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Analytical and numerical approaches to the spin-facilitated kinetic Ising model

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Abstract

A survey is presented of recent progress in studying kinetically constrained Ising models using numerical approaches, a Fock-space formulation of the master equation and a mode-coupling approach, respectively. To this end, numerical results of various kinds are discussed, and also in connection with some ideas related to the glass transition. Furthermore, kinetically facilitated Ising models, considered as candidates for describing systems with a pronounced cooperativity, are reformulated in a second-quantized formulation. Using this approach, kinetic constraints can be incorporated into an analytical study. We discuss in great detail a one-dimensional model and mean-field approaches of several kinds; finally, we give some ideas for a mode-coupling approach to the slow dynamics of such kinetically constrained Ising models in higher dimensions.

1. Introduction

A central topic in statistical physics is the qualitative and quantitative understanding of long-time phenomena in strongly interacting many-body systems, which exhibit an extreme slowing down in the structural relaxation dynamics near a certain point, e.g. a system-specific value of temperature, which defines a transition of the system state. Investigations of dense systems that show no long-range-ordered state or singular behaviour of static quantities, in contrast to the well-known critical slowing down of conventional phase transitions, are of particular interest. Such phenomena are purely kinetic in origin and characterized by a high cooperativity of local processes [1]. This means that the timescale for structural rearrangement increases dramatically for decreasing temperatures. Therefore, cooperativity leads to the slowing down of the relaxation with decreasing temperature. For a large class of dense systems some essential properties of the slowing-down regime, e.g. a stretched-exponential decay of autocorrelation functions and a non-Arrhenius temperature behaviour of relaxation times, seem to be characteristic or universal features for the primary relaxation process at sufficiently

long times. Such a scenario has often been observed for the main glass transition process in supercooled liquids, which is usually called the α-relaxation.

Furthermore, there exists also an interesting class of non-equilibrium models, which are related to facilitated kinetic spin systems. These models show a typical slowing down of the structural relaxation dynamics for long times caused by an increasing cooperativity of local spin-flip processes with decreasing temperature. Our interest in the present paper is focused on the f-spin-facilitated kinetic Ising models [2–4], originally introduced by Fredrickson and Andersen. These models are formulated on d-dimensional lattices. Each lattice point i is characterized by a spin variable σ_i with two possible states $\sigma_i = \pm 1$. The set of all observables $\boldsymbol{\sigma} = \{\sigma_i\}$ forms a configuration. The time evolution of the whole Ising spin system is described by a master equation

$$\frac{\partial P(\boldsymbol{\sigma}, t)}{\partial t} = \sum_{\sigma'} L(\boldsymbol{\sigma}, \boldsymbol{\sigma}') P(\boldsymbol{\sigma}, t) \tag{1}$$

where $P(\boldsymbol{\sigma}, t)$ is the probability that a certain configuration $\boldsymbol{\sigma}$ is realized at time t and $L(\boldsymbol{\sigma}, \boldsymbol{\sigma}')$ is the system-specific dynamical matrix. Hence, the underlying dynamics of the f-spin-facilitated kinetic Ising models is a stochastic one, which is given by a simple Glauber process [5], i.e. spin flips $\sigma_i = +1 \leftrightarrow \sigma_i = -1$ are controlled by the thermodynamic Gibbs measure and by self-induced topological restrictions. In particular, the topological conditions are explicitly taken into account such that an elementary flip at a given lattice point i is only allowed if the number of the nearest-neighbour lattice points in the spin-up state ($\sigma_i = +1$) is equal to or larger than a restriction number f with $0 < f < z$ (here z is the coordination number of the lattice). Such a model [2–4, 6] is referred to as an f-spin-facilitated Ising model on a d-dimensional lattice, and abbreviated as SFM[f, d]. Ising spins on different lattice sites are normally coupled only via kinetic constraints, i.e. there are no pair interactions between neighbouring spins. In this way, elementary single-spin-flip processes combined with geometrical restrictions lead inevitably to cooperative rearrangements in the underlying spin system.

2. Numerical simulations

Apart from experimental observations, numerical studies are very powerful methods for analysing the relaxation dynamics in systems with high intrinsic cooperativity.

Starting from a configuration $\sigma_i = +1$, thermal equilibrium is established after a certain relaxation time. However, this holds only provided that the restriction number f is sufficiently small. Otherwise, the kinetic rules prevent equilibrium from being attained and only a stationary state is achieved. On a d-dimensional square lattice, $f \leqslant d$ has to be satisfied in order to achieve thermal equilibrium. Therefore, the following statements are based mostly on investigations of the SFM[d, d].

Thermal equilibrium is defined independently of the kinetic restrictions by the Hamiltonian

$$\frac{H}{T} = -J \sum_{\langle ij \rangle} \sigma_i \sigma_j + h \sum_i \sigma_i \qquad \sigma_i = \pm 1. \tag{2}$$

The quantity h is always positive whereas the coupling constant is chosen to be $J = 0$ for most investigations. Formally, h scales with the inverse temperature.

2.1. Non-exponential decay

The autocorrelation function $C(t)$ and the global mobility $F(t)$ are used for the numerical analysis of the kinetics of the SFM[f, d] in most cases. While the autocorrelation function is

given by (L is the lattice size)

$$C(t) = \frac{1}{L^d} \sum_i \left[\langle \sigma_i(t)\sigma_i(0) \rangle - \langle \sigma_i(0) \rangle^2 \right] \tag{3}$$

the global mobility function $F(t)$ is the fraction of spins which remain unflipped after a time interval t, measured from some arbitrary initial time, has elapsed. At sufficiently low temperatures, both functions show a characteristic non-exponential decay [7–9]. However, the decay of $C(t)$ seems to be faster than the decay of $F(t)$.

The SFM[1, 1] shows a slightly modified exponential decay of $C(t)$ for all temperatures [10], whereas $\log F(t) \sim -\sqrt{t}$. These relations can also be verified analytically. The spin-autocorrelation function of the SFM[2, 2], on the other hand, shows a characteristic behaviour which can be fitted over a large time interval by a stretched-exponential decay

$$C(t) \sim \exp[-(t/\tau)^\gamma] \tag{4}$$

where the exponent γ is presumably non-universal. Recent simulations [9] suggest that the exponent is in the range $\gamma = 0.43$–0.54, with a weak dependence on temperature (or alternatively on h) and on the coupling strength J.

The global mobility function $F(t)$ can also be fitted using (4), but the exponent becomes very small for low temperatures [8]. It is an open question whether γ converges to 0 or to a finite value for $T \to 0$.

2.2. Relaxation times

The relaxation time τ can be obtained from the fit procedure using (4). One obtains a characteristic non-Arrhenius temperature behaviour which can be approximated, at least over a large temperature interval, by a Vogel–Fulcher law [7, 11] or by a power law [12] $\log \tau \sim T^{-\delta}$. It is remarkable that the relaxation regime of facilitated kinetic Ising models and the α-relaxation of real supercooled liquids exhibit qualitatively the same long-time behaviour. Consequently, cooperativity due to local kinetic constraints seems to be a fundamental physical mechanism for understanding the collective nature of the primary relaxation process in the slowing-down scenario of dense systems. However, as demonstrated previously [10] there is no indication of a real glass transition or a critical temperature as predicted by mode-coupling theory [13, 14]. In particular, the relaxation time of every equilibrated SFM[f, d] diverges only at zero temperature.

2.3. Control parameter

Any finite value of the coupling constant J generates correlations in the spatial distribution of the spins. There is then the question of whether this distribution has an essential influence on relaxation behaviour. We obtain the surprising result that neither J nor h influences the kinetics of the models alone. The relevant control parameter is the number of spins in the up and down state and therefore the magnetization $M(J, h)$ defined by all spins. If we plot the relaxation times as a function of this magnetization, all values collapse onto one curve.

