

Monte Carlo simulations of a generalized n -spin facilitated kinetic Ising model

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A kinetic Ising model is analyzed where spin variables correspond to lattice cells with mobile or immobile particles. Introducing additional restrictions for the flip processes according to the n spin-facilitated kinetic Ising model and using Monte Carlo methods, we study the freezing process under the influence of an additional nearest-neighbor interaction. The stretched exponential decay of the auto correlation function is observed and the exponent γ , as well as the relaxation time are determined depending on the activation energy h and the short-range coupling J . The magnetization corresponding to the density of immobile particles is found to be the controlling parameter for the dynamic evolution. [S0163-1829(99)08709-3]

I. INTRODUCTION

There is a continuous effort in describing of supercooled liquids using different approaches.¹⁻⁴ However the phenomenon is generally not complete understood. Supercooled fluids normally reveal a stretched exponential decay of typical (e.g., density-density) correlation functions and a non-Arrhenius behavior of the associated relaxation times. This slowing down in the dynamical behavior can be illustrated by a strongly curved trajectory in the Arrhenius plot (relaxation time τ versus the inverse temperature T^{-1}), empirically described by the well-known Williams-Landel-Ferry (WLF) relation.⁵ But in contrast to conventional phase transitions a long-range order is not developed.

The characteristic slowing down of the dynamics is usually explained by an increasing cooperativity of local processes with decreasing temperature.⁶ This behavior is an universal phenomena of the glass transition.⁷

Mode coupling theories^{1,8,9} (MCT) predict the existence of an ergodic behavior above a critical temperature T_c and a nonergodic behavior below T_c . Note that T_c is in the range between the melting temperature T_m and the glass temperature T_g , i.e., $T_m > T_c > T_g$. At T_c the system undergoes a sharp transition from an ergodic state to a state with partially frozen (density) fluctuations. The slow α process within the MCT is thought to correspond to the actual dynamic glass transition whereas the fast β process is often identified with a cage rattling or the boson peak.

Actually, the nonergodic state obtained from the original MCT below T_c are approximately stable only for a finite-time interval. Strongly cooperative processes lead to a slow decay of apparently frozen structures. This slow decay shows the typical above mentioned properties corresponding to the dynamics of the main glass transition (WLF-like behavior of the relaxation time, stretched exponential decay of the correlation function). This effect can be partially described in terms of an extended mode coupling theory^{10,11} introducing additional hopping processes.

There exist also various alternative descriptions,^{3,12} which explain the cooperative motion of the particles inside a supercooled liquid below T_c . One of these possibilities is the spin-facilitated Ising model,¹²⁻¹⁵ originally introduced by Fredrickson and Andersen. The basic idea of these models

consists of a coarse graining of space and time scales and simultaneously a reduction of the degrees of freedom. In detail that means:

(1) *Coarse graining of spatial scales.* The supercooled liquid is divided into cells in such a way that each cell contains a sufficiently large number of particles, which realize a representative number of molecular motions.

(2) *Reduction of the degrees of freedom.* Each cell will be characterized by only one degree of freedom, i.e., the cell structure enables us to attach to each cell an observable s_j (usually denoted as spin), which characterizes the actual dynamic state of particles inside the cell j . The usual realization is given by the local density ρ_j (particles per cell) with $s_j = 1$ if $\rho_j > \bar{\rho}$ and $s_j = -1$ if $\rho_j < \bar{\rho}$, where $\bar{\rho}$ is the averaged density of the system. This mapping implies consequently different mobilities of the particles inside such a cell, i.e., $s_j = 1$ corresponds to the immobile solidlike state and $s_j = -1$ to the mobile state of cell j . The set of all spin observables forms a configuration, the time expansion of the corresponding probability distribution obeys a master equation.

