\[
\chi(\omega, t) = \frac{1 - Q(t + 2\pi/\omega, t)}{T},
\]

where \( Q(\tilde{t}, t) \equiv 1/N \sum_i \langle S_i(t)S_i(\tilde{t}) \rangle \) stands for the (per site weighted) autocorrelation of spins at times \( \tilde{t}, t \) (corresponding to \( C(\tilde{t} - t, t) \) in our notation); and the period \( 2\pi/\omega \) is 16 Monte Carlo steps (MCS; cf. app. A.6), while the total simulated time is of order \( 10^3 \) MCS.

Interestingly, for \( n \gtrsim 8 \), \( \chi(\omega, t) \) in a \( T \)-cycling protocol \( T_1 \to T_2 \to T_1 \), which is also compared to the isothermal case(s), exhibits aging, rejuvenation, and memory effects similar to the experimental results in fig. 3.3. To mention merely two specific findings from the simulations, let us note that, as in the effective dynamics, rejuvenation is found to set in gradually as experienced length scales \( L_n \) (as chosen by the system’s ‘generation number’ \( n \)) well below the overlap length \( L^* \) of (3.17). Moreover, for not too large \( n \), \( \chi(\omega, t) \) in the second stage (at temperature \( T_2 \)) may fall below its isothermal counterpart \( \chi_{iso}(\omega, t)\), which is obtained for constant \( T_2 < T_1 \) during the whole simulated time \( t \).

Thus, counterintuitively at first sight, the system may eventually appear older after aging at the ‘wrong’ temperature \( T_1 \neq T_2 \). The clue is that the thermalized length scale \( L(t) \) grows faster for higher \( T \), which below \( L^* \) may outweigh the effects of temperature chaos, cf. the discussion in sec. 5.2.2. So, in principle, this is also compatible with the behavior observed in the effective dynamics, if one translates the length scales \( L(t) \) to actual times \( t \).

As for the approach as such, the choice of the observable \( Q(t + 2\pi/\omega, t) \) in (6.13) is appropriate, since any ‘virtual’ lower level spins, whose decimation may be conceived at the origin of the employed renormalized couplings, should fluctuate on a faster time scale and hence should not contribute to \( Q \). Furthermore, at any change of temperature \( \Delta T \), the abrupt replacement of couplings (by ones which, for \( L_n \gtrsim L^*(\Delta T) \), are only weakly correlated to the former) induces additional mobility in the system. However, it should be mentioned that this straightforward use of the renormalized bonds in MC simulations indeed raises some conceptual issues.

As already noted in the original study [Sas03], the temporal course graining implied by a ‘generation number’ \( n \) is certainly temperature dependent. Thus, strictly speaking, \( n \) should be adjusted (or, alternatively, the simulated time \( t \) rescaled) in the different stages of a \( T \)-shift or circle. Moreover, one may object [Dro04] that the method does not reproduce the growth of energy barriers \( B(L) \) in (3.15) with the exponent \( \psi = d - 1 \).

To substantiate the latter aspect, we present in fig. 6.6(a) the mean flip rate \( R_n(t) \) as introduced before for the fastest and second fastest spins in systems with different \( n \). Thereby, note that the spins of nominal level \( \alpha = 0 \) (or 1) are actually such of level \( n \) \((n+1)\), if one thinks of the respective system with bonds \( J_{ij}^{[n]} \) as being created through \( n \)-fold application of the RSRG transformation; cf. above. Quite obviously, when directly compared for
Figure 6.6: MC simulations with renormalized bonds: (a) Mean flip rate $R_{0,1}(t)$ for spins of (nominal) level 0 (open symbols) and 1 (filled symbols) as in fig. 6.5(b), whereby the bonds in the system are renormalized couplings $J^{(n)}_{ij}$ of level $n$ as indicated; $T/T_c = 0.8$. (b) Supposed ‘energy barrier’ $B(L) = -T \log[R_{0,1}(\infty)/\tau_0]$ as a function of the length scale $L_n/L_0 = 2^n$ pertaining to the employed bonds; cf. the inset of fig. 6.5(b). The data are for spins of level 0 (○) and 1 (□) in the respective system, whereas the bent curve (×) is based on rescaled rates $R_{0,1}$ as explained in the text. For comparison, we also display the bonds’ standard deviation $J^{(n)}$ (●).

different values of $n$, the data merely exhibit a mild increase of time scales for spin flips on growing length scales $L_n$. This is also reflected in part (b) of the figure, where energy barriers $B(L)$ are supposed identifiable in the same – admittedly simplistic – manner as in the inset of fig. 6.5(b).

Arguably more reasonable than the direct comparison is a rescaling of times $t$ for the simulations with different $n$. For this purpose, we identify the spins of (nominal) level 0 in a system with a certain $n$ with those of level 1 in the corresponding lattice with $(n-1)$-fold renormalized bonds. Then, using the observed ratio $R_i(\infty)/R_0(\infty)$ in each such system, every rate $R_{0,1}$ of spins of ‘intrinsic’ level $n, n+1$ may be converted recursively to the time unit of the original $(n = 0)$ model.

In fig. 6.6(b), as may be expected, the corresponding data for the supposed barriers $B(L)$ indeed exhibit a stronger increase than without the rescaling. Yet, note that the outcome of this plain and, first of all, illustrative procedure reflects neither the ‘real’ growth of barriers (3.15) (with exponent $\psi = d - 1$) nor that of the effective couplings (with the ‘stiffness’ exponent $\theta \simeq 0.26$) in a simple manner.

Other hybrid schemes

Clearly, one can think of further dynamical schemes combining kinetic simulations and renormalization concepts. For example, as suggested at the beginning of the present section, the system could be evolved efficiently up to an epoch $t_n$ by means of the effective dynamics, whereupon MC simulations of the above type may supply details of the dynamics on all
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smaller time scales during that epoch. As a by-product, such a procedure should yield a
more comprehensive comparison of the dynamics in both cases, which may be used e.g. to quantify precisely the growth of length scales \( L(t) \).

To conclude our survey of MC and related methods, let us sketch a proposal to address on
a more fundamental level the problem of recovering the original time scales in simulations
with renormalized couplings. Indeed, the RSRG transform in sec. 4.1.2 is derived within
the framework of equilibrium statistical mechanics, whereby invariance of the partition
function is the central requirement. While, certainly, this is reasonable also for the study
of the dynamics (in a way similar to the condition of detailed balance for generic flip rates), one may ask whether the transform can be supplemented to capture explicitly also dynamical consequences of the coarse graining.

To this aim, let us compare flip rates in the original system to those in the corresponding
once coarse grained one. Specifically, consider a ‘branch’ within a cluster of level 2 with
boundary spins \( S^{(2)} \) and \( S^{(3,...)} \) of level 2 and higher, a central spin \( S^{(1)} \) of level 1, and
2 \( \times \) 4 level 0 spins \( S_{i}^{(0)} \), \( 0 \leq i < 8 \), located between \( S^{(1)} \) and \( S^{(2,...)} \), respectively; see e.g. fig. 4.1(a). For simplicity, we will write the spins without their level indicated, and we assume flip rates of the form \( r = \tau^{-1} \exp[-\Delta E/2T] \). The latter makes the expressions in the following more compact, although, we emphasize, the calculation may be performed similarly (e.g.) for the above Metropolis rates.

If the fastest spins \( \{ S_{i} \} \) are taken to be thermalized in their local fields \( h_{i} = J_{\gamma i} S_{\gamma} + J_{xi} S_{c} \), where \( \gamma \in \{ a, b \} \) (whichever is the nearest neighbor of the respective spin), the (time-
averaged) flip rate of the central spin \( S_{c} \) for given \( S_{a,b} \) becomes

\[
r_{c} = \frac{\tau^{-1}}{\prod_{i} 2 \cosh(h_{i})} \sum_{S_{i} = \pm 1} \exp \left[ \sum_{i} h_{i} S_{i} \right] \exp \left[ -\sum_{i} J_{xi} S_{i} S_{c} \right],
\]

which contains the equilibrium distribution of spins \( S_{i} \), cf. the text preceding (5.4); \( \dot{J}_{c} \equiv J_{c}/T \) as before. Upon insertion of the fields \( h_{i} \), we obtain due to obvious cancellations in the exponents

\[
r_{c} = \tau^{-1} \prod_{i} \frac{\cosh \left[ J_{\gamma i} S_{\gamma} \right]}{\cosh \left[ J_{\gamma i} S_{\gamma} + J_{xi} S_{c} \right]}.
\]

If, on the other hand, the spins \( \{ S_{i} \} \) are decimated by means of the RSRG, the envisaged
spin \( S_{c} \) is coupled directly to \( S_{a,b} \) via renormalized bonds \( J'_{\gamma c} \) calculated from the original
\( \{ J_{\gamma i}, J_{xi} \} \) using (4.6), whence the flip rate

\[
r'_{c} = \tau^{-1} \exp \left[ -\left( J'_{\alpha c} S_{a} + J'_{b c} S_{b} \right) S_{c} \right] = \tau^{-1} \prod_{i} \left( \frac{\cosh \left[ J_{\gamma i} + J_{xi} \right]}{\cosh \left[ J_{\gamma i} - J_{xi} \right]} \right)^{-S_{\gamma} S_{c} / 2}
\]

The idea, then, is to factor the ratio

\[
\frac{r_{c}}{r'_{c}} = \prod_{i} \left( \frac{\cosh[2J_{\gamma i}] + 1}{\cosh[2J_{\gamma i}] + \cosh[2J_{xi}]} \right)^{1/2}
\]

This is a special case of so-called \( \alpha \)-rates \( r = \tau_{0}^{-1} \exp[-(\alpha E' - (1 - \alpha)E)/T] \), where \( E, E' \) denote the system’s energy before and after the flip, for the ‘symmetric’ choice \( \alpha = 1/2 \); cf. e.g. [Rin02].
into an individual attempt frequency, i.e. a specific prefactor for the actually employed flip rate at site $S_c$, when performing (e.g.) MC simulations using the renormalized bonds $J'_{\gamma c}$. In the effective dynamics, as another option, one might think about updating spins not only according to their level, but also according to their individual ‘effective mobility’ $r_c/r'_c$, which might yield a finer temporal resolution than our previous one, i.e. a single representative state per epoch.

As for the form of the ratio (6.17), we note that it does not depend on the value of $S_c$. On general grounds, this is because, as may easily be shown, the rates $r_c$ fulfill the condition of detailed balance as formulated with the renormalized couplings, i.e. $r_c(S_c = +1)/r_c(S_c = -1) = \exp[2(J'_{\alpha c} S_a + J'_{\beta c} S_b)]$, just as genuinely do the rates $r'_c$ – a fact which, besides, may give some confidence into the ansatz (6.14). According to some first, preliminary numerical tests, (6.17) may grow for increasing levels in a way reminiscent of the (standard deviation of) renormalized bonds.

Let us re-emphasize, however, that these are preliminary results. Within an extended study, it should be interesting to explore the properties of the ratio (6.17) and, in particular, its influence on the system’s dynamics when it is employed in simulations as suggested. Thereby, it might well turn out that the straightforward averaging in (6.14) needs to be refined. In any case, the preceding discussion should point out, by way of example, the potential for further development offered by the this type of approach to the dynamics of spin glasses – or also other complex systems.
7 Summary of Part II

Synopsis

The subject of Part II of the present work is a novel approach to the dynamics of spin glasses on large length and time scales, which makes use of renormalization concepts in real space.

In chap. 3, we started by discussing what are spin glasses in terms of real materials and outlined their characteristic features. Thereby, emphasis was laid on the sophisticated non-equilibrium phenomena, i.e. aging, rejuvenation and memory effects, by which the disordered frozen state below the critical temperature \( T_c \) exhibits its unusual, strongly prolonged dependence even on details of the material’s past (primarily thermal) treatment. A number of examples from diverse other ‘glassy’ systems, which display – though typically less pronounced – similar phenomena, should substantiate the broader scope of the problem as well as the prototypical role arguably to be ascribed to spin glasses.

Further, we gave a survey of theoretical approaches employed to describe these and other glassy systems. As a main point, we noted the crucial involvement of large length and time scales, which are notoriously difficult to reach in conventional computer simulations. Hence we formulated the goal of our approach to spin glass dynamics as reflected in the above subject description.

In chap. 4, we therefore chose to study the common spin glass model of Edwards and Anderson, i.e. Ising spins interacting via Gaussian distributed nearest neighbor exchange couplings, however on a particular hierarchical type of lattice. As essential feature, this allows for an exact renormalization transform in real space closely analogous to the Migdal-Kadanoff approximation on ordinary (hyper-)cubic lattices. The usefulness and insight to be gained from the model should be testified by the recital of a number previous applications to spin glasses.