2.4. Cooperative regions

The original definition of cooperatively rearranging regions [1] may be reasonable for the qualitative explanation of some processes in glasses, but it is too imprecise for a well-defined mathematical investigation of cooperative phenomena in the SFM[f, d]. Therefore, we make

the definition that there exists in the environment of each spin σ_i a time- and temperature-dependent region $R_i(t, T)$ containing the minimum number of other spins that must flip at least once before σ_i can flip. The spins in a cooperative region $R_i(t, T)$ form a connected cluster. The distribution function of the cluster sizes suggests a kinetic behaviour similar to the so-called heterogeneous dynamics which is known from supercooled liquids [12]. The temperature-dependent mean size of cooperative regions can also be fitted by a power law $\bar{R} \sim T^{-\beta}$.

2.5. Modifications

The SFM$[f, d]$ can be generalized by including additional degrees of freedom such as vacancies [8, 15] and new kinetic rules. The dynamical rules are then modified in such a manner that a vacant state favours the flip process. Furthermore, the vacancies are able to perform hoppings between nearest neighbours. As a result of these modifications a new relaxation process appears which can be identified with the Johari–Goldstein process observed in supercooled liquids. In [15] one can find an analytical approach for this modified SFM$[f, d]$ based on a three-state spin model.

The SFM can be further generalized by including chemical reactions [16], kinetic growth of an interface within the glass matrix [17] and the mixed-mobile-ion effects in glasses [18].

3. Fock-space approach

First we give a short review of the main ideas behind the Fock-space formalism, which is a very powerful method for analysing classical many-body systems with a stochastic dynamics given by a master equation on a lattice. The Fock-space approach is based on a quantum-like formulation of the underlying master equation written in terms of creation and annihilation operators. The Fock-space representation of the SFM$[2, d]$ can be made more transparent by interpreting the two spin orientations $\sigma_i = -1$ and 1 as empty and singly occupied sites corresponding to the occupation numbers $n_i = 0$ and 1 via $\sigma_i = 2n_i - 1$, respectively. With this, the SFM$[2, d]$ can be interpreted as a lattice gas with excluded volume; i.e. changes of the configuration $\boldsymbol{n} = \{n_i\}$ are possible only subject to the exclusion principle. Now, make the replacement $\boldsymbol{\sigma} \to \boldsymbol{n}$ in (1); following [19–22], the probability distribution $P(\boldsymbol{n}, t)$ can then be related quite generally to a state vector $|F(t)\rangle$ in a Fock space according to $P(\boldsymbol{n}, t) = \langle \boldsymbol{n}|F(t)\rangle$ and $|F(t)\rangle = \sum_{\boldsymbol{n}} P(\boldsymbol{n}, t)|\boldsymbol{n}\rangle$, respectively, where the basis vectors $|\boldsymbol{n}\rangle$ are composed of second-quantized operators. Using this representation, the Master equation (1) can be transformed to an equivalent evolution equation in a Fock space

$$\partial_t |F(t)\rangle = \hat{L}|F(t)\rangle. \tag{5}$$

The dynamical matrix $L(\boldsymbol{n}, \boldsymbol{n}')$ in (1) is mapped onto the operator $\hat{L} = \hat{L}(d, d^\dagger)$, which is given in a second-quantized form with d and $d^\dagger$ being the annihilation and creation operators, respectively. Originally, this transformation had been applied for the Bose case with unrestricted occupation numbers [19–21]. Here, we consider the case of restricted occupation numbers [22]. In order to preserve the restriction of the occupation number in the underlying dynamical equations, the commutation rules of the operators $\hat{d}$ and $\hat{d}^\dagger$ are chosen as those of Pauli operators [22–24]:

$$[\hat{d}_i, \hat{d}_j^\dagger] = \delta_{ij}(1 - 2\hat{d}_i^\dagger \hat{d}_i) \qquad [\hat{d}_i, \hat{d}_j] = [\hat{d}_i^\dagger, \hat{d}_j^\dagger] = 0 \qquad \hat{d}_i^2 = (\hat{d}_i^\dagger)^2 = 0. \tag{6}$$

In principle, as was shown firstly by Doi [19], the average of an arbitrary physical quantity $B(\boldsymbol{n})$ is given by the average of the corresponding operator $\hat{B}(t) = \sum_n |\boldsymbol{n}\rangle B(\boldsymbol{n})\langle \boldsymbol{n}|$ via

$$\langle \hat{B}(t)\rangle = \sum_{\boldsymbol{n}} P(\boldsymbol{n}, t)B(\boldsymbol{n}) = \langle s|\hat{B}|F(t)\rangle \tag{7}$$

using the reference state $\langle s| = \sum_n \langle \boldsymbol{n}|$. The normalization condition is expressed by the relation $\langle s|F(t)\rangle = 1$. A special feature of this Fock-space formulation is the fact that the mean value is linear in the corresponding state vector, while in quantum mechanics it is bilinear. In the same way, correlation functions can be expressed as

$$\langle \hat{A}(t)\hat{B}(t')\rangle = \sum_{\boldsymbol{n},\boldsymbol{n}'} A(\boldsymbol{n})P(\boldsymbol{n}, t; \boldsymbol{n}', t')B(\boldsymbol{n}') = \langle s|\hat{A}\exp\{\hat{L}(t-t')\}\hat{B}|F(t')\rangle.$$

Furthermore, because of (5) and (7) the evolution equation for an arbitrary operator $\hat{B}(t)$, for example the particle number operator, is given by [25]

$$\partial_t\langle \hat{B}\rangle = \langle s|\,[\hat{B}, \hat{L}]\,|F(t)\rangle \tag{8}$$

which can be extended immediately in order to allow one to write down the kinetic equations for time-dependent correlation functions. To derive the last equation we have used the relation $\langle s|\hat{L} = 0$, which is necessary to guarantee the conservation of the normalization condition. As a general result of the procedure, all the dynamical equations governed by the classical problem are completely determined by the commutation rules of the underlying operators and the structure of the evolution operator $\hat{L}$. Therefore, this method allows investigations of master equations for various evolution processes, e.g. aggregation, chemical reactions [26,27], non-linear diffusion [28] as well as the spin-facilitated kinetic Ising models. The decisive advantage of the Fock-space approach is given by a simple construction principle for each evolution operator $\hat{L}$ on the basis of creation and annihilation operators.

Thus, using the Fock-space formalism the master equation of the SFM[2, d] (with coupling constant $J = 0$ and $h = \varepsilon/2T$) can be expressed by the following evolution operator [29]

$$\hat{L} = \sum_{i,j,k} \kappa_{i|jk}\hat{D}_j\hat{D}_k\big[\beta(\hat{d}_i - \hat{D}_i) + \lambda(\hat{d}_i^{\dagger} - (1 - \hat{D}_i))\big] \tag{9}$$

with the particle number operator $\hat{D}_i = \hat{d}_i^{\dagger}\hat{d}_i$ (with $\hat{D}_i|\boldsymbol{n}\rangle = n_i|\boldsymbol{n}\rangle$) and temperature-dependent jump rates λ and β. The terms inside the square bracket in (9) represent a single-spin-flip process at lattice site i and the product of the particle number operators $\hat{D}_j\hat{D}_k$ represents the local constraints. Additionally, $\kappa_{i|jk}$ is a lattice function with $\kappa_{i|jk} = 1$ if $j \neq k$ and j and k are neighbours to lattice cell i. Due to detailed balance one obtains for the jump rates [25]

$$\beta = \nu^{-1}(T) \qquad \text{and} \qquad \lambda = \nu^{-1}(T)\exp(-\varepsilon/T) \tag{10}$$

where in general $\nu^{-1}(T)$ is an elementary temperature-dependent timescale represented by simple activation dynamics like $\nu^{-1}(T) = k\exp(-E_A/T)$ with the parameter k (measure of a microscopic timescale) and the activation energy barrier E_A (a useful tool for computer simulations). Without loss of generality, we set in our calculations $E_A = 0$. Therefore, the microscopic timescale is simply a constant. Moreover, ε is the energy difference between the up and down states. Using (8) with the operator $\hat{L}_i = \beta(\hat{d}_i - \hat{D}_i) + \lambda(\hat{d}_i^{\dagger} - (1 - \hat{D}_i))$ for free spins [25], the evolution equation for the averaged particle number operator reads

$$\partial_t\langle \hat{D}_i\rangle = -\beta\langle \hat{D}_i\rangle + \lambda\big(1 - \langle \hat{D}_i\rangle\big) \tag{11}$$

and the solution of this kinetic equation shows an exponential decrease characterized by the relaxation time $\tau_0 = (\lambda + \beta)^{-1}$. The stationary state $\partial_t\langle \hat{D}_i\rangle = 0$ corresponds to an equilibrium averaged occupation number $\bar{n}_{\mathrm{eq}} = \langle \hat{D}_j\rangle = \lambda/(\lambda + \beta)$, which is controlled by the ratio of the energy difference ε and the temperature T. The average magnetization follows directly from $M = 2\bar{n}_{\mathrm{eq}} - 1$.