(3) *Coarse graining of the time scale.* This step is based on the assumption that fast processes (e.g., the β process) are well separated from the slow α process. Hence, the original Liouville equation of the supercooled liquid can be projected onto a simple master equation without any memory terms. Therefore, the spin-facilitated kinetic Ising model is suitable for a description of a supercooled liquid well below T_c within the MCT and for sufficiently large time scales.

To make the time evolution of the glass configurations more transparent we use the argumentation following the idea of Fredrickson and Andersen,¹²⁻¹⁵ i.e., we suppose that the basic dynamics is a simple process $s_j = +1 \leftrightarrow s_j = -1$ controlled by the thermodynamical Gibbs measure and by self-induced topological restrictions. In particular, an elementary flip at a given cell is allowed only if the number of the nearest neighbored mobile cells ($s_j = -1$) is equal to or larger than a restriction number f with $0 < f < z$ (z : coordination number). So, elementary flip processes and geometrical restrictions lead to the cooperative rearrangement of the underlying system and therefore to a mesoscopic model describing a supercooled liquid below T_c . Such models¹²⁻¹⁵ are denoted as a f spin-facilitated Ising model on a

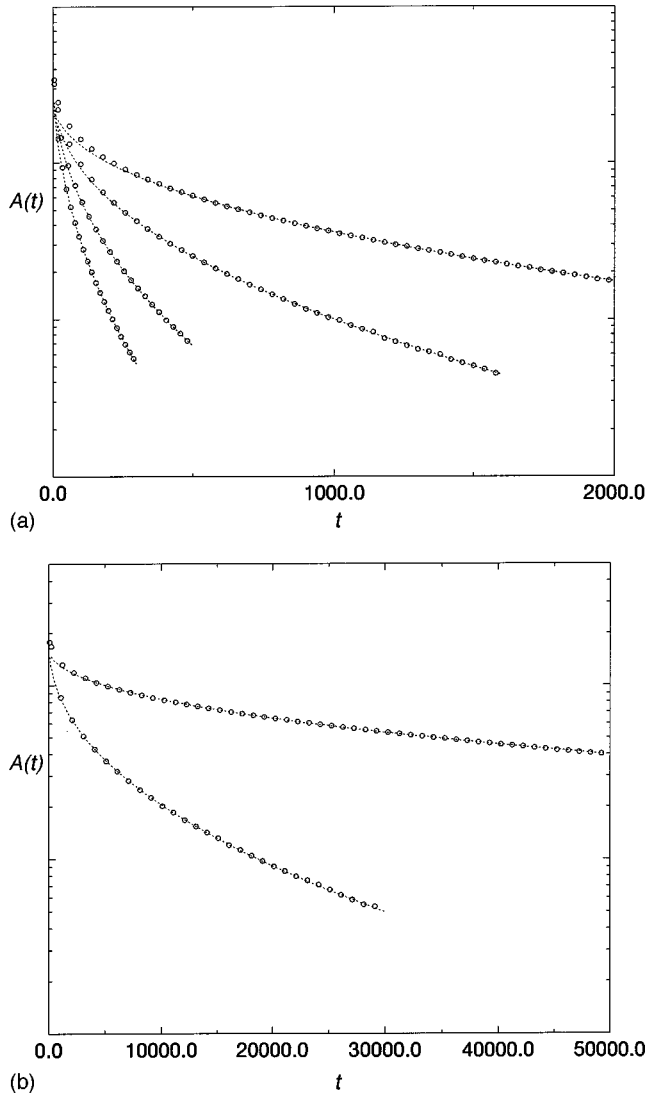


FIG. 1. The autocorrelation in semi-log scale with $h=0.40$ (a) at the couplings $J=0.10, 0.20, 0.30$, and 0.40 ; (b) at the couplings $J=0.50, 0.60$ (from below). The dotted lines represent the stretched exponential form fitted to the curves.

d -dimensional lattice, SFM[f, d]. The SFM[f, d] can be classified as an Ising-like model the kinetics of which is confined by restrictions of the ordering of nearest neighbors to a given lattice cell. These self-adapting environments influence in particular the long-time behavior of the spin-spin and therefore of the corresponding density-density correlation functions. These models were studied numerically^{16–19} (SFM[2,2]) and recently also analytically²⁰ (SFM[f, d]).