Still, our approach was among the first to use the hierarchical lattice to study spin glass dynamics. Thereby, as corroborated also by standard Monte Carlo simulations of the model (cf. below), the hierarchical topology evidently translates into a hierarchy of time scales or ‘epo\( \text{\text Hicks} \) on which distinct groups of spins are mobile, from which we developed our novel, computationally highly efficient scheme to capture the system’s effective dynamics across large length and time scales. Moreover, within the introduction of our central observed quantity, i.e. the spin autocorrelation function \( C(t_m, t_n) \), we anticipated that the effective dynamics is also amenable to an approximate analytical treatment.

In the main chap. 5, we then presented and analyzed in detail results for the spin autocorrelation. The usability of our approach was first demonstrated for isothermal aging,
whereby we found the system to closely follow scaling predictions from the droplet model of spin glasses, as e.g. for the scaling of energy gaps, and the linearity of the order parameter $q_{\text{EA}}(T)$ with temperature $T$ during a quasi-equilibrium regime as well as the scaling of $C(t_m, t_n)$ as function of explored length scales $L(t_m)/L(t_n)$ in the subsequent aging regime.

The analogy to the droplet model as well as generic experimental findings carries over to the aging and rejuvenation phenomena occurring upon a temperature shift or bond perturbation. In particular, rejuvenation sets in gradually on length scales well below the overlap length, i.e. the bond decorrelation length due to the temperature chaos effect. We thus identified weakly and strongly perturbed regimes with a scaling behavior for varying strength of the perturbation similar to experimental observations. In addition to that, the autocorrelation $C(t_m, t_n)$ as a function of length scale $L(t_m)$ comes out identical after temperature shifts $T_1 \rightarrow T_2$ and $T_2 \rightarrow T_1$. This nontrivial symmetry is especially remarkable since it was also detected in previous experimental studies employing thorough efforts to gauge relevant length scales $L(t)$.

Furthermore, we demonstrated that our approach may also be used to study memory phenomena in temperature or bond cycling protocols, whereby the overall behavior again reveals parallels to observations in real spin glasses. In our model, however, the occurrence of memory effects strictly depends on a specific order of length scales explored during the different stages, which points at limitations to recover within the hierarchical topology fine details of the domain structure – such as play a role in the so-called ghost domain scenario, cf. below.

The last chap. 6 was concerned with possible extensions of our model and its relation to other ‘dynamical’ approaches involving the hierarchical lattice. We first discussed the effects of an external magnetic field on the renormalization transform, whereby properties of the latter on the hierarchical lattice depart somewhat from those of the Migdal-Kadanoff approximation on conventional lattices. Still, for both the spin autocorrelation and the observed magnetization, the effective dynamics was demonstrated to reveal a much richer and more ‘realistic’ behavior (as compared to experiments) than surmised previously based on conventional Monte Carlo simulations. We also noted the form of the fluctuation dissipation relation, which might deserve further attention within an extended study of the magnetic field effects.

Moreover, we thoroughly analyzed the system’s behavior in conventional Monte Carlo simulations, whose severe practical limitations were found to issue precisely from the separation of time scales on which our effective dynamics relies. Likewise, we discussed the approach of Sasaki and Martin, which consists of Monte Carlo simulations using renormalized bonds. Its fine time resolution within a specific epoch is in a way complementary to the effective dynamics’ monitoring of the systems evolution across large length and time scales. We thus mentioned possibilities to combine both approaches within further developed ‘hybrid’ dynamical schemes and also sketched an idea to extend the renormalization transform in order to better capture the system’s dynamics.
Digression: relation to advanced domain growth scenarios

In order to assess, for the present, benefits as well as limitations of our approach, let us finally consider how it goes together with and what it may contribute to the currently most advanced spatial (domain growth) theoretical descriptions of spin glass dynamics. While many aspects are also interpretable within ‘alternative’ scenarios which do not rely on the chaos effect (cf. the corresponding remarks in [Sas03]), we will hereafter mainly refer to the theoretical framework [Yos03, Jön04] built on the ghost domain scenario, since the comparison to the latter appears most instructive especially in view of the results from the effective dynamics.

Fundamentally, domain growth theories constitute an attempt to capture the obvious complexity of the evolving spin glass state in a small number of characteristic parameters, for which one takes typical sizes $L_{T_{1,2}}(t)$ of domains of the respective equilibrium states $\Gamma_{1,2}$ (pertaining e.g. to different temperatures $T_{1,2}$). As a salient feature, our system on the hierarchical lattice naturally possesses distinct length scales, of which, moreover, also very large ones may be reached conveniently in simulations by use of the renormalization.

On this basis, as described above, our approach may indeed yield important insight into aging and rejuvenation processes – take e.g. the $T$-shift symmetry. As an already noted drawback, the (nominal) length scales in the system come in a discrete and exponentially growing hierarchy, which eventually has to be reinterpreted in view of the (quasi-)continuum of length and corresponding time scales of more ‘realistic’ models (on conventional lattices).

The inherent reduction of complexity, however, appears still more severe in the context of memory phenomena. An instant where this becomes particularly clear is the expression for the recovery time $t_{m^*}$, i.e. the time required by the system to ‘overgrow’ traces of the preceding second stage in a $T$-cycling protocol $T_1 \to T_2 \to T_1$ (see sec. 5.3). From straightforward length scale arguments which, as shown, genuinely hold in the effective dynamics, the appropriate condition is given by (5.25). Yet, in the refined scenario of the two last-mentioned references (to which we refer for details), the latter equation is argued to apply only in a weakly perturbed regime, whereas it should be replaced by

$$\frac{L_{T_1}(t_{m^*})}{L_{\Delta T}} = \left( \frac{L_{T_2}(t_{n'})}{L_{\Delta T}} \right)^{\frac{\lambda}{\lambda'}} \quad (7.1)$$

in case the system evolved beyond the overlap length\(^1\) $L_{\Delta T} \sim L'$ during the second stage $t_{n'}$. Therein, $\lambda$ is a novel dynamic exponent describing domain growth under biased initial conditions (i.e. ghost domains), while $\lambda'$ is in some analogy to our previous ‘$\lambda$’ in (5.10). Importantly, since typically $\lambda > \lambda'$, (7.1) may account for observed unusually large recovery times.

In fact, the basic improvement of this refined description is to consider not only length scales, i.e. domain sizes, but also properly defined ‘amplitudes’, i.e. the degree of ordering within the domains – as insinuated by the different shades of gray in the schematic fig. 5.10. Correspondingly, the theory allows to differentiate between the ‘outer growth’ (in size) and

\(^1\)Note that, to analyze experimental data, a renormalized length $L_{\Delta T}$ in (7.1) should be employed in order to account for typical finite cooling/heating times – which is yet another finding of [Yos03, Jön04].
the simultaneous ‘inner growth’ (in amplitude) of former ghost domains, i.e. a distinction otherwise lost in the definition of a coherence length $L(t)$ as single variable.

Obviously, as already noted by the end of chap. 5, such advanced theoretical scenarios involve fine details of the domain structure and its dynamics which, if present at all, appear only in a strongly modified form on the hierarchical lattice. This is clearly evident for the effective dynamics, which relies on a single, unambiguous $L(t)$; however, on a more fundamental level, certain limitations may also be ascribed to the topological peculiarities of the lattice discussed in sec. 4.1.1, in particular the restrictions on the form of domain walls.\footnote{Note that, after all, we found the effective dynamics to mimic rather well the ‘genuine’ long-term dynamics, i.e. it may not be considered the principal source of all of the discussed limitations.}

Additionally, though, it may be interesting to note that, for most aspects, it is rather that details may not be resolved on the hierarchical lattice than that the dynamical mechanisms on the latter were in some blatant discrepancy to those on conventional lattices. As an example, one may argue that inner and outer domain growth in principle do occur contemporaneously in our system, yet essentially become perceptible in sequence due to the discreteness and increasing separation of length scales. Thereby, what seems an oversubtle and purely notional argument might nevertheless be helpful in seizing the applicability of our model.

In a similar vein, one may note that the difficulties in recovering the sought–for non–equilibrium effects in our system by standard Monte Carlo simulations appear to resemble continual ambiguities in the interpretation of such simulations on conventional lattices (cf. below). Thus the deepened study of the relation of the Monte Carlo dynamics to ‘effective’ dynamical schemes might potentially yield insight well beyond the peculiarities of the hierarchical topology.

**Conclusions**

The potential of our approach may be estimated as follows: First, the model, which in a sense may be viewed in between a statistical analysis of the renormalization transform and a ‘fully’ detailed microscopic picture, properly displays and yields interesting insight into a number of aspects of the non–equilibrium dynamics of spin glasses. Thus, as noted, it should be interesting to see how far it may be extended with reasonable effort e.g. to ‘magnetic’ quantities.

Second, our approach, which lends itself to a semi–analytic description in the spirit of the droplet model, unequivocally shows aging, rejuvenation and memory effects. By contrast, corresponding simulations on conventional lattices and the proper interpretation of their outcome – not to mention that of experiments – generally prove difficult and are still under debate (see e.g. [Jim05, Mai05]). Hence, despite its limitations, our hierarchical system may serve as a working expository model for the corresponding theoretical scenarios.

Third, from a methodological point of view, the successful experience from employing renormalization concepts in the description of the system’s dynamics may motivate the development of further such approaches – although they might imply (at first sight) harsh
approximations and abstractions from the original system. Concerning spin glasses, instead of taking sites on a hierarchical lattice, one may e.g. think of considering dynamically identified clusters of spins on a conventional lattice. Such clusters could be distinguished by a (prevalently) fixed relative orientation of the inherent spins, as was done previously in some ground state search algorithms (see e.g. [Har04]). Clearly, many further ideas may be devised also for other complex or glassy systems with largely separated time scales.
7 Summary of Part II
Part III

Quasicrystalline order in two–dimensional binary dipolar systems
8 Motivation and static energy calculations

8.1 Introduction

Quasicrystals

Since the surprising discovery of sharp diffraction images with non-crystallographic symmetry in some rapidly quenched metal alloys by Shechtman et al. in 1984 [She84], quasicrystalline structures have attracted growing interest as an alternative type of structure of solid matter. As distinctive feature, quasicrystals possess long-range positional order in combination with a crystallographically ‘forbidden’ (e.g. fivefold) point group symmetry, which necessarily means aperiodic order. For two recent comprehensive reviews on these materials and their unusual properties, some of which will be mentioned in the following, we refer to [Suc02, Tre03]; an introduction and a more concise overview may be found in [Jan94] and [dBo05], respectively.

Basic characteristics of quasicrystalline structures may be understood by considering their representation as projections of a specific portion of a higher-dimensional regular lattice onto an embedded (hyper-)plane of irrational slope. In fig. 8.1, this is illustrated for a simple one-dimensional ‘prototype’ of a quasicrystal, i.e. the well-known Fibonacci chain (see e.g. [Jan94]), which consists of two types of segments ‘L’ and ‘S’ and is commonly constructed by iteratively applying the mapping \( \{ L \mapsto LS; S \mapsto L \} \) to an initial \( L \).\(^1\) An important observation from the illustrated projection procedure is the notion of quasiperiodicity: By approximating the irrational slope of the plane of projection, i.e. the golden mean \( \tau = (1 + \sqrt{5})/2 \), by a rational number \( \hat{\tau} \), one obtains a periodic structure similar to the original chain, i.e. a so-called periodic approximant. Thereby, the closer \( \hat{\tau} \) comes to \( \tau \), the better the approximation of the aperiodic structure (in whatever quantitative sense), yet the larger – as a tendency – the approximant’s period.

Indeed, quasiperiodicity may explain why quasicrystalline materials exhibit properties distinct from both amorphous solids and conventional crystals. Standard concepts in the description of crystals, e.g. the Brillouin zone and Bloch’s theorem for electronic states, apply, if at all, only approximately and in modified form. As practical examples, quasicrys-

\(^1\)Note that, since there are no non-crystallographic symmetries in dimension \( d = 1 \), the Fibonacci chain may, according to the above characterization, formally be regarded as a modulated crystal rather than a ‘real’ quasicrystal; cf. [Baa02].
Figure 8.1: Schematic illustration of the ‘projection method’ to obtain quasiperiodic structures: The Fibonacci chain results from projecting a particular stripe of a square lattice onto a parallel hyperplane, i.e., in this case a line. Obviously, when the slope of this line is irrational (as given here by the golden mean $\tau$), the resulting structure is aperiodic. Note that the indicated equation holds for both the lengths and the number ratio of segments $L$ and $S$ (cf. the references in the text).