4. One-dimensional model

In one dimension, only the restriction parameters $f = 0$, 1 and 2 need to be considered. However, since thermal equilibrium is not attainable for $f = 2$ and 0 corresponds to the trivial case of the paramagnetic gas, the remaining case $f = 1$ is the only physically relevant one. Therefore, we focus exclusively on the SFM[1, 1] in this section.

4.1. Short-time limit

The time evolution of the averaged occupation number $\bar{n}_i = \langle \hat{D}_j \rangle$ satisfies the equation

$$\partial_t \langle \hat{D}_i \rangle = \lambda\big(\langle \hat{D}_{i-1} \rangle + \langle \hat{D}_{i+1} \rangle\big) - (\lambda + \beta)\big[\langle \hat{D}_{i-1} \hat{D}_i \rangle + \langle \hat{D}_{i+1} \hat{D}_i \rangle\big].$$

The simplest way of truncating the hierarchy is the decoupling $\langle \hat{D}_{i-1} \hat{D}_i \rangle \approx \langle \hat{D}_{i-1} \rangle \langle \hat{D}_i \rangle = \bar{n}^2$ which leads to the mean-field solution

$$\bar{n}(t) = \frac{\bar{n}_{\mathrm{eq}}}{1 - (1 - \bar{n}_{\mathrm{eq}}) \exp\{-t/(2\tau_0)\}}.$$

Another possibility is based on a more accurate decoupling [30]. As result of this procedure the averaged occupation number $\bar{n}(t)$ satisfies an integral equation which can be solved in the short-time regime:

$$\bar{n}(t) = \bar{n}_{\mathrm{eq}} + 2(1 - \bar{n}_{\mathrm{eq}}) \exp\{-2t/\tau_0\} \frac{I_1(t/t_0)}{t/t_0}.$$

Here $I_1(x)$ is the first-order modified Bessel function and t_0 is a second relaxation constant defined by $t_0^{-1} = 4\sqrt{\lambda\beta}$. An exact solution is possible for $T = 0$. With the aim of obtaining this solution we use the cluster function

$$\Phi_i^m = \prod_{j=0}^{m} \hat{D}_{i+j} \equiv \hat{D}_i \hat{D}_{i+1} \cdots \hat{D}_{i+m}. \tag{12}$$

Such a function gives a non-vanishing contribution only when all the $m+1$ lattice points of the cluster are occupied, i.e. when all spins of the cluster are in the up state. Using the algebraic properties of the Pauli operators we can derive an exact evolution equation [30] for the $\langle \Phi_i^m \rangle$. In particular, considering as initial condition a parallel alignment $\langle \Phi^m \rangle(t = 0) = 1$ (for all $m > 0$), we get

$$\langle \Phi^m \rangle(t) = \exp\left\{-\frac{2mt}{\tau_0}\right\} \exp\{\mathrm{e}^{-t/\tau_0} - 1\}.$$

The spin-up cluster function reveals a double-exponential decay. In the initial time regime the conventional exponential decay dominates. But the concentration of isolated up spins $\langle \Phi^0 \rangle(t)$ shows a more pronounced slowing down that leads to non-ergodic behaviour, which is shown in the limit $t \to \infty$ by $\langle \Phi^0 \rangle(\infty) = \mathrm{e}^{-1}$. This result corresponds to the general fact that a crossover of an ergodic SFM[f, d] to non-ergodic behaviour occurs only for $T = 0$.

4.2. Long-time limit

The long-time limit cannot be described accurately for $T > 0$. However, at very low temperatures the SFM[1, 1] is characterized by a small concentration of single spins in the up state. Then two neighbouring spins can exchange their orientation. Blocks of up spins of length $L \geqslant 2$ are extremely rare and have only a short lifetime; they will therefore be neglected. As discussed in detail in [10] one should distinguish three types of elementary local process:

(i) *Diffusion*: a down spin next to an up spin changes its orientation with probability $\lambda \simeq \exp(-\varepsilon/T)$ and becomes an up spin. Then, the original up spin changes its state to a down spin. The result is an effective diffusion of a single up spin.

(ii) *Creation of up spins*: one of the two down spins next to the single up spin changes its state with probability λ. Then, one of the two down spins next to the up spin block of length 2 changes its state, so a spin-up block of length 3 is created. The central up spin of this block flips down with probability ≈ 1. Thus, one obtains two single up spins, separated by one down spin.

(iii) *Annihilation of up spins*: a single up spin can be next to another single up spin, separated only by one down spin. The down spin between the two up spins can then change its state and one obtains an up spin block of length 3. Finally, the length is reduced to 2 and then to 1.

Taking into account the three elementary processes described we are able to find the autocorrelation function

$$C(t) = \exp\left(-\frac{\alpha_1^2 t}{2}\right) \frac{\Phi(\pi\sqrt{\alpha_2 t/2})}{\sqrt{t}}. \tag{13}$$

Here α_1 and α_2 are constant parameters, and Φ is the complementary error function. This correlation function agrees very well with numerical simulations [10]. In the same manner, we have calculated the global memory function $F(t)$ introduced earlier, which is a measure of the fraction of spins which remain unflipped after some time t, measured from an arbitrary initial time. We get

$$F(t) \simeq \exp\left(-\frac{\sqrt{\bar{n}_{\mathrm{eq}} t}}{\xi_0}\right)$$

with $\xi_0 = -1/\ln(1-\bar{n}_{\mathrm{eq}})$. This analytical result is also in rather good agreement with simulations [10].

5. Mean-field approaches

In higher dimensions the Fock-space formulation for a non-equilibrium system based on a master equation is also applicable. However, in this case one can perform approximations of mean-field type. The typical structure of the basic evolution operator is given by equation (9); compare [29]. In terms of the lattice gas variable, the Hamiltonian equation (2) can be rewritten as

$$H = 2\sum_i (Jz + h)\hat{D}_i - 4J \sum_{\langle i,j \rangle} \hat{D}_i \hat{D}_j.$$

Here z is the coordination number of the lattice.

5.1. Standard mean-field approach

In the standard mean-field approximation the complete hierarchy for the averaged occupation number $\langle \hat{D}_i(t) \rangle$ is reduced to the following one:

$$\partial_t \bar{n} = \bar{n}^f \left[\lambda(1-\bar{n}) - \beta\bar{n}\right]. \tag{14}$$

Performing a linear stability analysis around the stationary state n_{eq} we get the relaxation time

$$\frac{\tau}{\tau_0} = \frac{\bar{n}_{\mathrm{eq}}^{-f}}{2[\cosh(\beta\varepsilon_0/2) - 2T^\star \bar{n}_{\mathrm{eq}} \exp(\beta\varepsilon_0/2)/T]}.$$

Here we have introduced $\varepsilon_0 = 2(h+T^\star(2\bar{n}_{\rm eq}-1))$ with the characteristic temperature $T^\star = Jz$. Furthermore, τ_0 stands for a microscopic timescale expressed by the inverse hopping rate ν and the coordination number. Due to the kinetic constraints, the relaxation time of the SFM[f, d] is defined by the relaxation time of the conventional kinetic Ising model without restrictions scaled with the factor $\bar{n}_{\rm eq}^{-f}$. When the interaction parameter J is zero, the relaxation time shows different behaviours for low and high temperatures:

$$\ln\left(\frac{\tau}{\tau_0}\right) \simeq \begin{cases} (f-1)\ln 2 + \dfrac{fh}{2T} & \text{for} \quad h/T \ll 1 \\ \dfrac{(2f-1)h}{2T} & \text{for} \quad h/T \gg 1. \end{cases} \tag{15}$$

Asymptotically there appear two Arrhenius trajectories. However, the low-temperature branch exhibits a slowing down compared to the high-temperature region, as shown by a smaller slope. The crossover of the two curves is roughly estimated to be $h/T = 2\ln 2$. In general, the relaxation process is a non-Arrhenius process.

