Fast Kawasaki spin exchange limit of spin-facilitated kinetic Ising models

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We study a Fock-space operator technique for describing the stochastic kinetics of a spin-facilitated kinetic Ising model. We focus in particular on the diffusion (fast Kawasaki exchange) limit in which the kinetics can be described by a single mean field evolution equation. We derive some general criteria for the approximative validity of mean field theory for the case of a nondiverging diffusion coefficient of the local spin states.

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A central topic in statistical physics is the qualitative and quantitative understanding of long-time phenomena in strongly interacting many-body systems. They exhibit an extreme slowing down without long range ordered states or other singular behavior of spatial quantities in contrast to the critical slowing down of conventional phase transitions. Such phenomena are characterized by a high cooperativity of local processes. For a large class of dense systems some essential properties of the slowing-down regime, e.g., a nonexponential decay of autocorrelation functions at sufficiently long times or a non-Arrhenius temperature dependence of relaxation times, seem to be universal or are at least a characteristic feature. For instance, such scenarios have been observed for the main glass transition process in supercooled liquids.

In this connection, facilitated kinetic spin systems belong to an interesting class of nonequilibrium models, which show a typical slowing down of the dynamics caused by an increasing cooperativity of local spin-flip processes with decreasing temperature. In this context our interest in the present paper is especially focused on studying the *f*-spin-facilitated kinetic Ising models [1–3], 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 underlying dynamic of the *f*-spin-facilitated kinetic Ising models is a stochastic one, which is given by a simple Glauber process [4], i.e., a spin flip $\sigma_i=+1\leftrightarrow\sigma_i=-1$ is controlled by the thermodynamic Gibbs measure and by self-induced topological restrictions. In particular, the topological conditions are explicitly taken into account so that an elementary flip at a given lattice point *i* is only allowed if the number of the nearest neighbored lattice points in the spin up state ($\sigma_i=+1$) is equal to or larger than a restriction number *f* with $0<f<z$ (*z* is the coordination number of the lattice). Such models [1–3,5] are denoted as *f*-spin-facilitated Ising models on a *d*-dimensional lattice, shortened to $S[f,d]$. In the original model, Ising spins on different lattice sites are coupled only via kinetic constraints, i.e., there are no pair interactions between neighbor spins. In this way, elementary single spin-flip processes are connected with geometrical restrictions which lead inevitably to cooperative rearrangements in the underlying spin system.

Identifying the up state as a molecule of component *A* and the down state as a molecule of type *B*, the $S[f,d]$ can be interpreted as a reversible autocatalytic chemical reaction $fA+B \rightleftharpoons (f+1)A$ on a *d*-dimensional lattice. A particle changes its constitution from *A* to *B* and *B* to *A* only in the presence of at least *fA* molecules in the nearest environment. Because of the slow dynamics of the $S[f,d]$, it can be used to model a glasslike behavior in a simple manner [6–9].

In this paper we study the influence of an additional diffusionlike nonactivated Kawasaki spin exchange between nearest neighbors. Such a process corresponds to diffusion with a certain diffusion coefficient D_{kaw} in the above introduced chemical picture $A+B \rightleftharpoons B+A$. The additional diffusion leads generally to an acceleration of the dynamics. It is well known that a sufficiently large diffusion coefficient allows the application of the kinetic theory for chemical processes. On the other hand, this classical theory of reaction-diffusion systems frequently breaks down in low dimensions and also for small diffusion coefficients. The aim of this paper is a discussion of the dynamical regimes that may be expected for our modified $S[f,d]$.

For our analytical treatment we use the Fock-space formalism, which is a very powerful method for analyzing classical many-body systems with a stochastic dynamic given by a master equation on a lattice. The Fock-space approach is based on a quantumlike formulation of the underlying master equation written in terms of creation and annihilation operators. The Fock-space representation of the modified $S[f,d]$ will be more transparent, interpreting the two spin orientations $\sigma_i=-1$ and $\sigma_i=1$ as empty and single occupied sites corresponding to the restricted occupation numbers $n_i=0$ and $n_i=1$ via $\sigma_i=2n_i-1$, respectively. The $S[f,d]$ can then be interpreted as a lattice gas with an excluded volume effect, i.e., changes of the configuration $\mathbf{n}=\{n_i\}$ are possible only under the presence of the exclusion principle. Following Refs. [10–15], the probability distribution $P(\mathbf{n},t)$ can be related generally to a state vector $|F(t)\rangle$ in a Fock space according to $P(\mathbf{n},t)=\langle n|F(t)\rangle$ and $|F(t)\rangle=\Sigma_{\mathbf{n}}P(\mathbf{n},t)|n\rangle$, respectively, where the base vectors $|\mathbf{n}\rangle$ are composed of second quantized operators. Using this representation, the time evolution in terms of a master equation can be trans-