Talline metal alloys have been found to exhibit a very low electric conductivity rising with temperature $T$ and falling with the degree of (quasicrystalline) order, while mechanical characteristics range from brittleness (at low $T$) to unusually high ductility and deformability (at higher $T$). Further observations in some of these materials, which are of potential technological interest, include low friction and wetting of surfaces, chemical resistance, high possible hydrogen storage, and high strength of quasicrystalline–crystalline composites (see the preceding references). Recently, quasicrystalline structures have been studied as photonic bandgap systems [Zoo00] and (photonic) laser materials [Not04]; however, these were ‘artificial’ (etched SiO$_2$) rather than generically grown atomic or molecular systems (cf. below).

Occurrence of quasicrystalline order

In the present Part III of this work, as detailed in the following, we shall less be concerned with properties of quasicrystals, whose comprehensive theoretical description still constitutes a challenge, but rather with the fundamental question which type of systems may exhibit this notable type of ordering, whereby, suiting to the general topic of ‘spin dynamics’, we will study this possibility for a (magnetic) dipolar system.

In fact, of the many systems with a quasicrystalline order identified by now in nature, most are ternary or, in a few cases, binary metal alloys. They fall into two broad classes, namely systems with icosahedral (i.e. ‘three-dimensional’ quasicrystalline) symmetry and such with dodecagonal, decagonal, or octagonal symmetry with respect to a distinguished axis, along which they are periodic (i.e. a stack of parallel sheets of atoms with in-plane quasicrystalline order). Very recently, though, quasicrystalline structures with fundamental building blocks much larger than single atoms have been found in micellar phases of dendrimers (i.e. tree-like molecules; [Zen04]). They represent a new mode of organization in soft matter and, as far as the scaling up of constituents may be continued, might become interesting in connection with the abovementioned photonic applications.
8.1 Introduction

On the theoretical side, the formation of quasicrystals could be reproduced in particular in simulations of two-dimensional binary mixtures with hard sphere or Lennard–Jones potentials [Wid87, Lan88] (for analogous approaches in $d = 3$, see e.g. [Rot90]). Thereby, the quasicrystalline structures were stabilized by tuning the distances corresponding to the minima of the interaction potentials to match the specific particle–particle distances.\footnote{Explicitly, the thus favored distances $d_{\alpha\beta}$ between neighboring particles $\alpha, \beta \in \{A, B\}$ of the two types $A$ and $B$ were chosen to obey $d_{AA}/d_{AB} = 2 \sin (\pi/5)$ and $d_{BB}/d_{AB} = 2 \sin (\pi/10)$, which may easily be seen to comply with a quasicrystalline reference structure with fivefold symmetry as presented in fig. 8.2.} Monte Carlo simulations and energy calculations revealed quasicrystalline order in these ‘tailor-made’ model systems to be a stable equilibrium state and (degenerate) ground state, while, in a physically quite reasonable scenario, molecular dynamics simulations yielded a quasicrystalline or an amorphous state depending on the cooling rate from high temperatures.

Recently, some evidence for local patterns with fivefold symmetry was found in experiments on two-dimensional binary mixtures of superparamagnetic colloidal particles [Zah99, Wen00, Kön03, Kön04]. The potential occurrence of quasicrystalline order in these systems, as a hint to which the reported observations could be interpreted, indeed would be very interesting for several reasons: First, colloidal systems are often considered as well controllable and conveniently observable model realizations of other condensed matter phenomena (on the atomic or molecular scale), which could thus be exploited also for the study of quasicrystals. Moreover, precisely because of consisting of such large building blocks (e.g. up to $\sim 50 \mu\text{m}$ in [Wen00]), the hypothetical colloidal quasicrystals could find their own specific applications; cf. above. For all of these purposes, a practically important task for a computational treatment would be to provide optimized parameters for the occurrence of the envisaged structures.

On a more fundamental level, still, it arises the intriguing question: Can there be quasicrystalline long–range order in a binary dipolar system despite the fact that the dipolar interaction potential does not possess tunable intrinsic length scales? Hereafter, we will tackle this problem as outlined in the following.

Outline of the present approach

Specifically, we investigate the occurrence of quasicrystalline order in binary mixtures of point dipoles with varying dipole strengths. As a two-dimensional reference structure with fivefold symmetry, we use the prominent rhombic binary tiling and decorate it with two types of dipoles as described the following section. In a first test of this structure as a possible system state, we then let it relax mechanically and thus identify a certain range of dipolar strength ratios $D$ (of the two types of particles) where the mechanically stable configuration preserves the long–range quasicrystalline order.

However, while such a configuration corresponds to a local minimum in the potential energy surface of the many–particle system, its global features are still undetermined. In one possible scenario, it could correspond to the ground state, a thermodynamically stable state within a certain parameter regime (of temperature, mixing ratio and dipolar strength ratio). Alternatively, it could correspond to a metastable state which, below some freezing
temperature, becomes separated from other states by free energy barriers diverging in the thermodynamic limit of infinite system size. This, indeed, would be reminiscent to the behavior of mean field spin glass models discussed in sec. 3.2.2 in Part II of the present work. In all of these cases, one would expect the dipolar system to evolve in time at some finite temperature and ultimately build up permanent long-range quasicrystalline order if its initial configuration belongs to the attraction basin of the quasicrystalline state. In another scenario, the mechanically relaxed quasicrystal structure could correspond to a metastable state from which the system escapes when it surmounts a finite free energy barrier. Nevertheless, it would then be interesting to see e.g. whether the quasicrystalline ordering exists locally in the disordered equilibrium state of the system.

To evaluate the global features as just discussed, we perform a number of investigations. So, by the end of the present chapter, we will compare the energy of several plausible alternative ground state structures with the energy of the quasicrystalline state. In the subsequent chap. 9, we shall go beyond mechanical equilibrium and static energy calculations in order to assess dynamical properties of the structure. In particular, we will study its stability, for varying dipole strength ratio, with respect to small random perturbations of the particle positions in the ideal decoration and, moreover, perform extensive Monte Carlo simulations to analyze the characteristics of quasicrystalline order at finite temperatures. Thereby, since conventional Monte Carlo techniques turned out to be ineffective for the thermalization of the system, we applied a parallel tempering protocol to reach thermodynamic equilibrium. Finally, our results will be discussed in chap. 10.

8.2 Model

8.2.1 System parameters and order parameter

Basic model description

Corresponding to the experimental situation [Wen00, Kö03], we consider particles moving in a plane with their dipolar moments all pointing in the same direction perpendicular to the plane. In experiments this can be realized by letting superparamagnetic particles float on a liquid meniscus in an external magnetic field.

To investigate quasicrystalline ordering, we choose as a reference structure the two-dimensional binary tiling, which consists of two types of rhombuses put together by certain matching rules [Gäh94]. The tiling, which is depicted in fig. 8.2, is a typical example for a quasicrystalline pattern with fivefold symmetry. Similarly as a crystal results from a periodically repeated unit cell decorated by atoms, we here obtain the ideal quasicrystalline reference structure by decorating the rhombuses as illustrated in the figure and described in [Lan86]. According to the number ratio of small to large rhombuses in the tiling, \( N_s / N_b = \tau - 1 \), where \( \tau \) is the golden mean as introduced above, we thus get a system of \( N_A \) strong A and \( N_B \) weak B dipoles with a mixing ratio \( x = N_A / (N_A + N_B) = \tau / (2 + \tau) \approx 0.447 \). The (overall) number density of dipoles is \( \rho \approx 1.231 \, a^{-2} \), where \( a \), later taken as our length
8.2 Model

Figure 8.2: The binary tiling decorated by strong (A) and weak (B) dipoles. The angles enclosed by the edges of its two rhombic building blocks are all multiples of $\pi/5$, which gives rise to the non-translation invariant 10-fold symmetry.

unit, is the edge of a rhomb as indicated in fig. 8.2.3

This decoration of the tiling was already used in previous simulations of binary Lennard–Jones and hard sphere systems [Lan86, Wid87, Lan88]. However, in contrast to these simulations, the potential of the dipolar system cannot be optimized in an obvious way to support the formation of the quasicrystalline structure.

The interaction energy of two parallel magnetic moments $m_1$ and $m_2$ at a distance $r$ is (in SI units)

$$E_{\text{int}} = \frac{\mu_0 m_1 m_2}{4\pi r^3}.$$  

(8.1)

In the following, let us switch to dimensionless quantities. As announced above, we choose as unit length the edge $a$ of a rhomb, i.e. the distance of two neighboring A– and B–dipoles in the reference structure. As unit of energy (and temperature $T$) we choose the interaction energy $(\mu_0/4\pi) \cdot (m_A m_B / a^3)$ of two such dipoles. After introducing the ratio $D \equiv m_A / m_B$ of dipole strengths $m_A$ and $m_B$, the interaction potentials of two A–dipoles, an A– and a B–dipole, and two B–dipoles at distance $r$ are

$$E_{AA} = Dr^{-3}, \quad E_{AB} = r^{-3}, \quad E_{BB} = D^{-1}r^{-3}.$$  

(8.2)

The dimensionless form clarifies that the dipole strength ratio $D$ is the only additional parameter in our problem besides the temperature $T$ and the (fixed) mixing ratio $x$. This is a direct consequence of the scale-free dipolar interaction potential (which, let us recall, gave rise to the in a related sense ‘scale-free’ field distributions considered in Part I of the present work).

3In full precision, we have $\rho = \tau / [\sin(2\pi/5) + (\tau - 1) \sin(\pi/5)] a^{-2}$. 

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**Order parameter(s)**

In order to quantify the degree of quasicrystalline order, we define the order parameter

\[
\phi = \frac{1}{2N_p} \sum_{j,k} \Theta(r_{\text{max}} - r_{j,k}) \exp(i \cdot 10 \alpha_{j,k}),
\]

where, as before, \(\Theta(.)\) is the usual step function (\(\Theta(x) = 1\) for \(x > 0\) and zero else), \(N_p = \sum_{j,k} \Theta(r_{\text{max}} - r_{j,k})\) is the number of pairs of dipoles with distances \(r_{j,k}\) smaller than \(r_{\text{max}} \equiv 1.15\), and \(\alpha_{j,k} \equiv \angle(r_{j,k}, \hat{e})\) is the bond angle between the pair vector \(r_{j,k}\) and an arbitrary but fixed direction \(\hat{e}\). Since the bond angles in the binary tiling are all multiples of \(\pi/5\) (cf. fig. 8.2), \(\phi = 1\) in the ideal quasicrystalline structure, while \(\phi = 0\) for a system without tenfold bond orientational order. The value \(r_{\text{max}}\) is slightly smaller than the distance of the two A particles in the fat rhomb shown in fig. 8.2. Eliminating these pairs from the sum in (8.3), on the one hand, allows us to encompass slightly displaced ‘nearest neighbors’ but, on the other hand, guarantees that bond angles different from multiples of \(\pi/5\) are not counted in the ideal quasicrystalline configuration.

In addition to (8.3), we also consider the \(n\)-fold local bond orientational order parameters

\[
\tilde{\phi}_n = \frac{1}{2N} \sum_{j} \frac{1}{N_j} \sum_{k} \Theta(r_{\text{max}} - r_{j,k}) \exp(i \cdot n \alpha_{j,k}),
\]

where \(N_j = \sum_k \Theta(r_{\text{max}} - r_{j,k})\) is the number of neighbors of dipole \(j\). Since the absolute value is taken before averaging over all \(N\) dipoles, the \(\tilde{\phi}_n\) are sensitive to the \(n\)-fold symmetric arrangement of nearest neighbors around any dipoles, but insensitive to the spatial variation of the bond orientations. Naturally, we have \(\tilde{\phi}_{10} \geq \phi\).

In this context, let us note that, for the analysis of the structures obtained in the dynamical (Monte Carlo) simulations, we will in sec. 9.2 consider a further quantity, i.e. the bond-order correlation function \(g_n(r)\) (see (9.2)), which, in a way, may be viewed as a generalization of the above parameters (8.3) and (8.4). As shall become clear then, this is because its (variable) argument \(r\) determines the distance over which it probes bond orientational symmetries.

**Rational approximants**

To conclude the exposition of our model system, let us discuss some practical aspects of the computer simulations. First, we note that, due to the missing translational invariance, standard periodic boundary conditions cannot be imposed to ideal quasicrystalline systems. To resolve this problem, it is convenient to use rational approximants of quasicrystals as described in [Ent88]. The basic principle underlying these was sketched in sec. 8.1 together with the projection method to obtain quasiperiodic patterns; for a structure as in fig. 8.2, one may e.g. start from a five-dimensional hypercubic lattice. Purely pragmatically, though, a rational approximant may be viewed as a rectangular part of the ideal quasicrystalline structure which is chosen such that it may be exposed to periodic boundary conditions without too much distorting the local quasicrystalline ordering.
From a number of different rational approximants, we mainly used a small one with 890 dipoles (398 A, 492 B dipoles) and size $24.80 \times 29.15$, and a larger one with 1700 dipoles (760 A, 940 B dipoles) and size $34.27 \times 40.29$. For both approximants, we find the order parameter $\phi \approx 0.9998$ close to the ideal value $\phi = 1$. We have no indication that the type of approximant is decisive for the conclusions obtained below.