For the present investigations, we generalize the usual SFM[f, d] by introduction of additional short-range interactions that favor (antiferromagnetic case) or prevent (ferromagnetic case) the formation of liquid-solid (mobile-immobile) interfaces. It is the aim to study a two-dimensional generalized SFM[2,2] using Monte Carlo simulations.

II. MODEL

We consider a generalized spin-facilitated Ising model with nearest-neighbor interactions in two dimensions. The

Hamiltonian of the model is the same as that of the standard two-dimensional Ising model with an external field

$$H = -h \left(J \sum_{\langle ij \rangle} s_i s_j + \sum_i s_i \right), \quad s_i = \pm 1. \quad (1)$$

In our notation, the inverse temperature T and the Boltzmann constant k has been absorbed into the field h . Physically, the field corresponds to the difference of the energy per cell in the liquid and the solid state. In our later discussions, for convenience, we simply denote $h=1/T$. Here the coupling constant J describes the nearest-neighbor interactions. In the case of $J=0$, the original Fredrickson model is recovered. As mentioned above, the dynamic evolution of the generalized SFM[2,2] is subjected to a topological constraint that a spin flip is only possible if

$$\frac{1}{2} \sum_i (1 + s_i) \leq f. \quad (2)$$

In our simulations, the Metropolis algorithm is used and f takes its typical value $f=d$ with d being the space dimension, here we chose $d=2$. To assure that the system evolves into the physical section of the phase space, the initial configuration is always taken to be $s_i \equiv -1$, which means that we start from the complete liquidlike state. After the system has reached its equilibrium, we measure the auto correlation function

$$A(t) = \frac{1}{L^d} \left\langle \sum_i s_i(t') s_i(t+t') \right\rangle, \quad (3)$$

with L being the lattice size. Practically an average over t' is made in the numerical measurements. The lattice sizes are taken to be $L=50$ or $L=100$ depending on h and J . Up to the time regime of our simulations, no visible finite-size effect has been observed. To achieve reliable results and estimated statistical errors, we have performed five runs of simulations. Total samples for average range from 50 000 to 500 000. More samples are for larger values of h and/or J .

III. RESULTS AND DISCUSSION

In the low-temperature regime, the original Fredrickson Andersen model gives rise to a drastic enhancement of the relaxation time, which is characterized inevitably with glassy materials. As demonstrated in Ref. 21 there is no indication for a real glass transition or a critical temperature as predicted by mode coupling theory.^{1,2} For large time t , empirical approaches suggest a stretched exponential decay of the auto correlation function

$$A(t) \sim \exp[-(t/\tau)^\gamma], \quad (4)$$

where the exponent γ is presumably not an universal exponent, i.e., weakly depending on h . As a function of the h (inverse temperature), the relaxation time τ increases faster than according to an exponential law $\ln \tau \sim c+h$ manifested as a non-Arrhenius behavior but there is no singularity $\tau \rightarrow \infty$ at finite temperatures as suggested by the Williams-Landel-Ferry relation.⁵ The exponent γ offers a weak dependence on h , which is also confirmed by our simulations

TABLE I. The correlation time τ and the exponent γ measured for different couplings J and h .