formed to an equivalent evolution equation in a Fock-space

$$\partial_t|F(t)\rangle = L|F(t)\rangle. \tag{1}$$

The dynamical matrix $L(\mathbf{n},n')$ of the master equation is mapped onto the operator $L=L(a,a^\dagger)$, which is given in a second quantized form with a and $a^\dagger$ being the annihilation and creation operators, respectively. Originally, this transformation was applied for the Bose case with unrestricted occupation numbers [10–12]. Here, we consider the case of restricted occupation numbers [13–15]. In order to preserve the restriction of the occupation number in the underlying dynamical equations, the commutation rules of the operators a and $a^\dagger$ are chosen as those of Pauli operators [13,16,17] with the commutation rules

$$[a_i,a_j^\dagger]=\delta_{ij}(1-2a_i^\dagger a_i), \quad [a_i,a_j]=[a_i^\dagger,a_i^\dagger]=0,$$
$$a_i^2=(a_i^\dagger)^2=0. \tag{2}$$

The master equation of the original $S[f,d]$ without Kawasaki spin exchange can be expressed by the following evolution operator [18]:

$$L_{\mathrm{SFM}}=\sum_i \left\{ \frac{1}{f!} \sum_{k_1,\ldots,k_f} \kappa_{i|k_1\cdots k_f} N_{k_1}N_{k_2}\cdots N_{k_f} \right\} \times[\beta(a_i-N_i)+\lambda(a_i^\dagger-(1-N_i))] \tag{3}$$

with the particle number operator $N_i=a_i^\dagger a_i$ and temperature dependent jump rates λ for the flip $\downarrow\rightarrow\uparrow$ and $\beta>\lambda$ for the inverse process. The temperature is defined by the ratio of the jump rates, $T\sim[\ln(\beta/\lambda)]^{-1}$. The inner terms of the square brackets in Eq. (3) represent a single spin-flip process on the lattice site i and the product of the particle number operators $N_{k_1}N_{k_2}\cdots N_{k_f}$ considers N_jN_k local constraints. Additionally, $\kappa_{i|k_1\cdots k_f}$ is a lattice function with $\kappa_{i|k_1\cdots k_f}=1$ if $k_1\neq k_2\neq\cdots\neq k_f$ and all k_α $(\alpha=1,\ldots,f)$ are neighbors to lattice site i, and $\kappa_{i|k_1\cdots k_f}=0$ otherwise.

Consideration of the Kawasaki spin exchange process requires an additional contribution to the evolution operator (3):

$$L_{\mathrm{kaw}}=\frac{1}{2}D_{\mathrm{kaw}}\sum_{i,j}\Theta_{ij}[a_i^\dagger a_j+a_j^\dagger a_i-(1-N_i)N_j -(1-N_j)N_i] \tag{4}$$

with the lattice function $\Theta_{ij}=1$ if i and j are neighboring lattice sites and $\Theta_{ij}=0$ otherwise. In principle, as was shown first by Doi [10], the average of an arbitrary physical quantity $B(\mathbf{n})$ is given by the average of the corresponding operator $B(t)=\Sigma_{\mathbf{n}}|\mathbf{n}\rangle B(\mathbf{n})\langle n|$ via [19]

$$\langle B(t)\rangle=\sum_{\mathbf{n}} P(\mathbf{n},\mathbf{t})\mathbf{B}(n)=\langle s|\mathbf{B}|\mathbf{F}(\mathbf{t})\rangle \tag{5}$$

using the reference state $\langle s|=\Sigma_{\mathbf{n}}\langle\mathbf{n}|$. The normalization condition is manifest in $\langle s|F(t)\rangle=1$ with the consequence [19] that the evolution operator always fulfills the necessary relation $\langle s|L=0$. Because of Eqs. (1) and (5) the evolution equation for an arbitrary operator $B(t)$, for example the particle number operator, is given by [19]