To speed up the computer simulations, we store the dipolar interaction energies (and the forces) in a large matrix by discretizing the set of possible distances $r_{i,j}$ and use a linear interpolation scheme between the matrix entries. As an advantage, the matrix has to be calculated only once for all the simulations, and the computer memory access is usually much faster than the repeated evaluation of the original mathematical expressions. Details of the method and how to take into account the periodic boundary conditions are outlined in app. B.1.

As an example, for the two approximants described above we use a $4044 \times 4754$ matrix for the energies and a $2022 \times 2377$ matrix for the forces. We find the underlying discretization of space for the energies or forces on a length scale of order $10^{-2}$--$10^{-3}$ to have no noticeable influence on our results.

### 8.2.2 Mechanical equilibrium

When considering the ideal quasicrystalline structure as a potential (meta-)stable state, the first question one should ask is whether this structure can be mechanically stable, i.e. whether the net forces on all the dipoles balance out or, in other words, whether the structure is a local or global minimum of the systems’ potential energy.\(^4\)

In fact, in simple periodic lattices (such as illustrated in fig. 8.3(a) in the next section), the vanishing net force on the dipoles and hence the equilibration of the system is immediately obvious from the lattices’ symmetry against inversion. More precisely, this is because each particle’s position constitutes a corresponding symmetry center. By contrast, this symmetry is missing in the quasicrystalline structure, which makes the question of mechanical equilibrium non-trivial. Eventually, we may only settle it by numerical simulations which actually fall into the realm of the dynamical investigations in chap. 9.

Note that in an infinite quasicrystal two different positions are never exactly equivalent. So the calculation of the long-range interaction with its surrounding dipoles can, strictly speaking, not be based on a finite piece of the ideal structure. Practically, however, as the dipolar interactions (8.1) or (8.2) decay as $r^{-3}$ with the distance $r$, the potential felt by the dipoles will largely be dominated by their local surroundings (see also next section). In this context, it is interesting to note that in the infinite binary tiling any arbitrarily large piece of it repeats (up to rotations) within a distance of the order of its size (see e.g. [Baa02]).

Numerically, we find the net forces acting on a dipole in the ideal structure to converge to a relative precision of $10^{-6}$ when taking into account neighboring particles up to a distance of $r_{\text{max}} \simeq 25$. Thereby, the resulting force components clearly show that there is no dipole strength ratio $D$ which would make the ideal structure a minimum of the potential

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\(^4\)Strictly speaking, zero net force can also mean a saddle point in the energy landscape; yet, such a state would obviously be unstable to any kind of perturbation.
energy surface. However, the significance of this finding is relativized when the dipoles are allowed to aberrate slightly from their mathematically precise positions as prescribed by the proposed decoration of the binary tiling. In particular, as will be detailed in sec. 9.1, when we let the ideal quasicrystalline structure relax via the method of steepest descent into a local potential minimum, we find that in the range \( 4 \lesssim D \lesssim 6.5 \) the positions of the dipoles are only very slightly shifted. Based on the steepest descent algorithm, though, we cannot state simple systematic rules for the decoration of the tiling such that the dipoles assume an exact mechanical equilibrium. Due to this fact and since the energy per dipole of the relaxed configurations and the ideal quasicrystal are almost the same, we retain the above decoration of the binary tiling as model reference structure.

### 8.3 Ground state calculations

Considering the quasicrystalline structure as a potential equilibrium state, we next investigate how this structure compares energetically to plausible alternative ordered states. To this end, we examine the energy per dipole in a binary dipolar system with the mixing ratio \( x \) and the total number density of dipoles \( \rho \) fixed to the values of the ideal quasicrystalline structure; see sec. 8.2.1.

To determine the energy per dipole \( E_* \) in the ideal quasicrystalline structure, we calculate the average energy of about \( 10^4 \) dipoles within a maximum distance \( r_{\text{max}} = 300 \). Specifically, if one takes into account neighboring dipoles up to a distance \( r_{\text{max}} \), the energy of an individual dipole in the structure scales as \( E \sim \int_0^{r_{\text{max}}} r^{-3} r dr \sim 1/r_{\text{max}} \) for large \( r_{\text{max}} \). In the range \( 50 \leq r_{\text{max}} \leq 300 \) used by us, this scaling relation becomes practically exact and may conveniently be used to extrapolate for \( r_{\text{max}} \to \infty \). In view of the discussion in the previous section, we should mention that the value of \( E_* \) may be up to a few parts per thousand higher than the energy per dipole in the slightly ‘relaxed’ quasicrystal. Note that the conclusions from this section will not alter due to such differences.

The irrational mixing ratio \( x \) requires alternative ordered structures based on regular lattices (with a rational \( N_A/N_B \) to be phase-separated, i.e. to be a combination of two distinct lattices, one with a higher mixing ratio than in the quasicrystal and another one with a lower mixing ratio. In the thermodynamic limit, we can neglect energy contributions from the interfacial boundaries between the two phases and optimize their respective lattice constants (or their ‘volume fractions’) to minimize the total energy per particle \( E_{i} \) of the two-phase state.

Some plausible alternative structures are depicted in fig. 8.3(a): (i) two hexagonal lattices (phase separation of A and B dipoles), (ii) a hexagonal A lattice with B dipoles in the triangle centers and a hexagonal A lattice, (iii) a hexagonal A lattice with B dipoles in every second triangle center and a hexagonal B lattice, (iv) a centered square lattice of A and B dipoles and a hexagonal lattice of B dipoles, and (v) a centered square lattice of A and B dipoles together with the centered A–B hexagonal lattice of (iii). The corresponding values \( E_{i} \) for dipole strength ratios \( 1 \leq D \leq 10 \) are shown in the graph in the figure relative to the value \( E_* \) of the quasicrystalline structure.

The figure shows the preference of hexagonal order and the degeneracy of structures (i)
8.3 Ground state calculations

Figure 8.3: (a) Alternative ordered structures and their energies per particle \( (E_t) \) compared to that of the quasicrystalline structure \( (E_s) \); plotted is the fraction \( E_t/E_s \) as a function of the ratio of dipole moments \( D \). (b) The same for a fictitious modified interaction potential \( \tilde{E}_{int} \sim r^{-\mu} \) with \( \mu = 2.1 \) (instead of 3).

and (ii) for \( D = 1 \), i.e., identical A and B dipoles. Furthermore, hexagonal ordering is preferred for \( D \lesssim 3 \), whereas for larger \( D \), partial tetragonal ordering seems energetically favorable. The quasicrystalline structure is closest to the optimum structure in the range \( 4 \lesssim D \lesssim 6 \), with an optimum around \( D \approx 4.5 \). However, the phase-separated structure (v) clearly has lower energy. Accordingly, the quasicrystalline structure cannot be the ground state of the dipolar mixture.

Still, we find that from a purely energetic point of view the quasicrystalline structure is a surprisingly competitive type of ordering for a binary mixture of dipoles within the appropriate mixing ratio and dipole strength ratio. In view of the fact that the alternative ordered structures (i)-(v) require a phase boundary, which possesses a surface energy, the quasicrystal may become the preferred structure in finite systems. Interestingly, as demonstrated in fig. 8.3(b), the quasicrystal becomes more favorable when the interaction potential \( E_{int} \sim r^{-3} \) is modified to a fictitious \( \tilde{E}_{int} \sim r^{-\mu} \) with \( \mu \) close to 2 (\( \mu > 2 \) for reasons of convergence; cf. also Part I of the present work). Albeit we are not aware of any realization of such a potential in nature, this indicates that other types of scale-free interactions more strongly support the quasicrystalline ordering.
8 Motivation and static energy calculations
9 Dynamic stability analysis

9.1 Stability analysis of reference structures

9.1.1 Steepest descent calculations

In the present chapter, we will study dynamical properties of the quasicrystalline structure. We start by testing its dynamical stability using the method of steepest descent. Thereby, the dipoles are displaced in small steps along their potential gradient with the step length being proportional to the modulus of the gradient vector. In numerics, this algorithm is used to find minima of multi-parametric functions (see e.g. [Pre95]). In physical terms, it describes the overdamped motion of the particles at zero temperature, which may be considered the simplest type of dynamics to be implemented in a system. Accordingly, the algorithm may be viewed both as another energy minimization procedure, which – by contrast to the one in sec. 8.3 – is not confined strictly to a preassigned structure, and as a basic, purely relaxational emulation of the system’s dynamics, which may be used e.g. to assess its mechanical stability. The method has been applied before to Lennard–Jones quasicrystals [Rot90].

In our simulations, the maximum step length (for the dipole experiencing the largest force) is limited to a fixed value of order 0.01–0.001. Typically, after $10^3$–$10^4$ steps, the relaxation is finished when the dipoles start to oscillate about fixed positions, where the amplitudes are of the order of the maximum step length.$^1$ We perform two types of steepest descent calculations for varying dipole strength ratio $D$: In the first one, we start from the ideal quasicrystalline positions of the dipoles, and in the second one, we perturb the ideal positions by Gaussian random noise of different strength.

The results of these simulations are somewhat hard to quantify since their concrete numerical outcome depends on the details of the algorithmic implementation, e.g. the value of the step length. The long-range dipolar potential and the lack of simple translational symmetry (which would let forces balance out trivially) give rise to a large number of dipole configurations being – mostly shallow – local energetic minima. Exactly which of the different minima will be reached in a steepest descent calculation depends on computational details and initial conditions. Nonetheless, robust trends in the relaxation behavior are revealed and we thus obtain a reliable picture of the systems’ dynamical stability.

$^1$We also tested protocols in which the step length was reduced in the course of the simulation in order to allow for a finer relaxation (cf. [Rot90]); however, for the mentioned range of step lengths, we found this to have very little influence on our following results.
Figure 9.1: Order parameter $\phi$ of (8.3) after steepest descent calculations starting from the ideal quasicrystalline structure as a function of the dipole strength ratio $D$. The data are for three different approximants as indicated (cf. text).

### 9.1.2 Results

#### Stability as a function of dipole strength ratio

Fig. 9.1 shows the order parameter $\phi$ as a function of the dipole strength ratio $D$ as obtained after steepest descent calculations starting from the ideal quasicrystalline structure. The data indicate that the ‘relaxed’ configurations stay closest to the ideal structure in a range $4.5 \lesssim D \lesssim 5.5$ with an optimum around $D \approx 4.8$. So the optimum range of $D$ essentially coincides with the range deduced from the comparison with alternative ordered structures in sec. 8.3.

This result is reproducible for different values of the step length and for different approximants (see fig. 9.1), though the values of the relaxed order parameter $\phi$ may differ. Thereby, it should be kept in mind that the magnitude of $\phi$ is very sensitive even to small displacements of the dipoles. For example, we find $\phi \approx 0.8$ if the dipole coordinates are randomly altered with respect to their values in the ideal quasicrystalline structure by a Gaussian noise with standard deviation $\sigma = 0.02$ (cf. fig. 9.2). Thus, in particular in view of the aforementioned potential experimental realizations, it is reasonable to consider the ‘relaxed’ structures to represent the ideal quasicrystalline configuration.

Still, this is not to say that the detailed behavior of $\phi$ was devoid of interesting information on the properties of the dipolar system. In particular, the reason why the medium approximant in fig. 9.1 has a smaller value of the order parameter is likely due to the fact that it corresponds to an approximation of $\tau \approx 1.618$ by the rational value $\tilde{\tau} = 7/4 = 1.75$, which is worse than the approximations $\tilde{\tau} = 5/3 = 1.6$ and $\tilde{\tau} = 8/5 = 1.6$ by the small and large approximants, respectively [Ent88]. The medium approximant therefore has the lowest quality with respect to representing the ideal quasicrystalline configuration. The
9.1 Stability analysis of reference structures

Figure 9.2: Average order parameter $\phi$ after steepest descent calculations starting from a perturbed structure for different values of $D$. $\phi$ is plotted as a function of the standard deviation $\sigma$ of the initial Gaussian displacements. As indicated, two curves (full circles) correspond to a medium-sized (1700 dipoles) and a large (2330 dipoles) approximant, as opposed to 890 dipoles (cf. fig. 9.1). For comparison, the order parameter $\phi$ due to pure Gaussian noise is shown, i.e. before the steepest descent relaxation starts.

Results for the small and large approximant suggest that finite size effects are not relevant here.

Recovery from perturbations

The above findings are also supported by the second type of steepest descent calculations, in which we test for metastability of the structure and ask if the quasicrystalline order recovers from small perturbations. Therefore, we apply Gaussian noise with zero mean and standard deviation $\sigma$ to each particle coordinate before letting the dipoles relax as before. The average order parameter $\phi$ of the final configurations is plotted in fig. 9.2 as a function of $\sigma$ for a few values of $D$ from the optimum range.