(h,J)	τ	γ	(h,J)	τ	γ
(0.2,0.0)	2.4(4)	0.54(2)	(0.2,0.4)	3.4(3)	0.48(1)
(0.4,0.0)	7.5(11)	0.48(2)	(0.2,0.6)	6.(1)	0.48(2)
(0.5,0.0)	16.(1)	0.47(1)	(0.2,0.8)	12.(1)	0.48(1)
(0.6,0.0)	33.(3)	0.44(1)	(0.2,1.0)	30.(3)	0.47(1)
(0.7,0.0)	89.(9)	0.42(2)	(0.2,1.2)	89.(8)	0.45(1)
(0.8,0.0)	378.(36)	0.44(1)	(0.2,1.4)	550.(52)	0.44(1)
(1.0,0.0)	10900.(1145)	0.44(2)	(0.2,1.6)	12000.(1040)	0.44(2)
(h,J)	τ	γ	(h,J)	τ	γ
(0.4,0.1)	14.(3)	0.48(2)	(0.5,0.1)	39.(3)	0.46(1)
(0.4,0.2)	26.(4)	0.47(2)	(0.5,0.2)	132.(9)	0.44(2)
(0.4,0.3)	64.(5)	0.45(1)	(0.5,0.3)	1185.(98)	0.45(2)
(0.4,0.4)	210.(17)	0.44(1)	(0.5,0.4)	19800.(1940)	0.43(2)
(0.4,0.5)	1620.(110)	0.45(2)			
(0.4,0.6)	21900.(800)	0.44(2)			
(h,J)	τ	γ	(h,J)	τ	γ
(0.8,0.02)	530.(54)	0.43(2)			
(0.8,0.04)	1250.(105)	0.44(1)			
(0.8,0.06)	3090.(201)	0.44(2)			
(0.8,0.08)	6950.(747)	0.44(2)			
(0.8,0.10)	17750.(1140)	0.44(2)			
(h,J)	τ	γ	(h,J)	τ	γ
(0.4,0.1)	14.(3)	0.48(2)	(1.0,-0.02)	2963.(274)	0.44(2)
(0.5,0.1)	39.(3)	0.46(1)	(1.0,-0.06)	840.(45)	0.44(2)
(0.6,0.1)	162.(17)	0.44(1)	(1.0,-0.10)	152(15)	0.39(1)
(0.7,0.1)	1220.(150)	0.45(2)	(1.0,-0.14)	57.(5)	0.39(1)
(0.8,0.1)	17750.(1140)	0.44(2)	(1.0,-0.18)	20(2)	0.38(1)
			(1.0,-0.22)	9.(1)	0.36(1)

presented below. For the generalized SFM[2,2] we are interested in the role of the extra coupling J . Physically, a positive exchange coupling $J > 0$ and $h > 0$ intend to support the creation of solid-solid pairs, which are partially frozen in, e.g., such a coupling tends to enhance the relaxation time.

We observe that for fixed h , even if it is small, the auto correlation decays in a stretched exponential form for large time t and more interestingly, the relaxation time τ increases also rapidly following a non-Arrhenius law when the coupling J increases. The exponent γ exhibits also a weak dependence on the coupling J . In Fig. 1, the auto correlation for $h=0.40$ and for different values of the coupling J is displayed in semilog scale with lines of circles. Obviously the decay is not an exponential one. The dotted lines are the stretched exponential fit to the curves. We see clearly the fit is rather good. The resulting relaxation time τ and the exponent γ for different h and J are listed in Table I. For comparison, results for the original Fredrickson Andersen model ($J=0$) are also included. In Fig. 2, we have plotted the correlation time τ as a function of the coupling J for different values of h in semilog scale. Clearly it is a non-Arrhenius behavior.

Our model has two parameters h and J . For large h and/or J , a strong freezing process manifested in a strong slowing down of the relaxation time is observed and the auto-correlation shows similar dependence on both h and J , respectively. Alternatively to the autocorrelation function let us consider the magnetization $M(h,J)$. This quantity is an

essential one also within our reinterpretation of the kinetic Ising model as an appropriate candidate to describe glasses. The magnetization corresponds to the density of the immobile particles. In Fig. 3, the dependence of the relaxation time τ and the exponent γ on $M(h,J)$ are depicted for different