The inclusion of mutual interaction between regions of different mobility leads, within the mean-field approximation, to a temperature- and state-dependent activation energy $\varepsilon = 2(h + T^\star(2\langle n\rangle - 1))$. The characteristic temperature $T^\star = Jz$ does not signal a second-order phase transition because the activation energy has to be a non-zero parameter. In the low-temperature limit $T \to 0$ the relaxation time tends to infinity:

$$\frac{\tau}{\tau_0} \simeq \exp\left((z-1)\frac{\tilde{h}_0}{2T}\right)$$

with

$$\tilde{h}_0 = 2\left\{h + T^\star\left[1 - 2\exp\left(-2\frac{h}{T} - 2\frac{T^\star}{T}\right)\right]\right\}.$$

Note that the activation energy is always non-zero and hence the increase of the relaxation time τ remains finite. There is no real phase transition at finite temperature.

5.2. *Spatial fluctuations*

Up to now we have neglected spatial fluctuations originating from the static coupling strength J. In the spirit of the mean-field approach we can include lowest-order gradient terms by using the relation

$$\sum_{l(i)} J_{il}\hat{D}_l = \sum_{r(i)} J_{ir}(\hat{D}_r - \hat{D}_i) + zJ\hat{D}_i \approx J(\nabla^2\hat{D}_i + z\hat{D}_i).$$

As the result, we get an evolution equation for the averaged density field $\bar{n}(\boldsymbol{x}, t)$ in the form

$$\partial_t\bar{n} = \bar{n}^f\,(\lambda(1-\bar{n}) - \beta\bar{n}) + 2J/T\bar{n}^f\,(\lambda(1-\bar{n}) + \beta\bar{n})\,\nabla^2\bar{n}. \tag{16}$$

To study the influence of spatial fluctuations we make the ansatz $\bar{n}(\boldsymbol{x}, t) = \bar{n}(t) + m(\boldsymbol{x}, t)$ where $\bar{n}(t)$ satisfies equation (14). To lowest order, the function $m(\boldsymbol{x}, t)$ satisfies a diffusion-like equation, but with a time-dependent diffusivity [29]. From this we can estimate the perimeter L of the spin-down regions which are self-organized by the underlying restrictions. In the long-time limit the result is

$$L^2 \simeq \frac{2J}{fT}\ln(t/t_0).$$

The perimeter is small in the initial time regime and increases on a logarithmic scale in the long-time limit. Let us remark that we are not very deep inside the regime of high cooperativity; there, we would expect a more pronounced increase related to the very strong decrease of the mobility.

5.3. Nucleation processes

As a consequence of the complex relaxational behaviour we have also studied the nucleation process in an environment determined by the SFM[f, d]. When a glass former is cooled down, the formation of droplets of the solid phase is prevented. So, we have modified the Lifshitz–Slyozov–Wagner (LSW) theory [31, 32] and have shown that the nucleation radius of droplets remains finite at low temperatures. Using the low-temperature limit for the relaxation time, equation (15), we get the averaged droplet radius

$$\bar{R} \simeq \kappa^{-1/3} \left\{ \mathrm{Ei}\left(\frac{(2f-1)h}{2T^\star} \right) + \frac{2T^\star}{(2f-1)h} \exp\left[-(2f-1)h/T^\star \right] \right\}^{1/3} \tag{17}$$

where $\mathrm{Ei}(x)$ is the exponential integral. In contrast to the conventional LSW case, the droplet radius remains finite; droplets of finite size both appear and disappear. Nucleation is strongly reduced, i.e. a sufficiently fast cooling procedure prevents crystallization.

6. Mode-coupling theory

6.1. Projection formalism

The general basis for an analytical study of long-time phenomena in arbitrary physical systems via a mode-coupling approach is given by the Mori–Zwanzig projection operator formalism [33]. Here we investigate the slow-relaxation regime of model classes where the underlying microscopic dynamics is stochastic due to an irreversible master equation [34]. For that purpose we derive in a mathematically consistent way the evolution equations for a complete set of relevant observables by using a projection formalism in the Fock space. Therefore, these projection equations, which should represent the irreversible nature of the underlying dynamics, form a general basis for analytical studies of the whole class of facilitated kinetic Ising models. Here, we restrict our investigation to the analysis of the SFM[2, d]. This means that, simultaneously with the general derivation of the projection equations, we will specify all quantities in the form in which they apply to the SFM[2, d].

6.1.1. Relevant operators. The dynamics of an arbitrary physical system can be described systematically by a reasonable set of relevant operators. It seems to be a suitable choice [33] to use a system-specific relevant observable and its time derivatives up to a certain order as a reasonable set of relevant operators, where the system-specific relevant observable itself is determined by the zeroth time derivative. For the investigation of the SFM[2, d] and moreover for several facilitated kinetic Ising models, the system-specific relevant observable or slow-system variable is given by the normalized local deviations of the spin configuration from the thermodynamic average. In principle, from the mathematical point of view the upper borderline of all possible time derivatives $\beta = 0, 1, \ldots, g_{\max}$ is usually a finite integer number ($g_{\max} < \infty$), but $g_{\max} \to \infty$ is also well defined, so

$$\hat{\eta}_i^{(0)}(t) = \hat{\eta}_i(t) = \frac{\hat{D}_i(t) - \bar{n}_{\mathrm{eq}}}{\sqrt{\bar{n}_{\mathrm{eq}}(1-\bar{n}_{\mathrm{eq}})}} \qquad \text{and} \qquad \hat{\eta}_i^{(\beta)}(t) = \frac{\partial^\beta \hat{\eta}_i(t)}{\partial t^\beta} = \hat{\eta}_i(t)\hat{L}^\beta. \tag{18}$$

These covariant operators must be extended by adding the corresponding contravariant operators in order to define a scalar product $\left(\hat{\eta}_i^{(\alpha)}, \tilde{\eta}_i^{(\beta)}\right) = \left\langle \hat{\eta}_i^{(\alpha)} \tilde{\eta}_i^{(\beta)} \right\rangle$, which corresponds to an arbitrary correlation between a covariant and a contravariant operator. In the case of the SFM[2, d] the contravariant operators are determined by

$$\tilde{\eta}_i^{(0)}(t) = \hat{\eta}_i(t) \qquad \text{and} \qquad \tilde{\eta}_i^{(\beta)}(t) = \hat{L}^\beta \hat{\eta}_i(t). \tag{19}$$

Using (18) and (19) we construct the backward projection operator $\hat{P}$:

$$\cdots \hat{P} = \sum_{\alpha,\beta,i,j} \langle \cdots \tilde{\eta}_i^{(\alpha)} \rangle g_{ij}^{\alpha\beta} \hat{\eta}_j^{(\beta)} \qquad \text{with} \sum_{\alpha,i} \langle \hat{\eta}_k^{(\gamma)} \tilde{\eta}_i^{(\alpha)} \rangle g_{ij}^{\alpha\beta} = \delta^{\gamma\beta}\delta_{kj} \tag{20}$$

with $\alpha, \beta, \ldots \in [0, g_{\max}]$. The projection operator leads to an identical mapping of the relevant operators onto itself, i.e. $\hat{\eta}_k^{(\gamma)} \hat{P} = \hat{\eta}_k^{(\gamma)}$. The orthogonal projection operator $\hat{Q}$ is given by $\hat{Q} = 1 - \hat{P}$ with $\hat{\eta}_k^{(\gamma)} \hat{Q} = 0$. We remark that, as a consequence of the Fock-space approach, all operators act to the left.