$$\partial_t\langle B\rangle=\langle s|[B,L]|F(t)\rangle, \tag{6}$$

which can be extended immediately in order to write down the kinetic equations for time-dependent correlation functions. As a general result of the procedure, all the dynamical equations describing the classical problem are completely determined by the commutation rules of the underlying operators and the structure of the evolution operator L. Thus, this method allows investigations of master equations for various evolution processes, e.g., aggregation, chemical reactions [14,15], nonlinear diffusion [20], as well as the spin-facilitated kinetic Ising models. Note that the decisive advantage of the Fock-space approach is given by the simple construction principles for each evolution operator L on the basis of creation and annihilation operators [21].

Using Eq. (6) with the Liouville operator $L=L_{\mathrm{S}}+L_{\mathrm{K}}$, the evolution equation for the averaged particle number operator reads

$$\partial_t\langle N_i\rangle=\lambda(\langle E_i[N]\rangle-\langle E_i[N]N_i\rangle)-\beta\langle E_i[N]N_i\rangle +D_{\mathrm{K}}\sum_j \Theta_{ij}[\langle N_j\rangle-\langle N_i\rangle] \tag{7}$$

with

$$E_i[N]=\frac{1}{f!}\sum_{k_1,\ldots,k_f}\kappa_{i|k_1\cdots k_f}N_{k_1}N_{k_2}\cdots N_{k_f}. \tag{8}$$

Unfortunately, this equation contains higher moments of spins at neighbor lattice sites. The evolution equation of these moments contains the next higher correlations, i.e., one obtains an infinitely large hierarchy of evolution equations. But in the case of a diverging diffusion coefficient $D_{\mathrm{K}}\rightarrow\infty$, the strong mixing of up and down spins results [22] in the decoupling of higher correlations $\langle N_iN_j\rangle\rightarrow\langle N_i\rangle\langle N_j\rangle$, and a homogenization of the local spin distributions $\langle N_i\rangle\rightarrow\overline{N}$. Thus, the homogeneous magnetization $\overline{\sigma}=2\overline{N}-1$ satisfies the mean field equation

$$\frac{\partial\overline{\sigma}}{\partial t}=\binom{z}{f}\left(\frac{\overline{\sigma}+1}{2}\right)^f[\lambda(1-\overline{\sigma})-\beta(1+\overline{\sigma})] \tag{9}$$

with z the coordination number of the lattice. The equilibrium state is given by $\overline{\sigma}_{\mathrm{eq}}=(\lambda-\beta)/(\beta+\lambda)$. A small fluctuation $\delta\overline{\sigma}=\overline{\sigma}-\overline{\sigma}_{\mathrm{eq}}$ approaches equilibrium exponentially $\delta\overline{\sigma}\simeq\exp(-t/\tau)$ with the relaxation time

$$\tau=\binom{z}{f}^{-1}\frac{(\lambda+\beta)^{f-1}}{\lambda^f} \tag{10}$$

whereas large fluctuations relax with an algebraic inverse power law in t. In particular, the low temperature limit $\lambda/\beta\rightarrow 0$ is defined by a purely algebraic decay

$$\overline{\sigma}+1=2\left[f\beta\binom{z}{f}t\right]^{-1/f}\sim t^{-1/f} \tag{11}$$

which remains after the passing of the initial regime.

The decoupling of the higher moments to break up the infinite hierarchy of evolution equations holds for large diffusion coefficients D_K. This decoupling procedure may be reasonable though to describe small fluctuations from equilibrium, because the Hamiltonian of both the original and the modified $S[f,d]$ is that of a simple paramagnetic gas. Thus, the decoupling remains valid at equilibrium and we have to deal with $\langle N_i N_j\rangle=\overline{N}^2+\overline{N}(1-\overline{N})\delta_{ij}$. Similar arguments can be used at sufficiently high temperatures, i.e., for $\lambda\approx\beta$. However, far from equilibrium and at sufficiently low temperatures we should use another concept. Let us find a criterion for the application of the mean field equation (9) to the modified $S[f,d]$. The dynamics of this model can be mapped onto the autocatalytic picture introduced above. Here, the relaxation into the equilibrium is mainly determined by diffusion processes $A+B\rightleftharpoons B+A$ and the annihilation and creation processes $(f+1)A\rightleftharpoons fA+B$. To determine the survival rate of A particles within a mean field approximation, note that in a lifetime t_L each A particle will succesfully encounter a site with at least f neighbor sites occupied by A particles. Consequently, in a time $\Delta t\sim t_L$ the concentration decrement of a large fluctuation $\Delta\overline{N}$ will be of order $\overline{N}$. Using this gives the mean field rate equation