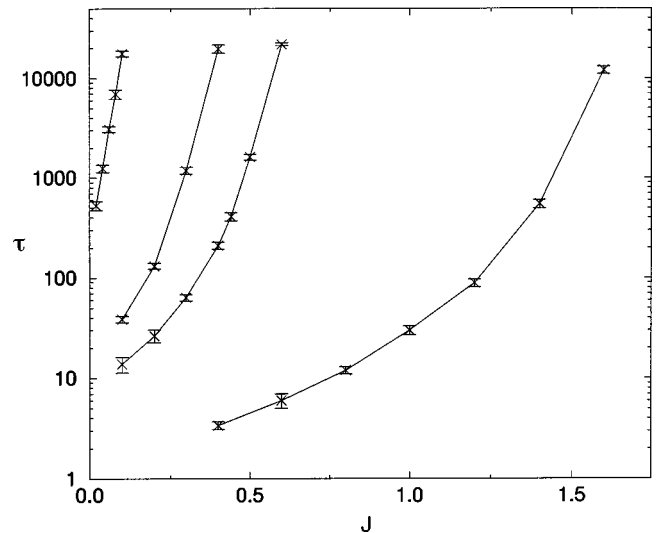
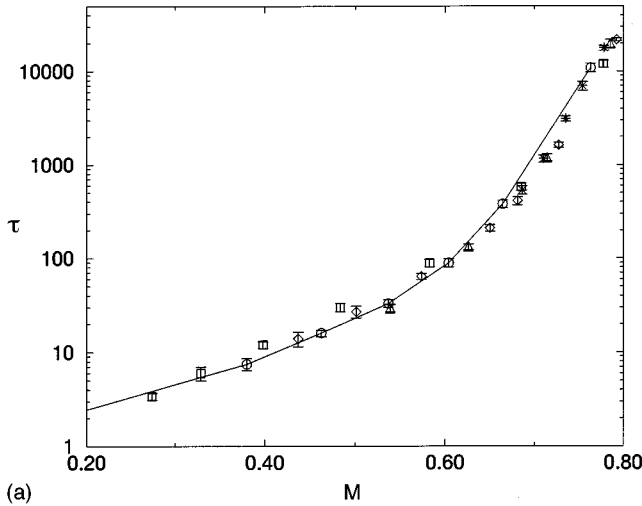
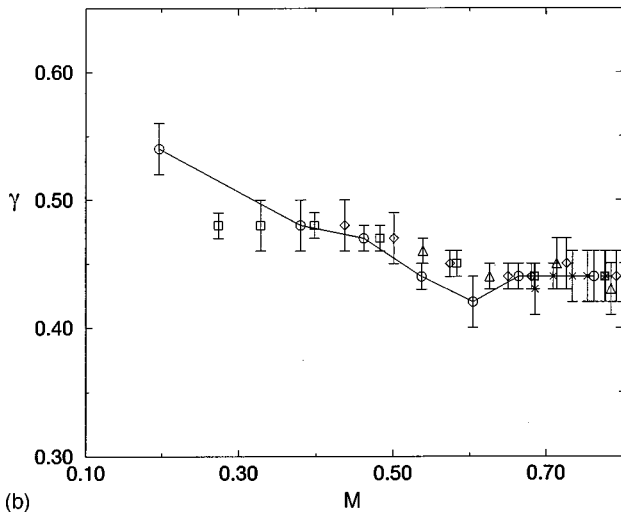


FIG. 2. The dependence of the correlation time τ on the coupling constant J . The corresponding fields are $h=0.80, 0.50, 0.40$, and 0.20 , respectively (from left). All curves show a non-Arrhenius behavior.