Up to an average initial displacement $\sigma \lesssim 0.15$, the system relaxes back to the quasicrystalline structure, whereas for larger $\sigma$, numerous defects remain as reflected by the decreasing average order parameter $\phi$. This behavior is reminiscent of a modified Lindemann-like criterion, which predicts the melting of a 2D dipole crystal at an average relative displacement of 0.12 in units of the lattice constant [Bed85]. We also note that due to the initial Gaussian noise the order parameter decreases below 0.1 at $\sigma = 0.15$, from which it recovers its optimum value around 0.9. Hence, the quasicrystalline state proves relatively robust against random perturbations.

Note that the order parameter $\phi$ in fig. 9.2 shows an equivalent behavior with respect to the different approximants as already discussed in connection with fig. 9.1. So, to conclude,
fig. 9.1 and fig. 9.2 suggest that the (relative) stability of the quasicrystalline binary tiling within the identified range of favorable dipole strength ratios $D$ is an intrinsic property of the ordered structure rather than a finite size effect.

9.2 Behavior at finite temperatures

9.2.1 Monte Carlo simulations

To assess the behavior of the binary system at finite temperatures we choose Monte Carlo (MC) simulations [All90]. While computationally less costly than e.g. Langevin dynamics or even more detailed schemes, the dynamical MC simulations allow us to explore thermo-dynamical equilibrium states and, with a reasonable choice of jump trials, should also yield a realistic scenario of the systems’ evolution.

As a jump trial in our simulations, a particle is chosen at random and imposed a Gaussian distributed displacement. The trial is accepted according to the usual Metropolis rule (cf. sec. 6.2). The standard deviation of the Gaussian in the range 0.001–0.1 is adjusted dynamically to ensure an efficient acceptance rate of trials in the range 10–60%. One MC step (MCS) corresponds to one attempted move per particle.

For sufficiently low temperatures, the outcome of these standard MC simulations goes well together with the results from the steepest descent simulations. For example, starting from the ideal or slightly perturbed quasicrystalline structure at $T = 0.005$, we find the order parameter first to decay and then to fluctuate around $\phi \simeq 0.75$, which confirms that (nearly) quasicrystalline order represents a local minimum in the free energy.

However, especially at slightly higher $T$, the degree and speed of the order parameter relaxation vary strongly from run to run in the MC simulations. In fig. 9.3, example trajectories of $\phi$ are shown for $T = 0.01$ and 0.04. The quasicrystalline structure is relatively stable at $T = 0.01$, but the attained value of $\phi$ depends erratically on the initial conditions and the realization of the stochastic MC simulation. As an example, in fig. 9.3 the order parameter emerging from an initially perturbed structure (run 2) unexpectedly exceeds the order parameter emerging from the ideal quasicrystalline structure (run 1). Moreover, at $T = 0.04$ a slow and unpredictable decay of $\phi$ becomes observable on the time scale reached in the simulation. Even after excessively long runs of more than $10^6$ MCS, it remains unclear whether the system has reached an equilibrated state.

9.2.2 Parallel tempering

To accelerate thermalization in the simulations, there are different possibilities. One of them is to introduce ‘artificial’ multi particle flips as additional MC moves, as was previously done for the Lennard–Jones system in [Wid87]. In an extra series of our simulations, we tested an elementary version of such flips, whereby every 100 MCS the positions of an A and a B dipole are interchanged and the new configuration is evolved for several MCS before the whole move is either accepted or rejected. In general, we find the relaxation of $\phi$ to be faster, but the overall behavior remains unaltered.
For the main part of our simulations, we use the standard local moves of single particles, which might be closer to the dynamics of the experimental systems, but we apply a more sophisticated thermalization scheme known as parallel tempering, which has been applied before e.g. to obtain equilibrium thermodynamical properties of spin glass models [Huk96, New99]. Basically, the idea of the method is to circumvent trapping of a system's dynamics in local energetic minima at low temperatures by occasionally interchanging the configuration with the one of the same system simulated in parallel at higher temperatures. Here, we consider 26 copies of our system at temperatures $2.5 \times 10^{-4} = T_1 > T_2 > \ldots > T_{25} = 0.071$. Every 2000 MCS, the particle configurations at adjacent temperatures $T_i, T_{i+1}$ are interchanged with a Metropolis type rate,

$$w_{i,i+1} = \begin{cases} \exp \left( \frac{1}{T_i} \left( \frac{1}{T_{i+1}} \right) (E_{i+1} - E_i) \right) & \text{if } E_{i+1} > E_i \\ 1 & \text{else} \end{cases}$$

where $E_i$ and $E_{i+1}$ are the energies of the configurations. It can be shown that this scheme allows different configurations to occur with their correct Boltzmann weight at any of the temperatures $T_i$.

In our implementation of the algorithm, a control program on a single PC keeps track of the configurations simulated at the various temperatures $T_i$. Once the assignment of a certain configuration to a $T_i$ has been made for the next 2000 MCS, it is passed as independent computing job to our queuing system. So the simulations can be carried out on a variable number of available CPUs which may also differ in speed. For the approximant
Figure 9.4: Order parameters $\phi$ and $\tilde{\phi}_n$ for several $n$ as a function of temperature $T$ as obtained from the parallel tempering MC simulations. The dipole strength ratio is $D = 4.75$. Note the jump in the ordinate.

with 890 dipoles (cf. sec. 8.2.1), which was used for the simulations in the following, one MCS for a single configuration takes about 0.5s on a contemporary Intel Pentium IV 2.8 GHz CPU.

We find the parallel tempering algorithm to be highly effective in thermalizing our ensemble of 26 systems. This relatively large number allows us to keep the spacing between the $T_i$ small and thus to change configurations frequently while still covering a large range of temperatures from possible ordering to apparently fluid–like behavior. After typically 300 rounds (i.e. $300 \times 2000$ MCS), we see no qualitative differences any more between the extremes of an ensemble started from the ideal quasicrystalline structure and one started from random initial positions.

9.2.3 Results: local ordering

In fig. 9.4, the mean order parameters $\phi$ and $\tilde{\phi}_n$ for several $n$ are displayed as a function of temperature $T$. The values are obtained from particle configurations at the respective $T_i$, each taken at the end of the 2000 MCS cycles of the parallel tempering scheme. The simulations are carried out at a dipole strength ratio $D = 4.75$ and started from random initial positions. The behavior is very similar for $D = 4.5$ and 5. Apart from larger initial fluctuations of the order parameters $\phi$ and $\tilde{\phi}_{10}$, the same holds for simulations started from ordered initial positions.

Additionally, in fig. 9.5, part of an example configuration from the lower temperature range in the parallel-tempering ensemble is shown. In this representative example, no long-range quasicrystalline ordering is discernable. It thus complies to the results in fig. 9.4, where the order parameter $\phi$ may be seen to stay very small throughout the whole temperature range and there is no signature of a phase transition.
Figure 9.5: Configuration from the parallel tempering MC simulations for $D = 4.75$. The temperature is $T = 0.00134$, the order parameter $\phi \simeq 0.07$ and energy per particle $E \simeq 8.087$. The part of the system shown is at the same scale as in fig. 8.2. The lines highlight local quasicrystalline ordering and are drawn whenever the bond angles between nearest neighbors of A (B) particles are even (odd) multiples of $2\pi/10$ (within an error of 10%).

As another observation, we find that the mean potential energy per particle in the final configurations approximately fulfills $E(T) \simeq 8.083 + T$ at low $T$, where the simple dependence on $T$ can be understood from a harmonic approximation around the configurations with lowest energy. Thus, at low temperatures, typical configurational energies are below both the energy of the ideal quasicrystalline structure ($E_\ast \simeq 8.103$ for $D = 4.75$) and the one reached in the steepest descent calculations, cf. sec. 9.1. On the other hand, $E$ is still above the energy of the optimum phase separated structure ($E_{(w)} \simeq 8.078$) found in the ground state energy calculations in sec. 8.3. This is not surprising, since the necessity of a phase boundary might prevent the dipolar mixture from reaching its possible ground state as deduced for infinite system size.

Based on these results, we can exclude the spontaneous occurrence of long–range quasicrystalline order in a thermodynamically stable phase. Moreover, irrespective of the (nearly) quasicrystalline structure being stable against small mechanical perturbations, we think that it may not represent a thermodynamically metastable state. Such a state, as discussed in sec. 8.1, would become separated from other states by an infinite free energy barrier in the limit of infinite system size.

Refined structural analysis

While the overall structure of the obtained configurations is amorphous, we find – indeed in accordance with experimental observations [Wen00, Kön03] – that there occur small domains with particles arranged as in the quasicrystal. In the exemplary structure in fig. 9.5, this is visualized by lines being drawn between neighboring particles as far as their constellation matches the one of the ideal quasicrystalline structure in fig. 8.2. As can be seen from the value of $\tilde{\phi}_3$ and $\tilde{\phi}_{10}$ compared to $\tilde{\phi}_n$ with $n = 4, 8$ and 6, 12 in fig. 9.4, there is a tendency towards a preferred local 5– or 10–fold symmetry at lower temperatures.
Figure 9.6: Bond–order correlation function $g_n(r)$ for $n = 5$ (triangles) and $n = 10$ (squares). The average is over 100 configurations at a temperature $T = 0.00134$ as in fig. 9.5. The open and full symbols distinguish the initial configurations in the MC simulations; ideal quasicrystal (open symbols) and random positions of the dipoles (full symbols).

To support these statements further, we introduce the bond–order correlation function

$$g_n(|\mathbf{r} - \mathbf{r}'|) = |\langle e^{i\alpha(\mathbf{r}) - \alpha(\mathbf{r}')} \rangle|,$$  

(9.2)

where the angle $\alpha(\mathbf{r})$ gives the orientation of a bond between two dipoles at location $\mathbf{r}$ and $\langle \ldots \rangle$ means both an average over all pairs of bonds at a distance $r = |\mathbf{r} - \mathbf{r}'|$ and over different configurations taken from the MC simulations. As already noted in sec. 8.2.1, the function $g_n(r)$ complements the order parameters $\phi$ and $\phi_n$ in that it gives an overview of the bond–orientational ordering over a variable distance $r$.

In fig. 9.6, we plot $g_n(r)$ with $n = 5$ and $n = 10$ for both ideal quasicrystalline and random positions of the dipoles as initial configurations in the parallel tempering simulations. Evidently, the equilibrium bond–order correlation function does not depend on the initial condition – apart from deviations for small distances $r$ due to statistical errors. Moreover, the graphs clearly corroborate that the quasicrystalline order is restricted to small domains as stated above.
10 Summary of Part III

Synopsis

The subject of Part III of the present work is an investigation of the possible occurrence and characteristics of quasicrystalline order in two-dimensional mixtures of point dipoles with two sorts of dipole moments.

At the outset of chap. 8, we started by a brief introduction to quasicrystals as a novel type of solid materials, whose structure is characterized by long-range, yet non-crystallographic (i.e. non-translation invariant) positional order. We described the notions of quasiperiodicity and periodic approximants of these structures as well as some of the unusual physical properties of quasicrystalline materials. As for the theoretical understanding of their formation, we pointed out the recovery of quasicrystalline order in simulations of two-dimensional binary mixtures, whereby, however, length parameters of the assumed short-range interaction potentials among the particles were specifically chosen to favor this type of order.

Motivated by recent experimental findings, we hence turned to the study of the dipolar systems as described above, where quasicrystalline ordering would be particularly intriguing because the scale-free dipolar interaction potential does not possess any tunable intrinsic length scale. Within a preview of our approach, we then propounded possible characteristics of the envisaged quasicrystalline state—such as mechanical and thermodynamic stability—as well as methods to assess them.

In sec. 8.2, we presented in detail our model system. As quasicrystalline reference structure, we chose the prominent rhombic binary tiling decorated by two types of particles, which was also used in the aforementioned earlier simulations. With the mixing ratio of the two particle species thus fixed, the ratio $D$ of their dipole strengths and temperature $T$ are the only free parameters in the model. Further, we introduced different bond–orientational order parameters to quantify the degree of quasicrystalline ordering. By contrast to conventional crystals, the basic question of mechanical equilibrium (i.e. whether forces balance out) turns out to be non-trivial due to the lack of inversion symmetry. Numerically, though, we found that for $4 \lesssim D \lesssim 6.5$ the particles equilibrate mechanically at positions shifted only very slightly from the proposed decoration (cf. below).