6.1.2. Basic equations. The evolution equation (5) leads to the formal solution $|F(t)\rangle = \exp\{\hat{L}t\}|F(0)\rangle$. The dependence of $|F(t)\rangle$ on time can be transferred to an arbitrary operator, by analogy with the transformation of Schrödinger's representation into the Heisenberg picture. Therefore, time-dependent operators can by expressed by $\hat{B}(t) = \hat{B}\exp\{\hat{L}t\}$. Accordingly, the starting point for the derivation of the projection equations for the relevant observables, in analogy with the derivation of the well-known Mori–Zwanzig equations [33, 35] for time-reversible classical or quantum mechanical equations of motion, is given by the following time evolution of $\hat{\eta}_k^{(\gamma)}(t)$ in the Fock space:

$$\frac{\partial \hat{\eta}_k^{(\gamma)}(t)}{\partial t} = \hat{\eta}_k^{(\gamma)}(t)\hat{L}. \tag{21}$$

The application of the identity $1 = \exp\{-\hat{L}t\}(\hat{P} + \hat{Q})\exp\{\hat{L}t\}$ onto the operator $\hat{L}$ from the right-hand side leads to a formal splitting into a relevant and an irrelevant part. Note that $\hat{P}$ realizes a projection onto the subspace $L_\parallel$ of relevant operators, whereas $\hat{Q}$ projects onto the linearly independent subspace $L_\perp$ of all other operators. Hence,

$$\frac{\partial \hat{\eta}_k^{(\gamma)}(t)}{\partial t} = -\sum_{\beta,j} \Omega_{kj}^{(\gamma\beta)} \hat{\eta}_j^{(\beta)}(t) + \hat{\eta}_k^{(\gamma)} \hat{L}\hat{Q}\exp\{\hat{L}t\} \tag{22}$$

with the frequency matrix

$$\Omega_{kj}^{(\gamma\beta)} = -\sum_{\alpha,i} \langle \hat{\eta}_k^{(\gamma)} \hat{L} \tilde{\eta}_i^{(\alpha)} \rangle g_{ij}^{\alpha\beta}. \tag{23}$$

The second term of (22) can be rewritten by using an identical transformation of $\exp\{\hat{L}t\}$ into an integral expression:

$$\exp\{\hat{L}t\} = \int_0^t \mathrm{d}t' \, \exp\{\hat{L}\hat{Q}(t-t')\}\hat{L}\hat{P}\exp\{\hat{L}t'\} + \exp\{\hat{L}\hat{Q}t\}. \tag{24}$$

This relation allows the derivation of rigorous projection equations similar in structure to the usual Mori–Zwanzig equations [33, 35]:

$$\frac{\partial \hat{\eta}_k^{(\gamma)}(t)}{\partial t} = -\sum_{\beta,j} \Omega_{kj}^{(\gamma\beta)} \hat{\eta}_j^{(\beta)}(t) + \int_0^t \mathrm{d}t' \sum_{\beta,j} K_{kj}^{(\gamma\beta)}(t-t')\hat{\eta}_j^{(\beta)}(t') + \hat{f}_k^{(\gamma)}(t) \tag{25}$$

with the residual forces

$$\hat{f}_k^{(\gamma)}(t) = \hat{\eta}_k^{(\gamma)} \hat{L}\hat{Q}\exp\{\hat{L}\hat{Q}t\} = \hat{f}_k^{(\gamma)}\exp\{\hat{L}\hat{Q}t\} \tag{26}$$

which are characterized by the properties $\hat{f}_k^{(\gamma)}(t)\hat{Q} = \hat{f}_k^{(\gamma)}(t)$ and $\hat{f}_k^{(\gamma)}(t)\hat{P} = 0$, and the memory matrix

$$K_{kj}^{(\gamma\beta)}(t-t') = \sum_{\alpha,i} \langle \hat{\eta}_k^{(\gamma)} \hat{L}\hat{Q}\exp\{\hat{L}\hat{Q}(t-t')\}\hat{L}\tilde{\eta}_i^{(\alpha)} \rangle g_{ij}^{\alpha\beta}. \tag{27}$$

The comparison between the projection equations (25) and the standard Mori–Zwanzig equations [33] shows a formal equivalence, because both types of equation contain frequency terms, memory terms and residual forces with a similar mathematical structure. But there is a fundamental difference which can be studied directly by inspecting the memory kernel. On the one hand, the memory terms of the usual Mori–Zwanzig equations can be written always as a correlation function of the residual forces. This relation can be interpreted as a representation of the fluctuation-dissipation theorem, and it is causally connected with the fact that the standard Mori–Zwanzig equations are related to reversible classical or quantum mechanical equations. By way of contrast, it is easy to see that the memory terms (27) cannot be completely constructed from the residual forces (26). The cause is the irreversible character of the underlying master equation.

6.1.3. Projection equations for a reduced set of relevant observables. Now, we restrict the set of relevant operators by setting $g_{\max} = 1$. This is because the relevant observables $\hat{\eta}_i^{(0)}(t) = \hat{\eta}_i(t)$ and $\hat{\eta}_i^{(1)}(t) = \hat{\eta}_i(t)\hat{L}$ represent a suitable set of relevant operators for characterizing completely the long-time dynamics of the SFM[2, d]. This choice corresponds to experience in mechanical systems, which are completely determined by spatial coordinates and velocities. With this choice, the general system of projection equations (25) becomes

$$\begin{aligned}\frac{\partial}{\partial t}\hat{\eta}_k^{(0)}(t) &= \sum_j \sum_{\beta=0,1}\left[-\Omega_{kj}^{(0\beta)}\hat{\eta}_j^{(\beta)}(t) + \int_0^t \mathrm{d}t'\, K_{kj}^{(0\beta)}(t-t')\hat{\eta}_j^{(\beta)}(t')\right] + \hat{f}_k^{(0)}(t)\\ \frac{\partial}{\partial t}\hat{\eta}_k^{(1)}(t) &= \sum_j \sum_{\beta=0,1}\left[-\Omega_{kj}^{(1\beta)}\hat{\eta}_j^{(\beta)}(t) + \int_0^t \mathrm{d}t'\, K_{kj}^{(1\beta)}(t-t')\hat{\eta}_j^{(\beta)}(t')\right] + \hat{f}_k^{(1)}(t).\end{aligned} \tag{28}$$

Note that $g_{\max} = 0$ is used in [36–38], which leads to an apparent ergodic–non-ergodic transition in disagreement with numerical predictions. On using the relations $\Omega_{kj}^{(0\beta)} = -\delta_{kj}\delta^{1\beta}$, $\hat{f}_k^{(0)}(t) = 0$ and $K_{kj}^{(0\beta)}(t-t') = 0$, the first equation of (28) is reduced to the identity $\partial_t\hat{\eta}_k^{(0)}(t) = \partial_t\hat{\eta}_k(t) = \hat{\eta}_k^{(1)}(t)$ and thus the second equation can be rewritten as

$$\begin{aligned}\frac{\partial^2\hat{\eta}_k(t)}{\partial t^2} &= -\sum_j\left[\Omega_{kj}^{(10)}\hat{\eta}_j(t) + \Omega_{kj}^{(11)}\frac{\partial\hat{\eta}_j(t)}{\partial t}\right]\\ &\quad + \sum_j\int_0^t \mathrm{d}t'\left[K_{kj}^{(10)}(t-t')\hat{\eta}_j(t) + K_{kj}^{(11)}(t-t')\frac{\partial\hat{\eta}_j(t')}{\partial t'}\right] + \hat{f}_k^{(1)}(t).\end{aligned} \tag{29}$$

The result is a second-order differential equation which reflects the complete dynamics of the relevant observables.