$$\frac{\partial\overline{N}}{\partial t}\approx\frac{\Delta\overline{N}}{\Delta t}\sim-\frac{\overline{N}}{t_L}\sim-(\beta-\lambda)\overline{N}^{1+f} \tag{12}$$

and we obtain $t_L\sim(\beta-\lambda)^{-1}\overline{N}^{-f}$. During this time, an A particle has visited a volume V_L. On the other hand, the mean field approach is valid, if each particle visits a lot of possible reaction centers (lattice sites with f A particles in their nearest environment) before the autocatalytic reaction takes place. The concentration of such centers is $\overline{N}^f$. Thus, the condition for an application of the mean field approach reads $V_L\overline{N}^f\gg1$. If ξ^2 is the mean square displacement of an A particle within its lifetime, $\xi^2\sim D_K t_L$, the visited volume is given by $V_L\sim\xi^d$ for $d<2$, $V_L\sim\xi^2/\ln\xi$ for $d=2$, and $V_L\sim D_K t_L$ for $d>2$. These relations follow from the fact that the trajectory of a random walker is compact for $d\leqslant2$, i.e., the particle visits almost all sites inside a sphere of radius ξ. For $d>2$, diffusion trajectories are noncompact and the visited volume is proportional to the length of the path, i.e., $V_L\sim D_K t$. Hence, for $d<2$ the mean field approch is valid if

$$D_K t_L\sim D_K(\beta-\lambda)^{-1}\overline{N}^{-f}\gg\xi^2\sim\overline{N}^{-2f/d}, \tag{13}$$

i.e., the condition $D_K(\beta-\lambda)\gg\overline{N}^{f(1-2/d)}$ must be satisfied if a mean field approach is to make sense. For $d>2$, we obtain independently from the concentration the result $D_K(\beta-\lambda)\gg1$ while for $d=2$ the condition $D_K(\beta-\lambda)\gg-f\ln\overline{N}$ has to be taken into account. The failure of the mean field theory for low concentrations and $d\leqslant2$ follows from the fact that the $f+1$ A particles involved in one possible elementary reaction have very many contacts among each other before a contact with another A particle occurs. However, the mean field concept requires a lot of contacts with different A particles. Finally, the limit of high concentrations $\overline{N}\to1$ leads independently of the dimension to the condition $D_K(\beta-\lambda)\gg1$.

It should be remarked that for slow spin exchange processes D_K has to be replaced by D_{eff} considering that combinations of annihilation and creation processes contribute also to an effective diffusion in the autocatalytic particle picture. This thermally activated effect is of the order of magnitude of the rate λ and can be neglected in the previous scaling arguments. The inequality becomes independent of the number $\overline{N}$ above the critical dimension and mean field approaches are successful for $D_K\gg\beta$. Therefore, the relaxation behavior is then characterized by an algebraic decay far from equilibrium, whereas an exponential decay dominates the small fluctuations from the equilibrium state. For $D_K\sim\beta$ we observe a typical slowing down of the dynamics which may be explained by other approaches, for example mode coupling techniques [23] or field theoretical perturbation theories [24].

In particular, for $f=1$, scaling arguments can be used also for the derivation of the correct decay outside the mean field regime. The $S[1,d]$ corresponds to the biparticle reactions $A+A\rightleftharpoons A+B$ and diffusion steps $A+B\rightleftharpoons B+A$. Equilibrium will be reached for temperature $T>0$ independently of the diffusion coefficient D_K. For $T=0$ and $D_K=0$ the dynamics stops at a nonergodic phase while for $D_K>0$ the $S[1,d]$ relaxes into the ordered equilibrium state defined by $\overline{\sigma}=-1$. The approach into the nonergodic state can be demonstrated by the rigorous result [18]; in this connection see also [25] for $d=1$. For instance, the decay of the ordered phase $\overline{\sigma}=1$ is given by $\overline{\sigma}(t)=2\exp\{\exp(-\beta t)-1\}-1$. In the long-time limit this results in a pronounced slowing down, leading to a nonergodic behavior which is manifested in that for $t\to\infty$ the quantity $\overline{\sigma}(t)$ remains finite with $\overline{\sigma}(\infty)=2e^{-1}-1$.