On this basis, we next compared the energy per dipole in the quasicrystalline structure to that in a number of plausible alternative ordered states, which have to consist of two phases to account for the irrational mixing ratio. Although the quasicrystalline structure turned out not to be the ground state of the infinite dipolar mixture (i.e. neglecting phase boundaries in the alternative states), we found it notably close to the optimum for the same range of $D$–values as above. Hence little additional stimuli might suffice to make...
quasicrystalline order energetically favorable – which, interestingly, is also furthered by tentatively modifying the exponent in the interaction potential (see further below).

The subsequent chap. 9 was concerned with dynamical properties of the quasicrystalline structure as assessed by different simulation methods. As a simple, purely relaxational dynamical scheme, steepest descent calculations were employed to identify the aforementioned range of suitable dipole strength ratios \( D \), for which the reference structure – subject to minor displacements of the dipoles – is in mechanical equilibrium. The quasicrystalline structure then also proved mechanically stable in that it recovered from moderately strong random noise, which was applied to the dipoles’ positions prior to the steepest descent calculations. Thereby, we found no indication of finite size effects, yet an apparent correlation to the quality of the used periodic approximant of the quasicrystalline structure. Also, the tolerance to perturbations was found in some similarity to predictions from a modified Lindemann criterion for the melting of crystals.

In order to obtain a more comprehensive picture of the system’s dynamics and to assess thermodynamical equilibrium states, we finally used Monte Carlo simulations. In their conventional form, these simulations corroborated the reference structure being metastable, but turned out very ineffective for the system’s thermalization, which, moreover, was not improved substantially by allowing for additional multi particle flips. As a more elaborate thermalization method, we therefore implemented a parallel tempering scheme. From an extensive series of simulations, the thus equilibrated systems were found amorphous in their overall structure. In accordance with experimental observations, however, we found distinct local quasicrystalline ordering, which was analyzed in particular by means of the specifically introduced bond–order correlation functions.

**Further discussion and outlook**

Our results from the parallel tempering method provide ample evidence that the quasicrystalline binary tiling with two sorts of dipoles does not correspond to a thermodynamical equilibrium state. Nevertheless, we can identify a range for the dipole strength ratio \( D \) where a long–range quasicrystalline structure corresponds to a local minimum in the potential energy landscape of the system. According to the steepest descent calculations, this local minimum has an attraction basin covering Gaussian fluctuations of the particle positions up to 15\% of the distance of two neighboring A– and B–dipoles in the ideal reference structure. Our simulations, however, do not indicate that the barriers separating the local minimum from other minima increase with the system size. Therefore we do not expect that such a frozen state with quasicrystalline order exists, although we cannot strictly exclude this possibility.\(^1\)

In any case, at low temperatures the times for the system to escape the local minimum by surmounting free energy barriers can become rather long. This is clearly seen in the kinetics modeled by conventional Monte Carlo simulations. Hence structures with long–range

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\(^1\)Our conclusion is based on the variation of the system size in steepest descent and parallel tempering MC simulations. Strictly speaking, it would require elaborate standard MC simulations (for sufficient statistics) in a large enough variety of system sizes, which is beyond the scope of the present work.
quasicrystalline order might be kinetically stable over sufficiently long time to make them an interesting subject for further study and eventual applications. What ‘sufficiently long’ in this context means could be tested in experiments. Today’s optical tweezer techniques allow colloidal particles to be placed at defined positions. Accordingly, one could prepare a quasicrystalline pattern and monitor its stability. Moreover, special boundary conditions and external stimuli may support the formation of quasicrystalline structures. We also found that a modified scale–free interaction potential \( \propto r^{-2+\epsilon} \) leads, for \( \epsilon > 0 \) becoming small, to ground state energies of the quasicrystal that within numerical error bars cannot be distinguished from the most favorable phase-separated lattice structures investigated in sec. 8.3.

In agreement with experiments, we find that even in the disordered ground state of the binary dipole system, local bond–orientational order with quasicrystalline 5– and 10–fold symmetry is preferred. We made quantitative predictions for the temperature behavior of several local bond–orientational order parameters, which can be tested in experiments by using, for example, two-dimensional binary mixtures of superparamagnetic colloidal particles [Zah99, Kön03, Kön04].
10 Summary of Part III
11 Zusammenfassung (Short summary in German)


Hier wird untersucht, inwieweit in einem binären dipolaren System stabile oder zumindest metastabile quasikristalline Ordnung auftreten kann. Als Referenzstruktur wird eine binäre rhombische Bedeckung der Ebene ähnlich dem bekannten Penrose-Muster verwendet. Aus statistischen Energieberechnungen folgt, daß diese im Vergleich mit alternativen geordneten Strukturen insbesondere in endlichen Systemen energetisch nahezu gleichwertig sein kann. 'Steepest-descent'-Simulationen zeigen eine überraschende Stabilität gegenüber aufgeprägter zufälliger Unordnung. Die Thermalisierung der dipolaren Systeme in Monte Carlo-Simulationen erweist sich als sehr schwierig; die 'parallel tempering'-Methode liefert aber dennoch verläßliche thermische Mittelwerte. Danach ist die quasikristalline Ordnung in einem freien System keine thermodynamisch stabile Phase; abhängig von den Parametern tritt jedoch spontan lokale quasikristalline Ordnung auf.
Appendix
A Appendices to Part II

A.1 Details of the effective dynamics simulations

In the following, we note some details of the computer simulations of the effective dynamics, which are technical in nature, yet greatly enhance the practicability of this approach.

For our simulations, we consider system sizes in the range of $L/L_0 = 2^{15}$ to $2^{20}$, i.e. 15 to 20 steps in the iterative construction of the hierarchical lattice. While the effective dynamics, at the price of limited time and length resolution, takes away most of the computational effort necessary to simulate the dynamics of such a system (as compared e.g. to MC simulations, see sec. 6.2), the sheer amount of $\sim 10^9$ to $10^{12}$ Gaussian bonds and a similar number of spins to be stored and linked constitutes a certain challenge for common contemporary computers.

Therefore, it should be realized that, for the effective dynamics, parts of the lattice may ‘exist’ merely in the form of an effective (renormalized) coupling. As an example, the cluster marked gray in fig. 4.3 could be replaced by an effective bond (of level 2) from the outset. Obviously, this possibility can be employed to regulate – within wide limits – the number $N_\alpha$ of spins of a certain level $\alpha$ which one aims to consider explicitly in the simulation.

Note that the method is particularly unproblematic for the study of local quantities, i.e. those pertaining to single spins, such as the spin autocorrelation or magnetization. (Non-local quantities, e.g. spatial correlations, besides would hinge on the definition of distance in the hierarchical topology.)

In our simulations, we have randomized the described replacement of clusters by effective bonds. Thereby, for simplicity, we have chosen $N_\alpha = const.$ in most cases, although this is suboptimal for the statistical accuracy of ultimately per site or per bond weighted quantities. The data displayed in the main text are typically based on 10 trajectories (i.e. realizations of the dynamics) for $N_\alpha = 200$ spins in each of $\gtrsim 5000$ simulated systems (i.e. realizations of the random bonds). Thus, as far as not noted otherwise, the statistical uncertainty is commonly below the symbol sizes in the plots.

To efficiently obtain those renormalized bonds which are present from the outset, we use the ‘pool method’ [Ban87]: Instead of calculating the bond, say of level $n$, from $8^n$ Gaussian random bonds, we draw it from a ‘pool’ (or reservoir) of $N_{pool} \gg 1$ such bonds. The reservoir itself has been obtained beforehand by choosing randomly each $p = 8$ bonds from the corresponding pool of level $n - 1$ bonds, which in turn is based on the $(n - 2)$-pool etc. For sufficient statistics of the couplings, we apply the method with $N_{pool} = 2 \cdot 10^5$ only for bonds of level $n \geq 4$. Also, the abovementioned randomization of the topology (as
Figure A.1: Schematically: Two replica A and B of a cluster with twisted boundary spins $S_{i,j}$.

... should assist that purpose.

In the renormalization in presence of a magnetic field $H$, the pool method is somewhat susceptible to a finite size effect manifesting in an increasing mean $\mu_H$ of the fields conveyed by the bonds, see sec. 6.1.1. We have taken additional measures, e.g. the randomization of the fields’ signs, and performed some tests to ensure that the results of the effective dynamics simulations are not affected.

**A.2 Cluster overlap under twisted boundaries**

Here, we shall prove that the overlap of a cluster of level $\alpha$ under a twisted-boundary condition, i.e. before and after one of its boundary spins $S_i^{(\alpha)}$, $S_j^{(\beta)}$ ($\beta > \alpha$) is flipped, vanishes when averaged over thermal fluctuations and the bond disorder or, alternatively, via self-averaging in a large system.

For this purpose, let us consider two replica of the cluster as depicted in fig. A.1: replica A with boundary spins $(S_i, S_j)$ and B with $(-S_i, S_j)$, where the indication of the spins’ levels ($\alpha, \beta$) is left out for brevity. The average overlap, i.e. the contribution to the spin autocorrelation (4.10) of the spins $S_k \in \{S^{(\alpha-1,\alpha-2,\ldots,0)}\}$ within the cluster, is

\[ C(A, B) = C((S_i, S_j), (-S_i, S_j)) = \sum_k w_{(k)} \langle S_k(A) S_k(B) \rangle, \]

whereby details such as the weighting of different spins don’t matter in the present context.

Now, we first note that

\[ C((S_i, S_j), (-S_i, S_j)) = C((S_i, S_j), (S_i, -S_j)), \]

since, after the disorder average, $S_i$ and $S_j$ play a symmetrical role for the cluster – or, put differently, for every configuration of bonds in the cluster there is another, ‘mirrored’ one. Second, a global flip of all spins (including the boundary spins) of one replica, say B, simply changes the sign of the overlap, $C(A, B) = -C(A, -B)$, or

\[ C((S_i, S_j), (-S_i, S_j)) = -C((S_i, S_j), (S_i, -S_j)). \]

This is because the statistical weight of a spin configuration $S_k$ is unchanged under a global spin flip due to the corresponding invariance of the Hamiltonian (in absence of a magnetic field). Combining (A.2) and (A.3), we conclude $C((S_i, S_j), (-S_i, S_j)) = 0$.

This result for the overlap of the whole cluster carries over to that of individual spins $S_k$, i.e. to single summands in (A.1), because, as already noted in sec. 4.2.2, these are...
equal after the averaging. Alternatively, one may argue as above: For every realization of bonds (and of the dynamics), in which $S_i$ follows the flip of boundary spin $S_j$, there is a ‘mirrored’ realization, in which it keeps unchanged as $S_j$. Besides, in the RSRG both of these bond configurations yield an identical effective coupling $J_{ij}^{\alpha}$ between $S_i$ and $S_j$, cf. the corresponding symmetry in (4.4) ff.

Finally, let us note that the vanishing of the overlap under twisted boundaries is a quite general result in no way restricted to a spin glass on the hierarchical lattice. The only requirements are a ‘symmetric’ distribution of the overlapping components (spins) with respect to the boundary conditions and invariance of the Hamiltonian under a global spin flip. Thus, our result also holds e.g. on a cubic lattice and/or for a ferromagnet.

A.3 Refined scaling of the normalized spin autocorrelation

In the following, we shall particularize the approximation which, as noted in sec. 5.2, is contained in the proposed short-time scaling form (5.21) for the normalized correlation $\hat{C}_\alpha(t_m)$ obtained in a temperature shift or bond perturbation protocol. As in the main text, the discussion is only for the case of per bond weighting of spins for simplicity.

In fact, straightforward insertion of the approximate rates $\bar{r}_{\sigma}(\delta) \sim \hat{\rho}(0)[L_{\sigma}/L^*(\delta)]^\zeta$ in (5.2) yields

$$\hat{C}_\alpha(t_m) = 1 + c^\phi - c^\phi \hat{\rho}(0) \frac{L(t_m)}{L^*(\delta)}^\zeta \tag{A.4}$$

with constants (for a specific protocol) $c^\phi$, $c^\phi$. Hence the approximation in (5.21) lies in the neglect of the contribution $c^\phi$, which reads explicitly

$$c^\phi \sim \frac{1}{2^{\phi} - 1} \hat{\rho}(0) \delta. \tag{A.5}$$

That we merely have this asymptotic relation for $c^\phi$, i.e. that it contains (at least) an unknown constant, clearly is because the rates $\bar{r}_{\sigma}(\delta)$ stem from a scaling argument. Numerically, we find $c^\phi \simeq 0.3 \delta$ to do a very good job in bringing the bond perturbation ($T = 0$) data in fig. 5.7(a) to a straight line for $(L(t_m)/L^*(\delta))^\zeta \lesssim 3$. Besides, the low numerical value for $c^\phi$ explains why the scaling without it also works moderately well.