6.1.4. Projection equations for correlation functions. An important role for investigations of slow-relaxation phenomena is played by the time-dependent equilibrium correlation functions of the relevant observables, which are in general defined by $\Phi_{kl}(t) = \langle\hat{\eta}_k(t)\tilde{\eta}_l(0)\rangle$. For facilitated kinetic Ising models like the SFM[2, d], the structure of these correlation functions can be characterized by

$$\Phi_{kl}(t) = \langle\hat{\eta}_k(t)\hat{\eta}_l(0)\rangle = \langle s|\,\hat{\eta}_k\exp\{\hat{L}t\}\hat{\eta}_l|F(0)\rangle. \tag{30}$$

Therefore, these correlation functions are equivalent to the normalized spin–spin correlation

$$\Phi_{kl}(t) = \frac{\langle\sigma_k(t)\sigma_l(0)\rangle - \bar{n}_{\mathrm{eq}}^2}{\bar{n}_{\mathrm{eq}}(1-\bar{n}_{\mathrm{eq}})}$$

which should be similar to the normalized density–density correlation of the underlying supercooled liquid, i.e. $\Phi_{kl}(t) \sim \langle \delta\rho(\boldsymbol{r}, t)\, \delta\rho(\boldsymbol{r}', 0)\rangle$. The evolution equation of $\Phi_{kl}(t)$ follows from (29) by a right-hand multiplication with $\hat{\eta}_l$ and a subsequent determination of the average. The contributions of the residual forces $\hat{f}_n^{(1)}(t)$ vanish identically. As a result, one obtains an exact second-order integro-differential equation [34]

$$\frac{\partial^2 \Phi_{kl}}{\partial t^2} = -\sum_j \left[\Omega_{kj}^{(10)} \Phi_{jl} + \Omega_{kj}^{(11)} \frac{\partial \Phi_{jl}}{\partial t} \right] + \int_0^t \mathrm{d}t' \left[K_{kj}^{(10)}(t-t') \Phi_{jl}(t') + K_{kj}^{(11)}(t-t') \frac{\partial \Phi_{jl}(t')}{\partial t} \right]. \tag{31}$$

As usual it is more convenient to transform the evolution equation (31) via a Fourier transform over space and time into an algebraic equation. In the present paper all further calculations are based on a hypercubic lattice structure with N sites, but application to another lattice type is always possible. In the following we assume that the correlation functions, frequency matrices, memory matrices are spatially homogeneous and isotropic, e.g. $\Phi_{nm}(t) = \Phi(|\boldsymbol{n} - \boldsymbol{m}|, t)$. The underlying lattice structure becomes irrelevant on sufficiently large spatial scales compared with the lattice constant and this can be mathematically expressed by the continuum limit, which corresponds to $|\boldsymbol{n} - \boldsymbol{m}| \to \infty$ or small wave-vectors $\boldsymbol{q}$ in the Fourier space. In this sense it is sufficient to perform all further analytical calculations within a continuum approximation. Finally, the Laplace transformation with respect to the time leads to the fundamental algebraic equation

$$\Phi(q, p) = \frac{\Phi_0(q)}{p + \dfrac{N\Omega^{(10)}(q) - NK^{(10)}(q, p) - pg_0(q)}{p + N\Omega^{(11)}(q) - NK^{(11)}(q, p) + g_0(q)}} \tag{32}$$

with the initial conditions $\Phi(q, 0) = \Phi_0(q)$ and $\dot{\Phi}(q, 0) = \dot{\Phi}_0(q)$ as well as the abbreviation $g_0(q) = \dot{\Phi}_0(q)/\Phi_0(q)$. In principle, the projection equations (32) are valid for any arbitrary physical system which can be described by irreversible master equations. Hence, the way to construct a mode-coupling approach, based on the projection equations, for facilitated kinetic Ising models is quite straightforward—by calculating explicitly the frequency matrices and memory matrices for the thermodynamic equilibrium. Here, it should be remarked that the following analytical calculations of the SFM[2, d] show that $\Omega^{(11)}(q) \neq 0$ and $K^{(10)}(q, p) \neq 0$, which is a consequence of the irreversible nature of the master equations. By way of contrast, the usual Mori–Zwanzig equations [33] are founded on reversible Liouville operators, leading immediately to $\Omega^{(11)}(q) = 0$ and $K^{(10)}(q, p) = 0$.

6.2. Determination of the memory matrices

6.2.1. Complete and orthogonal basis. In principle, under the assumption of a vanishing interaction between spins related to the Hamiltonian $H = \sum_i \varepsilon\sigma_i$, all operators acting on the Fock space can be represented by a complete set of orthogonal base operators. For facilitated kinetic Ising spin models the determination of such an operator-space basis is possible, by considering the underlying $\hat{d}_i, \hat{d}_i^{\dagger}$-(pseudo-fermionic) algebra (6). Now the base operators can be expressed as all possible products of the operators $\hat{\eta}_i$ introduced above (18). A base operator is denoted as $\hat{B}_{\boldsymbol{N}_n}^{(n)}$. The index n corresponds to the order of the product; $\boldsymbol{N}_n$ is an n-dimensional vector indicating the lattice sites concerned. Hence, the structure of the basis is given by

$$\begin{aligned}
\hat{B}^{(0)} &= 1 \\
\hat{B}_i^{(1)} &= \hat{\eta}_i \\
\hat{B}_{ij}^{(2)} &= \hat{\eta}_i \hat{\eta}_j \qquad \text{for } i < j \\
\hat{B}_{ijk}^{(3)} &= \hat{\eta}_i \hat{\eta}_j \hat{\eta}_k \qquad \text{for } i < j < k \\
&\vdots
\end{aligned}$$

Note that because of the commutation relation $[\hat{\eta}_i, \hat{\eta}_j] = 0$, the components of $\boldsymbol{N}_n$ can be ordered. The case of two or more equivalent indices is excluded because $\hat{\eta}_i^2 = (1 - 2\bar{n}_{\mathrm{eq}})/[\bar{n}_{\mathrm{eq}}(1 - \bar{n}_{\mathrm{eq}})]^{1/2}\hat{\eta}_i + 1$, i.e. quadratic or higher powers of each operator $\hat{\eta}_i$ can always be reduced to a linear representation. The base operators are orthogonal to each other, which means that

$$\left\langle \hat{B}_{N_n}^{(n)} \hat{B}_{N_m}^{(m)} \right\rangle = \delta^{nm} \delta_{N_n, N_m}. \tag{33}$$

This relation is based on the fact that each equilibrium average of operators on various lattice sites decomposes into a product of averages with respect to these sites, e.g. $\langle \hat{\eta}_i^2 \cdots \hat{\eta}_j \cdots \hat{\eta}_k^2 \cdots \rangle = \langle \hat{\eta}_i^2 \rangle \cdots \langle \hat{\eta}_j \rangle \cdots \langle \hat{\eta}_k^2 \rangle \cdots$. In general, this important relation is valid for all facilitated kinetic Ising spin models if the neighbour–neighbour interaction vanishes. Consequently, the orthogonality relation (33) follows immediately because of the fundamental features $\langle \hat{\eta}_i \rangle = 0$ and $\langle \hat{\eta}_i^2 \rangle = 1$. Thus, the basis $\tilde{B} = \{\hat{B}_{N_n}^{(n)}\}$ is orthogonal.

The completeness of $\tilde{B} = \{\hat{B}_{N_n}^{(n)}\}$ is to be understood in relation to the reference state $\langle s|$, i.e. the following equation is fulfilled for an arbitrary operator $\hat{X}$:

$$\langle s| \hat{X} = \sum_n \sum_{N_n} \left\langle \hat{X} \hat{B}_{N_n}^{(n)} \right\rangle \langle s| \hat{B}_{N_n}^{(n)}. \tag{34}$$

The mathematical proof of this property is given in [34].