The behavior of the modified $S[1,d]$ with $D_K>0$ and for $T=0$ is similar to a single species coalescence model corresponding to a reversible diffusion $A+B\rightleftharpoons B+A$ and an irreversible annihilation $A+A\to A+B$ with active particles A and passive particles B. For high concentrations of A, i.e., for large $\overline{N}$ a classical mean field regime may exist if $D_K/\beta\gg\overline{N}^{-1}$. As discussed above, after the passing of the inital regime, the decay is given by $\overline{N}\sim t^{-1}$. This reaction controlled regime holds for intermediate times as long as $t\ll D_K/\beta$. For longer times the system undergoes a crossover to the diffusion controlled regime. The decay of this regime can be obtained also from scaling arguments. Because of the recurrence of random walks for $d\leqslant2$ the number m of visited lattice sites is given by $m\sim t^{d/2}$ for $d<2$ and $m\sim t$ for $d>2$. At the critical dimension $d=2$ one obtains a logarithmic correction $m\sim t/\ln t$. On the other hand, each active particle occupies a volume $V\sim\overline{N}^{-1}$. Thus, a reaction between

two active particles A occurs if $V \sim m$ and we obtain $\overline{N} \sim t^{-d/2}$ for $d<2$ and $\overline{N} \sim \ln t/t$ for $d=2$. Above the critical dimension we obtain $\overline{N} \sim t^{-1}$.

Numerical simulations within the $S[1,1]$ starting from the ordered spin up phase support these arguments; see Fig. 1. The initial regime can be fitted very well by the above mentioned solution [18]. For a slow spin exchange, the pronounced plateau $\overline{N}=e^{-1}$ indicates a metastable nonergodic state. This nonergodic phase decays due to the spin exchange dynamics. The asymptotic decay is determined by the expected $t^{-1/2}$ behavior. A classical regime with $\overline{N}=t^{-1}$ can be observed as an intermediate regime beween the initial decay and the $t^{-1/2}$ law only for fast Kawasaki processes.

In the case of finite but small temperatures we obtain a similar behavior. But here, the asymptotic behavior of the decay $\overline{N}(t)$ is determined by an exponential approach to the equilibrium state. This behavior gradually dominates the other regimes with increasing temperature.

We have pointed out the remarkable simplification in the description of lattice reaction-diffusion processes in the limit of an infinitely large diffusion coefficient. In effect the infinite hierarchy of evolution equations for the coupled occupation number correlations can be replaced by a single mean field evolution equation. We have confirmed this by numerical simulations and given criteria for the approximate applicability in limiting cases of the mean field theory for large but finite diffusion coefficient.

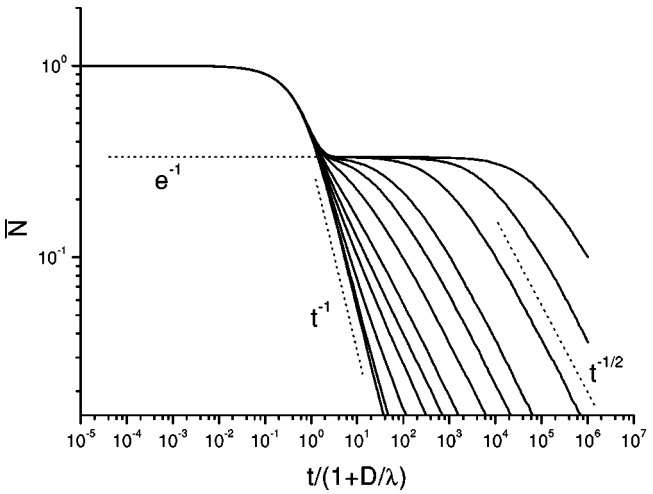

FIG. 1. Mean particle number $\overline{N}$ versus $t/(1+D\lambda^{-1})$ for $T=0$ and for different $D=D_K$. For a rapid Kawasaki exchange of spins the decay follows a t^{-1} law. For smaller diffusion coefficient the decay follows the $t^{-1/2}$ law. As one goes from left to right in this figure the diffusion coefficient decreases.

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