For $T > 0$, the situation is more complicated, since there the isothermal correlation $C_{\alpha_l}(t_m)$ enters via the scaling (5.15). Similarly as in (A.4), we find for the onset of its decay

$$C_{\alpha_l}(t_m) = 1 + c^\phi T - c^\phi \hat{\rho}(0) T \left( \frac{L(t_m)}{L_0} \right)^{-\theta} \tag{A.6}$$

with an expected $c^\phi = (2^{-\theta} - 1)^{-1} \cdot \hat{\rho}(0)/(Jc)$ and $c^\phi = 2^{-\theta} c^\phi / \hat{\rho}(0)$, in which $c \simeq 3.22$ from sec. 5.1. Indeed, this behavior may be retrieved in the data for $C_{\alpha_l}(t_m)$ in fig. 5.4, whereby the prediction for $c^\phi$ proves correct to a few percent, while that for $c^\phi$ is $\sim 20\%$ off the numerically obtained value. Note that, by contrast to $c^\phi$, $c^\phi$, the ‘isothermal’ factors $c^\phi$, $c^\phi$ are negative, which formally comes from the exchange of the exponent $\zeta$ by $-\theta$ in expressions like the fraction in (A.5).
A Appendices to Part II

Now, if we assume that (A.4) (understood for $T = 0$) and (A.6) (for $T = T_2$) are independent decay mechanisms for $C_\infty(t_m)$ (from a shift $(T_1, T_2)$) and put everything together in the definition (5.15) of the normalized correlation function $\hat{C}_\infty(t_m)$, we obtain approximately

$$\hat{C}_\infty(t_m) \simeq 1 + \left[ \frac{c^\Delta T_c}{2} + c^\delta \right] \delta - c^\delta \tilde{\rho}(0) \left( \frac{L(t_m)}{L(\delta)} \right)^c . \quad (A.7)$$

Therein, the term in square brackets, which is the analogue of $c^\Delta$ in (A.4), is (slightly) reduced by the additional contribution $\propto c^\Lambda$, which might be a hint as to why the ‘approximate’ scaling (5.21) works better here than for the $T = 0$-data.

However, we emphasize that the preceding arguments are to some degree speculative and obviously suffer from a number of approximations. More extensive numerical checks, on the other hand, are complicated by the limited time resolution of the effective dynamics, which does not allow for an arbitrary ‘magnification’ of any considered scaling regime.

After all, small times $t_m$ are clearly not the realm of the effective dynamics, so that the identification of an ‘effective’ scaling (5.21) of $\hat{C}_\infty(t_m)$ in some range of intermediate $t_m$ might well summarize the gain in general insight to be obtained from this particular aspect.

## A.4 Explicit formulas for the MK renormalization in a field

Here we give explicitly some expressions for the RSRG on the hierarchical lattice in presence of an external magnetic field which, for conciseness, have been left out in the main text.

As noted in sec. 6.1.1, effective (renormalized) bonds and fields for dimension $d \geq 1$, i.e. $p' = 2^{d-1} \geq 1$ parallel paths between two higher level spins $S_{i,j}$, are obtained by adding the contributions of each one-dimensional path as given by (6.4) and (6.5). With the full notation as in (4.6), we have in the $n$-th step of the renormalization

$$J_{ij}^{(n)} = \frac{1}{4} \sum_{k=1}^{p'} \log \left[ \frac{\cosh \left( J_{ik}^{(m)} + J_{kj}^{(m)} + H_k^{(m)} \right) \cosh \left( J_{ik}^{(m)} + J_{kj}^{(m)} - H_k^{(m)} \right) \cosh \left( J_{ik}^{(m)} - J_{kj}^{(m)} + H_k^{(m)} \right) \cosh \left( J_{ik}^{(m)} - J_{kj}^{(m)} - H_k^{(m)} \right)}{\cosh \left( J_{ik}^{(m)} - J_{kj}^{(m)} + H_k^{(m)} \right) \cosh \left( J_{ik}^{(m)} - J_{kj}^{(m)} - H_k^{(m)} \right)} \right] , \quad (A.8)$$

$$H_i^{(n)} = H_i^{(m)} + \frac{1}{2} \sum_{k=1}^{p'} \log \left[ \frac{\cosh \left( J_{ik}^{(m)} + J_{kj}^{(m)} + H_k^{(m)} \right) \cosh \left( J_{ik}^{(m)} + J_{kj}^{(m)} - H_k^{(m)} \right) \cosh \left( J_{ik}^{(m)} - J_{kj}^{(m)} + H_k^{(m)} \right) \cosh \left( J_{ik}^{(m)} - J_{kj}^{(m)} - H_k^{(m)} \right)}{\cosh \left( J_{ik}^{(m)} + J_{kj}^{(m)} - H_k^{(m)} \right) \cosh \left( J_{ik}^{(m)} + J_{kj}^{(m)} - H_k^{(m)} \right)} \right] , \quad (A.9)$$

where $m \equiv n - 1$ and the expression for $H_j^{(n)}$ is obtained by interchanging indices $i$ and $j$ in the last line.

In the numerical treatment of the RSRG, the approximations

$$\log [\cosh(x)] = \begin{cases} \frac{x^2}{2} - \frac{x^4}{12} + O(x^6) \quad & \text{for } x \ll 1, \\ x - \log(2) + e^{-2x} + O(e^{-4x}) \quad & \text{for } x \gg 1 \end{cases} \quad (A.10)$$

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are useful to ensure numerical stability. For $T \to 0$, the second case applies, which yields the low-temperature limit of (6.4) and (6.5),

$$J' = \frac{1}{2} \left( \sigma^+ \max \{|J_1 + J_2|, |H_0|\} - \sigma^- \max \{|J_1 - J_2|, |H_0|\} \right),$$

$$H'_1 = H_1 + \frac{1}{2} \left( \sigma^- \min \{|J_1 + J_2|, |H_0|\} + \sigma^- \min \{|J_1 - J_2|, |H_0|\} \right),$$

where $\sigma^+ \equiv \text{sgn}((J_1 \pm J_2)H_0)$ and $H'_2$ may be obtained by exchanging indices as explained before. The generalization to $d > 1$, i.e. summation over parallel paths as in (A.8), (A.9), is obvious.

### A.5 Fluctuation dissipation theorem

In the following, we demonstrate that, as claimed in sec. 6.1.2, the fluctuation dissipation theorem (FDT) for thermal equilibrium takes the simple form (6.9) in our system on the hierarchical lattice, provided the same weighting factors $w_\alpha$ are employed in the definition of the spin autocorrelation function (4.10) and the magnetization (6.7).

In a general form, which is given e.g. in [Kub95], the FDT may be written

$$\langle b(t) \rangle - \langle b \rangle_0 = -\frac{1}{T} \int_{-\infty}^{t} ds \, f(s) \left\langle \left( \frac{d}{dt} b(t-s) \right) a(0) \right\rangle_0,$$  

where $b$ is the quantity to be observed and $a$ that by which the external field $f(t)$ couples to the Hamiltonian, i.e. $\mathcal{H}$ contains a term $-a(t)f(t)$. Angle brackets $\langle .. \rangle$ denote thermal averaging in the presence of $f(t)$, while those with index zero explicitly stand for thermal averaging without field. In the notation of (6.10), the term in brackets in the above integral is the response function $R(t-s)$, which here depends only on the time difference $t-s$ (cf. below).

For the zfc magnetization in our system, $f(t)$ is the external magnetic field $H(t) = H\Theta(t-t')$ and $b(t)$ is the magnetization $\tilde{M}(t)$ of (6.7), whose average without field vanishes. Since the field is applied uniformly to all sites (cf. sec. 6.1.1), the factor $a(t)$ in the Hamiltonian is the sum of all spins, $\tilde{M}(t) \equiv \sum_i S_i(t)$.

Thus (A.13) gives

$$\chi_{\text{zfc}}(t) = \frac{M_{\text{zfc}}(t)}{H} = \frac{1}{T} \left( \left\langle M(0)\tilde{M}(0) \right\rangle_0 - \left\langle M(t-t')\tilde{M}(0) \right\rangle_0 \right),$$

where we have introduced $\chi_{\text{zfc}}$ as in the main text. Therein, by the definitions of $M$ and $\tilde{M}$, each of the averages $\langle .. \rangle_0$ contains a double sum over all spins. Since there is no net magnetization without field, i.e. there is an equal number of positive and negative spins $S_i$, only the diagonal terms of the double sum contribute to the first average (with equal time arguments). More generally, due to the symmetric distribution of bonds (cf. sec. 3.2.1), dynamical correlations of different spins (at distinct times) cannot exhibit a
preferred direction either; so the second average also reduces to its diagonal terms. Hence we get
\[ T_{\chi^{(a)}(t)} = \left( \sum_{\alpha} w_{\alpha} \sum_{i_\alpha} \left(S_{i_\alpha}^{(\alpha)}(0)\right)^2 \right)_0 - \left( \sum_{\alpha} w_{\alpha} \sum_{i_\alpha} S_{i_\alpha}^{(\alpha)}(t - t') S_{i_\alpha}^{(\alpha)}(0) \right)_0. \] (A.15)

Note that the (normalized) weighting factors \( w_{\alpha} \) appear only once in each average, of which the first equals unity, while the second gives the spin autocorrelation \( C(t - t') \).

Then there are merely two formal differences left to (6.9): First, the quantities, which are time translation invariant in the equilibrium setting considered here, are written as two-time functions (depending explicitly on the waiting time \( t' \)); second, in the corresponding two-time notation, the variable \( t \) refers to the time passed since the end of the waiting time \( t' \), which in turn is taken as reference time for the autocorrelation function; cf. (4.10).

### A.6 Dynamic MC algorithms

Here we briefly outline our experience with different implementations of the MC simulations for our spin glass model, the results of which are discussed in sec. 6.2. In fact, algorithms for such kinetic MC simulations exist in several variants, see e.g. [Bin79].

In the well-known ‘discrete time’ (dt) MC algorithm, as described deliberately formally, each elementary step consists of a trial to perform one of the currently possible events in the system – such as a flip of a randomly or sequentially chosen spin. The trial is accepted with a probability corresponding to the assumed flip rate \( r \), which may be e.g. of Metropolis type as given in the main text. The time unit then is usually taken to be one Monte Carlo step (MCS), i.e. (an average of) one trial per independently realizable event, which in our case is identical to the time \( \tau_0 \).

For the connectivity weighted dynamics as in [Ric00], in particular, a spin of level \( \alpha \) is updated \( 4^\alpha \) times in each MCS, which is equivalent to a modified attempt frequency \( 1/\tau_0^{(\alpha)} = 4^\alpha/\tau_0 \) for the rates as mentioned in the main text.

An alternative to the dt MC algorithm is given by ‘continuous time’ (ct) methods which, instead of relying on accepted or rejected trials, employ a suitably chosen stochastic time step \( \Delta t \) to the next event to be performed. Practically, \( \Delta t \) may be drawn from the exponential distribution \( p(\Delta t) = R \exp[-R\Delta t] \), where \( R = \sum_i r_i \) is the sum over the rates of all possible events \( i \), one of which has to be chosen subsequently according to its probability \( p(r_i) = r_i/R \).

In the recently proposed ‘waiting time method’ [Dal01], as another ct procedure, each possible next event \( i \) is assigned an estimated time \( t_i = t + \Delta t_i \), where \( t \) is the current time in the simulation and the individual waiting time \( \Delta t_i \) is drawn from \( p(t_i) = r_i \exp[-r_i t_i] \). Then the event with the smallest \( t_i \) is performed and the simulated time \( t \) put forward to \( t_i \) (corresponding to a time step \( \Delta t = \Delta t_i \)).

---

1 This holds if one chooses as acceptance probability for the trial \( r \cdot \tau_0 = \min\{1, \exp[-\Delta E/T]\} \), i.e. the standard expression of the main text without the prefactor \( \tau_0^{-1} \).

2 We remark that, while the number of updates is claimed to be weighted by the connectivity \( z = 2 \times 4^\alpha \) in the last-mentioned reference, the data actually seem to correspond to the above choice (without the inessential factor 2).
Clearly, which method is most efficient depends on details of the simulated dynamics. In the presence of long range interactions, i.e. when a single event principally modifies the rates of all others, the $\text{dt}$ algorithm is the only practicable choice, since it requires to consider only a single rate $r_i$, namely that of the attempted event, at a time (cf. the MC simulations in Part III of the present work).

Apart from that, ct algorithms tend to be more efficient in situations where the acceptance rate of the trials would be low for a majority of the events. Moreover, since there is no discrete (fixed) minimal time step corresponding to a flip trial, they adapt unconditionally to varying time scales in the course of the simulation. The latter point may be of some interest for conceivable ‘hybrid’ dynamical schemes as outlined in sec. 6.2. As for the two mentioned ct methods, the first one may be more efficient than the ‘waiting time method’ when many of the $r_i$ are identical, such that the choice according to $p(r_i)$ is simplified, and vice versa.