6.2.2. Decomposition of the memory terms. Now, on the basis of the complete and orthogonal basis $\{\hat{B}_{N_n}^{(n)}\}$ we are able to decompose the memory matrix (27) which can be written as

$$K_{kj}^{(\gamma\beta)}(t) = \sum_{\alpha,i} \left\langle \hat{\eta}_k \hat{L}^{\gamma+1} \hat{Q} \exp\{\hat{Q}\hat{L}\hat{Q}t\} \hat{Q} \hat{L}^{\alpha+1} \hat{\eta}_i \right\rangle g_{ij}^{\alpha\beta} \tag{35}$$

for the SFM$[2, d]$ by using the relations (18) and (19) (note that $\hat{Q}^2 = \hat{Q}$). As is known from quantum mechanics, it is possible to rewrite (35) by inserting the completeness relation $\hat{1} = \sum_n \sum_{N_n} |\hat{B}_{N_n}^{(n)}\rangle\langle\hat{B}_{N_n}^{(n)}|$, which leads to

$$K_{kj}^{(\gamma\beta)}(t) = \sum_{\alpha,i} \sum_{n,m} \sum_{N_n, N_m} H_{k,N_n}^{\gamma(n)} \langle \hat{B}_{N_n}^{(n)} \exp\{\hat{Q}\hat{L}\hat{Q}t\} \hat{B}_{N_m}^{(m)} \rangle \tilde{H}_{N_m,i}^{(m)\alpha} g_{ij}^{\alpha\beta} \tag{36}$$

with the coefficients

$$H_{k,N_n}^{\gamma(n)} = \left\langle \hat{\eta}_k \hat{L}^{\gamma+1} \hat{Q} \hat{B}_{N_n}^{(n)} \right\rangle \qquad \text{and} \qquad \tilde{H}_{N_m,i}^{(m)\alpha} = \left\langle \hat{B}_{N_m}^{(m)} \hat{Q} \hat{L}^{\alpha+1} \hat{\eta}_i \right\rangle. \tag{37}$$

These coefficients can be determined exactly for the SFM$[2, d]$ by simple algebraic calculations. If one writes down explicitly the form of the coefficients (37), it is easy to see that $H_{k,N_n}^{\gamma(n)} = \tilde{H}_{N_m,i}^{(m)\alpha} = 0$ for $n = 0, 1$. Moreover, due to the choice of the relevant observables ($\gamma = 0, 1$) the coefficients (37) also vanish identically for the base operators with $n > 5$. Hence, non-vanishing contributions to the memory matrix (36) are determined by a finite number of base operators, where the leading term is given by $n = 2$. By inserting the

unit operator $\hat{1} = \exp\{-\hat{L}t\}\exp\{\hat{L}t\}$ and the completeness relation into the memory kernel $\langle \hat{B}^{(n)}_{N_n} \exp\{\hat{Q}\hat{L}\hat{Q}t\}\hat{B}^{(m)}_{N_m}\rangle$. One obtains

$$\langle \hat{B}^{(n)}_{N_n} \exp\{\hat{Q}\hat{L}\hat{Q}t\}\hat{B}^{(m)}_{N_m}\rangle = \sum_p \sum_{N_p} \langle \hat{B}^{(n)}_{N_n} \exp\{\hat{Q}\hat{L}\hat{Q}t\}\exp\{-\hat{L}t\}\hat{B}^{(p)}_{N_p}\rangle\langle \hat{B}^{(p)}_{N_p}(t)\hat{B}^{(m)}_{N_m}\rangle \tag{38}$$

where the averages $\langle \hat{B}^{(p)}_{N_p}(t)\hat{B}^{(m)}_{N_m}\rangle$ are usual many-point correlation functions, e.g. $\langle \hat{\eta}_i(t)\hat{\eta}_j(t)\hat{\eta}_k\hat{\eta}_l\rangle$ for $p = m = 2$.

6.2.3. Mode-coupling approximation. An exact determination of the memory kernel (38) is not possible at long times. Therefore, we need a suitable approximation [39] to make progress. The exact decomposition of the memory kernel in (38) can be interpreted as a separation of fast and slow timescales. By physical intuition, in the long-time limit the approximation strategy for the memory kernel will consist in the elimination of the fast modes as a suitable approximation of the correlation function.

First we analyse the function $\Psi^{(np)}_{N_nN_p}(t) = \langle \hat{B}^{(n)}_{N_n} \exp\{\hat{Q}\hat{L}\hat{Q}t\}\exp\{-\hat{L}t\}\hat{B}^{(p)}_{N_p}\rangle$ of the kernel (38). In general, $\hat{B}^{(n)}_{N_n}\exp\{\hat{Q}\hat{L}\hat{Q}t\}$ can be expected to show a significant evolution on a very short timescale in comparison to the characteristic timescale related to $\hat{B}^{(n)}_{N_n}\exp\{\hat{L}t\}$. Note that, while the evolution operator $\hat{L}$ contains all relevant timescales, the operator $\hat{Q}\hat{L}\hat{Q}$ is mainly determined by contributions related to short timescales [33]. Consequently, it can be assumed that at long times the time dependence of $\Psi^{(np)}_{N_nN_p}(t)$ is weak in comparison to the decay of the correlation function $\langle \hat{B}^{(p)}_{N_p}(t)\hat{B}^{(m)}_{N_m}\rangle$, which is related only to the time evolution factor $\exp\{\hat{L}t\}$. Hence, at long times we can expand $\Psi^{(np)}_{N_nN_p}(t)$ in powers of time t via a Taylor expansion

$$\Psi^{(np)}_{N_nN_p}(t) = \sum_{M=0}^{\infty} \Lambda^{(np),M}_{N_nN_p}\frac{t^M}{M!}$$

with

$$\Lambda^{(np),M}_{N_nN_p} = \left\langle \hat{B}^{(n)}_{N_n}\frac{\partial^M}{\partial t^M}\big[\exp\{\hat{Q}\hat{L}\hat{Q}t\}\exp\{-\hat{L}t\}\big]_{t=0}\hat{B}^{(p)}_{N_p}\right\rangle.$$

The first coefficient $\Lambda^{(np),M}_{N_nN_p}$ can be determined by simple calculations

$$\Lambda^{(np),0}_{N_nN_p} = \langle \hat{B}^{(n)}_{N_n}\hat{B}^{(p)}_{N_p}\rangle = \delta^{np}\delta_{N_n,N_p}.$$

So far, we have restricted our investigation to the simplest case, i.e. we assume $\Lambda^{(np),M}_{N_nN_p} = 0$ for $M \geqslant 1$. It should be remarked that an extension to higher-order terms is possible without any problems. We omit consideration of higher-order terms only for clarity of the calculations. Furthermore, the results obtained already show a reasonable agreement with numerical simulations.

Clearly, the main problem consists in a reasonable approximation of the many-point correlation function $\langle \hat{B}^{(p)}_{N_p}(t)\hat{B}^{(m)}_{N_m}\rangle$. This function decays in products of simple pair correlation functions if the distances between the corresponding lattice points (defined by the vectors $\boldsymbol{N}_p$ and $\boldsymbol{N}_m$) are sufficiently large:

$$\langle \hat{B}^{(p)}_{i_1i_2\ldots i_p}(t)\hat{B}^{(m)}_{j_1j_2\ldots j_p}\rangle \simeq \frac{1}{p!}(\Phi_{i_1j_1}(t)\Phi_{i_2j_2}(t)\cdots\Phi_{i_pj_p}(t) + \text{permutations}). \tag{39}$$

This asymptotic limit is correct for infinitely large (or at least sufficiently large) distances between the lattice sites $i_1, i_2, \ldots$. We use this limit case as an approximation for an arbitrary set of lattice sites $\{\boldsymbol{N}_p, \boldsymbol{N}_m\}$. This approximation is equivalent to the decomposition of higher

static correlation functions into simple pair correlation functions. A similar approach has been used, for example, to create self-consistent equations for the static structure factor [40].

As mentioned above, the base operators with $2 \leqslant n \leqslant 5$ lead only to non-vanishing coefficients (37) and thus to non-zero terms for the memory matrix (36). The leading term or the main contribution is given by $n = 2$, related to the base operator $\hat{B}^{(2)}_{ij} = \hat{\eta}_i \hat{\eta}_j$. The base operators $\hat{B}^{(n)}_{N_n}$ with $3 \leqslant n \leqslant 5$, on the other hand, yield only small additional contributions, which can be neglected. In view of this, we now restrict the basis $\tilde{B} = \{\hat{B}^{(n)}_{N_n}\}$ to operator elements with $n \leqslant 2$. Thus, the memory matrix (36) can be approximated by

$$K^{(\gamma\beta)}_{kj}(t) \approx \frac{1}{2} \sum_{\alpha,i} \sum_{i_1 i_2 j_1 j_2} H^{\gamma(2)}_{k,i_1 i_2} \left[\Phi_{i_1 j_1}(t) \Phi_{i_2 j_2}(t) + \Phi_{i_1 j_2}(t) \Phi_{i_2 j_1}(t) \right] \tilde{H}^{(2)\alpha}_{j_1 j_2, i} g^{\alpha\beta}_{ij} \tag{40}$$

at long times.