In our implementation of MC dynamics on the hierarchical lattice, we find the dt algorithm considerably faster (about a factor 3 at $T/T_c = 0.8$) than the ‘waiting time method’. Thereby, acceptance rates for trials in the former are around 0.2. For the connectivity weighted dynamics, the competitive advantage of the dt method reduces to a factor $\sim 1.7$. Note, however, that these numbers refer to a ‘multipurpose’ implementation of the ‘waiting time’ algorithm. In particular, the search tree (to find the smallest $t_i$) has not been adapted specifically to peculiarities of our system, e.g. faster flips of low level spins, which might eventually offer some optimization potential.

In terms of absolute runtimes, the simulation of a single configuration (of random bonds and stochastic dynamics; system size $L/L_0 = 2^5$; dt algorithm) up to $t/\tau_0 = 10^7$ as in fig. 6.5(b) takes about 17h on a contemporary Intel Pentium IV 2.8 GHz CPU. We note that, due to the strong increase of relevant time scales even on a logarithmic scale, growing computing power seems not likely to redress the basic limitations of conventional MC simulations of the present system as discussed in sec. 6.2.
B Appendix to Part III

B.1 Energy and force calculations in the simulations

B.1.1 General procedure

For an efficient calculation of the energy $E(x, y)$ of (or force $F(x, y) = -\nabla E(x, y)$ acting on) a pair of dipoles $i, j$ with distance vector $\mathbf{r}_{i,j} = (x, y)$, including their images in the periodically continued systems of the simulation box, we store the corresponding values on a fine grid of pair vectors and use a linear interpolation to obtain the values for $\mathbf{r}_{i,j}$ in the continuum. According to the ‘minimum image convention’, possible distances in $x$- and $y$-direction fall in the range $-L_x/2 \leq x < L_x/2$ and $-L_y/2 \leq y < L_y/2$, where $L_x$ and $L_y$ are the lengths of the system in the $x$- and $y$-direction. We define by $\gamma \equiv L_x/L_y$ the aspect ratio. Due to symmetry, $E(x, y)$ is an even function of $x$ and $y$, while the force components $F_x(x, y) = -\partial_x E(x, y)$, $F_y(x, y) = -\partial_y E(x, y)$ are odd functions of $x$, $y$, and even functions of $y$, $x$, respectively. These symmetries are used to reduce the storage needs for the matrices of energy values and force components on the grid.

For short notation, we use in this appendix $L_y = 1$ as our length unit and $\mu_0 m_i m_j / 4\pi L_y^3$ as our energy unit, where $m_i$ and $m_j$ are the magnetic moments of the two dipoles with pair vector $\mathbf{r}_{i,j}$ (transformation to the units used in the main text follows after elementary rescaling). Then we have

$$E(x, y) = \sum_{\mu, \nu = -\infty}^{\infty} \frac{1}{[(x + \gamma \mu)^2 + (y + \nu)^2]^{3/2}}. \quad (B.1)$$

The numerical calculation of this absolutely convergent series can be done by different means, for example by employing a two-dimensional variant of the Ewald summation (see e.g. [All90]) or a simple extrapolation scheme, cf. sec. 8.3. We applied a method developed previously in our group [Rin98b], where the series in (B.1) is decomposed into an inner part for distances $\sqrt{(x + \gamma \mu)^2 + (y + \nu)^2}$ smaller than a cutoff radius $r_m$, and a remaining outer part, $E = E_{\text{in}} + E_{\text{out}}$. The inner part $E_{\text{in}}$ is calculated by explicitly performing the summation, while the outer part $E_{\text{out}}$ is approximated by an integral. An analogous decomposition is done for the force components. In the following we discuss the integral approximation for the outer parts and their numerical evaluation.
### B.1.2 Integral approximation for the energy

Defining for \( \alpha = 0, 1 \)

\[
\nu_\alpha(\mu) = \begin{cases} 
\text{ceil} \left( \frac{\sqrt{\frac{r_m^2 - \gamma^2 \mu^2}{\alpha}}}{\sqrt{\gamma^2 \mu^2}} \right) & \text{if } \gamma |\mu| < r_m \\
\alpha & \text{else,}
\end{cases}
\]  

where \( \text{ceil}(x) \) is the lowest integer number larger than or equal to \( x \), we obtain

\[
E_{\text{out}} = \sum_{\mu=-\infty}^{\infty} \left[ \sum_{\nu=-\infty}^{\nu_{\mu}(\mu)} \left[ \ldots \right]^{-3/2} + \sum_{\nu=\nu_{1}(\mu)}^{\infty} \left[ \ldots \right]^{-3/2} \right] 
\]  

\[
\int_{-\infty}^{\infty} d\mu \left[ \int_{-\infty}^{\nu_{0}(\mu)} d\nu \left[ \ldots \right]^{-3/2} + \int_{\nu_{1}(\mu)}^{\infty} d\nu \left[ \ldots \right]^{-3/2} + \right. 
\]

\[
\frac{1}{2} \left( \left( x + \gamma \mu \right)^2 + \left( y - \nu_0(\mu) \right)^2 \right)^{-3/2} + \left( x + \gamma \mu \right)^2 + \left( y + \nu_1(\mu) \right)^2 \right)^{-3/2} \right) \right] \]  

\[
\equiv \int_{-\infty}^{\infty} d\mu f(\mu; x, y). \]  

Here \( \left[ \ldots \right] \) stands for \( \left( x + \gamma \mu \right)^2 + \left( y + \nu \right)^2 \). The two integrals over \( \nu \) together yield

\[
\int d\nu \ldots = \frac{1}{\left( x + \gamma \mu \right)^2} \left( 2 - \frac{\nu_0(\mu) - y}{\sqrt{(x + \gamma \mu)^2 + (y - \nu_0(\mu))^2}} - \right. 
\]

\[
- \frac{\nu_1(\mu) + y}{\sqrt{(x + \gamma \mu)^2 + (y + \nu_1(\mu))^2}} \right). \]  

Due to the piecewise definition of \( \nu_\alpha(\mu) \), it is useful to split the remaining integral over \( \mu \) in (B.5) according to its boundaries, with

\[
\int_{-\infty}^{\infty} d\mu f(\mu; x, y) = \int_{|\mu|\leq r_m / \gamma} d\mu f(\mu; x, y) + \int_{|\mu|\geq r_m / \gamma} d\mu f(\mu; x, y) \equiv E_{\text{out,1}} + E_{\text{out,2}}. \]  

The first part \( E_{\text{out,1}} \) is calculated analytically. With the abbreviations

\[
a_{\pm} \equiv \sqrt{1 + \left( \frac{y}{r_m \pm x} \right)^2}, \quad b_{\pm} \equiv \sqrt{1 + \left( \frac{y + 1}{r_m \pm x} \right)^2}, \]  

it reads

\[
E_{\text{out,1}} = \frac{1}{\gamma} \left[ \frac{2r_m}{r_m^2 - x^2} - \frac{2}{y} - \frac{a_- - a_+}{2} + \frac{b_- - b_+}{y + 1} + \frac{a_- - a_+}{2y^2} + \frac{b_- - b_+}{2(y + 1)^2} \right]. \]  

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If \( y = 0, \) the limit \( y \to 0 \) should be taken explicitly to avoid numerical instabilities,

\[
\lim_{y \to 0} E_{\text{out,1}} = \frac{1}{\gamma} \left[ 2 \left( \frac{1}{r_m - x} + \frac{1}{r_m + x} \right) + (2 - b_{\text{m}} - b_{\text{p}}) + \frac{1}{4} \left( \frac{1}{(r_m - x)^2} + \frac{1}{(r_m + x)^2} \right) + \left( 1 - \frac{1}{2b_{\text{m}}} - \frac{1}{2b_{\text{p}}} \right) \right]. \tag{B.10}
\]

The second part \( E_{\text{out,2}} \) is approximated by a sum,

\[
E_{\text{out,2}} \simeq \sum_{\mu = -\mu_1 + 1}^{\mu_1 - 1} f(\mu; x, y) + \left( \frac{1}{2} - \varepsilon \right) \left[ f(-\mu_1; x, y) + f(\mu_1; x, y) \right], \tag{B.11}
\]

where \( \mu_1 \equiv \text{ceil}(r_m/\gamma) \) and \( \varepsilon \equiv \mu_1 - r_m/\gamma. \) For numerical stability, here the limit \( x \to 0 \) of the addend with \( \mu = 0 \) should be considered explicitly,

\[
\lim_{x \to 0} f(0; x, y) = \frac{1}{2} \left[ (r_m - y)^{-3} + (r_m + y)^{-3} + (r_m - y)^{-2} + (r_m + y)^{-2} \right]. \tag{B.12}
\]

In the numerics, the evaluation of (B.11) can conveniently be combined with the summation for the inner part \( E_{\text{in}}. \) For the energy matrix (and for the force calculation discussed in the next section), we use \( r_m = 10. \) If \( E \) is to be calculated repeatedly in a simulation, \( r_m \approx 5 \) might be a reasonable choice. In ample tests in the range \( 0.8 \leq \gamma \leq 1.2, \) we find the relative error of the integral approximation of the energy not to exceed \( 8 \times 10^{-4} \) for \( r_m = 5 \) and \( 10^{-4} \) for \( r_m = 10. \)

### B.1.3 Integral approximation for the force

The outer part \( F_{x}^{\text{out}} \) of the \( x \) component force is

\[
F_{x}^{\text{out}} = \int_{-\infty}^{\infty} d\mu \ldots
\]

\[
\times \left[ \frac{1}{x + \gamma \mu} \left( \frac{y - v_0(\mu)}{[(x + \gamma \mu)^2 + (y - v_0(\mu))^2]^{3/2}} - \frac{y + v_1(\mu)}{[(x + \gamma \mu)^2 + (y + v_1(\mu))^2]^{3/2}} \right) + \frac{2}{(x + \gamma \mu)^3} \left( \frac{y - v_0(\mu)}{[(x + \gamma \mu)^2 + (y - v_0(\mu))^2]^{1/2}} - \frac{y + v_1(\mu)}{[(x + \gamma \mu)^2 + (y + v_1(\mu))^2]^{1/2}} \right) + \frac{3(x + \gamma \mu)^2}{2} \left( \frac{1}{[(x + \gamma \mu)^2 + (y - v_0(\mu))^2]^{5/2}} + \frac{1}{[(x + \gamma \mu)^2 + (y + v_1(\mu))^2]^{5/2}} \right) \right] \tag{B.13}
\]

\[
\equiv \int_{-\infty}^{\infty} d\mu g(\mu; x, y). \tag{B.14}
\]
Analogous to (B.7), the integral over $\mu$ is split into two parts, of which the first one yields

$$
F_{x,1}^{\text{out}} = \frac{1}{\gamma} \left\{ \frac{1}{y} \left[ \frac{1}{|r_m - x|} \left( \frac{1}{a_-} - a_- \right) - \frac{1}{|r_m + x|} \left( \frac{1}{a_+} - a_+ \right) \right] 
- \frac{1}{y + 1} \left[ \frac{1}{|r_m - x|} \left( \frac{1}{b_-} - b_- \right) - \frac{1}{|r_m + x|} \left( \frac{1}{b_+} - b_+ \right) \right] 
- \frac{1}{2|r_m - x|^3} \left( \frac{1}{a_-^2} + \frac{1}{b_-^2} \right) + \frac{1}{2|r_m + x|^3} \left( \frac{1}{a_+^2} + \frac{1}{b_+^2} \right) 
+ 2 \left( \frac{1}{(r_m + x)^2} - \frac{1}{(r_m - x)^2} \right) \right\},
$$

while the second part is approximated by a sum analogous to (B.11),

$$
F_{x,2}^{\text{out}} \simeq \sum_{\mu = -\mu_1 + 1}^{\mu_1 - 1} g(\mu; x, y) + \left( \frac{1}{2} - e \right) \left[ g(-\mu_1; x, y) + g(\mu_1; x, y) \right].
$$

Since the approximation turns out to be inaccurate in the neighborhood of $x = 0$ and $x = 0.5\gamma$, it is advantageous to perform the force calculation in the ranges $|x|/\gamma \lesssim 5 \times 10^{-4}$ and $0.5 - |x|/\gamma \lesssim 10^{-2}$ by explicit summation.

The corresponding expressions for the force component $F_y^{\text{out}}$ can be obtained from the expressions for $F_x^{\text{out}}$ by interchanging $x$ and $y$, replacing $\gamma$ by $1/\gamma$, and rescaling by $1/\gamma^4$, i.e. $F_y^{\text{out}}(x, y; \gamma) = 1/\gamma^4 F_x^{\text{out}}(y/\gamma, x/\gamma, 1/\gamma)$. 

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