According to the classification scheme of Hohenberg and Halperin [41], the SFM[2, d] is a so-called model A and therefore the frequency and memory matrices do not vanish for $q \to 0$. In this sense, it is a suitable simplification to restrict the long-time analysis for the non-conserved irreversible dynamics of the SFM[2, d] to the limit $q \to 0$. Then the suppression of the wave-vector dependence leads to $\Phi_{nm}(t) = \varphi(t)\delta_{nm} = (1/N) \sum_{\boldsymbol{q}} \varphi(t) \exp\{\mathrm{i}\boldsymbol{q}(\boldsymbol{n} - \boldsymbol{m})\}$. On using the relation $g_0(q) = g_0(0) = -\Gamma^1_0/\tau_0$ with $\Gamma^1_0 = 2z(z-1)[\bar{n}_{\mathrm{eq}}(1-\bar{n}_{\mathrm{eq}})]^{1/2}$ (see [34]), the algebraic evolution equation (32) can be written as

$$\varphi(p) = \left[p + \frac{\Gamma^1_0}{\tau_0} - \frac{1}{\tau_0} \frac{\Gamma^1_0 N \Omega^{(11)}(0)\tau_0 - N\Omega^{(10)}(0)\tau_0^2 - (\Gamma^1_0)^2}{p\tau_0 + N\Omega^{(11)}(0)\tau_0 - \lambda \Xi(p) - \Gamma^1_0} \right]^{-1} \tag{41}$$

with the initial condition $\varphi(0) = 1$ and the Laplace transform

$$\Xi(p) = \int_0^\infty (\mathrm{d}t/\tau_0)\ \varphi^2(t) \exp\{-pt\} \qquad \mathrm{Im}\ z > 0.$$

6.3. Ergodicity and non-exponential decay

Now, using the algebraic mode-coupling equation (41) we are able to analyse the characteristic slowing down of the dynamics of the SFM[2, d] for decreasing temperature. Following the philosophy of the investigations of mode-coupling theories [36, 38, 42], the first question that arises concerns the existence of ergodicity and non-ergodicity: is there a critical temperature $T^\star$ such that the correlation function $\varphi(t)$ shows an incomplete decay $\varphi(t \to \infty) = f_\infty \neq 0$ for $T \leqslant T^\star$? Equivalently, does the function $\varphi(p)$ have a pole at $p = 0$ for $T \leqslant T^\star$? This question is analogous to that of the determination of a kinetic phase transition from an ergodic state into a non-ergodic state for supercooled liquids. To answer it, we split the correlation function into a non-ergodic part f_∞ and a contribution $\varphi_{\mathrm{erg}}(t)$ via the ansatz $\varphi(t) = f_\infty + \varphi_{\mathrm{erg}}(t)$. Thus, the function $\varphi_{\mathrm{erg}}(t)$ describes the remaining ergodic part of the SFM[2, d], i.e. $\varphi_{\mathrm{erg}}(t \to \infty) = 0$. The Laplace transformation leads to

$$\varphi(p) = \frac{f_\infty}{p} + \varphi_{\mathrm{erg}}(p) \tag{42}$$

with $\lim_{p\to 0} p\varphi_{\mathrm{erg}}(p) = 0$. Furthermore, the term $\Xi(p)$ can expressed as

$$\Xi(p) = \frac{f^2_\infty}{p\tau_0} + \Xi_{\mathrm{erg}}(p) \tag{43}$$

with $\lim_{p\to 0} p\,\Xi_{\mathrm{erg}}(p) = 0$. Using the ansatz (42) and (43), the evolution equation (41) can be written as

$$f_\infty = \lim_{p\to 0} p\varphi(p) = \lim_{p\to 0} \left[1 + \frac{\Gamma^1_0}{p\tau_0} + \frac{\Gamma^1_0 N \Omega^{(11)}(0)\tau_0 - N\Omega^{(10)}(0)\tau_0^2 - (\Gamma^1_0)^2}{\lambda f^2_\infty} \right]^{-1}. \tag{44}$$

One sees immediately that the non-ergodicity part f_∞ has a non-vanishing value only if $\Gamma_0^1 = 0$. Otherwise, the only solution of (44) is given by $f_\infty = 0$, i.e. the SFM[2, d] is an ergodic system if $\Gamma_0^1 \neq 0$. Clearly, the value of Γ_0^1 vanishes [34] only for $T = 0$:

$$\Gamma_0^1 = 0 \qquad \Longleftrightarrow \qquad T = 0.$$

Thus, the analytical calculations based on a mode-coupling approach exhibit a non-ergodic state of the SFM[2, d] only at zero temperature. Additionally, we obtain that the frequency term $\Omega^{(10)}(0)$ vanishes for $T = 0$. Hence, the non-ergodic part is given by

$$f_\infty = 1$$

i.e. an initial equilibrium configuration at $T = 0$ shows no structural relaxations throughout the total observation time. Consequently, in the language of ergodic–non-ergodic transitions the SFM[2, d] realizes a kinetic phase transition from an ergodic state to a non-ergodic state at the critical temperature $T^\star = 0$. In other words, each arbitrary equilibrium configuration is frozen at $T = T^\star = 0$.

The present analytical calculation exhibits the important result that the SFM[2, d] is ergodic for all finite temperatures $T > 0$, which is in complete agreement with previous numerical results [6–8, 43]. As expected, a mode-coupling approach seems to be the right formalism for the investigation of slow-relaxation dynamics of the SFM[2, d], in contrast to a renormalized perturbation theory [2, 3] which predicted an incorrect kinetic phase transition at a finite critical temperature.

In the next step, we analyse the slow-relaxation behaviour near the critical temperature $T^\star$, i.e. at finite temperatures $T > 0$. To this end, we introduce the relaxation time $\tau_c = \varphi(0)$. On the basis of equation (41), τ_c is given by

$$\tau_c = \tau_0 \frac{N\Omega^{(11)}(0)\tau_0 - \lambda\Xi(0) - \Gamma_0^1}{N\Omega^{(10)}(0)\tau_0^2 - \lambda\Gamma_0^1\Xi(0)}. \tag{45}$$

Using this notation, the algebraic equation (41) can be written as

$$\varphi(p) = \left[p + \tau_c^{-1} + \left(\frac{\Gamma_0^1}{\tau_0} - \tau_c^{-1} \right) \left[1 + \frac{\varsigma}{p\tau_0 + \lambda\,[\Xi(0) - \Xi(p)]} \right]^{-1} \right]^{-1} \tag{46}$$

with the coefficient

$$\varsigma = N\Omega^{(11)}(0)\tau_0 - \lambda\Xi(0) - \Gamma_0^1. \tag{47}$$

Therefore, equations (45)–(47) are a closed, non-linear system of equations, which can be solved by standard numerical methods. We obtain that the correlation function $\varphi(t)$ shows with decreasing temperature a pronounced stretched decay over some decades, while an exponential-like decay is obtained for high temperatures [34]. This stretching can be illustrated by a simple argument. Short times ($t \to 0$ or $p \to \infty$) are related to a behaviour $\varphi(p) \simeq (p + \Gamma_0^1/\tau_0)^{-1}$ or $\varphi(t) \simeq \exp\{-\Gamma_0^1 t/\tau_0\}$. On the other hand, the long-time regime ($t \to \infty$ or $p \to 0$) is characterized by $\varphi(p) \simeq (p + \tau_c^{-1})^{-1}$ or $\varphi(t) \simeq \exp\{-t/\tau_c\}$. Because $\Gamma_0^1/\tau_0 \gg \tau_c^{-1}$, we expect a typical crossover between the two regimes characterized by a stretched decay. Therefore, the present analysis of SFM[2, d] shows a typical stretched-exponential decay of the autocorrelation function $\varphi(t)$ for temperature decreasing towards $T^\star$, which depends in a natural way on T.

7. Conclusions

In the present paper we have introduced various theoretical and numerical approaches to the slow relaxation and cooperative rearrangements in the SFM[f, d]. We have demonstrated that

spin-facilitated kinetic Ising models show the essential characteristics of supercooled liquids. From this point of view, and from the fact that the spin-lattice structure allows the application of various theoretical techniques, the SFM[f, d] is an outstanding candidate for the description of cooperative processes.

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