(a)

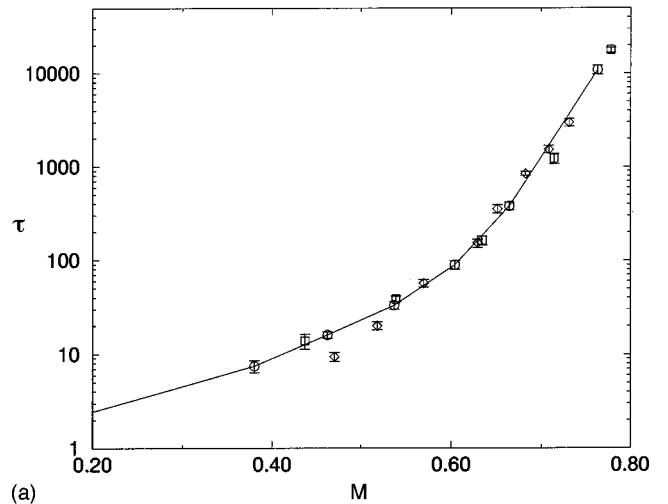


(b)

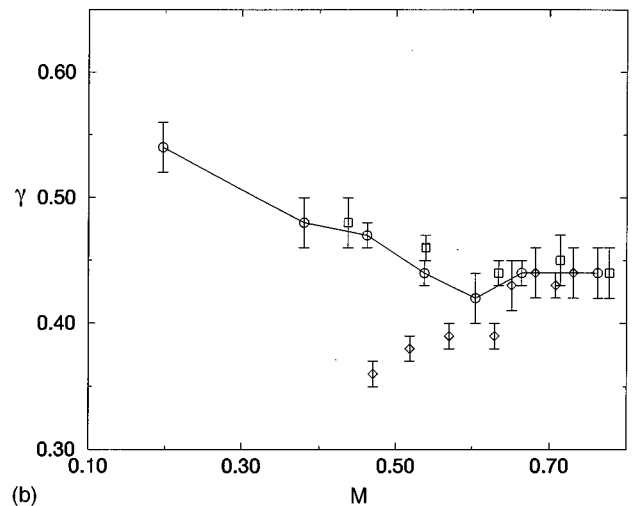
FIG. 3. The collapse of (a) correlation times τ in semilog scale and (b) the exponent γ . Circles with a solid line, squares, diamonds, triangles, and stars correspond to $J=0.00, h=0.20, h=0.40, h=0.50,$ and $h=0.80,$ respectively. Data are taken from the first five blocks of Table I.

couplings J and different fields h . A nice collapse of the data is observed. A nonzero coupling J practically induces a short-range spatial correlation. However, our results show that such a short spatial correlation length does not change dramatically the properties of the glass system.

The interaction can also be antiferromagnetic, i.e., with a negative coupling constant J . In this case, the relaxation time τ decreases when the magnitude of J becomes larger. This can be seen in the last block of Table I. However, we again find a nice collapse of the data with negative J and positive J when the magnetization M is chosen to be the scaling variable. This is shown in Fig. 4(a). The situation is slightly different for the exponent γ . In Fig. 4(b), for small $|J|$, it joins to the data points with positive J . But for bigger $|J|$, i.e., small M and with short relaxation times τ , the exponent γ decreases rather than increases as in the case of positive J . This phenomenon is also understandable. A static antiferromagnetic coupling favors the coexistence of both liquid and solidlike regions in the neighborhood. But the limit of $J \rightarrow -\infty$ for fixed h does not exactly correspond to a high-temperature state.



(a)



(b)

FIG. 4. The collapse of (a) correlation times τ in semilog scale and (b) the exponent γ . Circles with a solid line, squares, and diamonds correspond to $J=0.00, J=0.10,$ and $h=1.00$ with negative J , respectively. Data are taken from the last two blocks of Table I.

IV. CONCLUSIONS

We obtain as a main result that the dynamics of the generalized SFM[2,2] is controlled only by the magnetization $M(h, J)$, i.e., the density of up (or down) spins. When we plot the correlation time τ as a function of $M(h, J)$, all data points for different values of h and J collapse to a single curve. The collapse of the data points for the exponent γ is also observed except for the case with a strong antiferromagnetic coupling. These results show that the stretched exponential behavior is rather universal.

Our simulations have not yet covered the regime near the critical point of the standard Ising model ($h \rightarrow 0$ but hJ remains finite at its critical value). In this critical regime, both the glass transition and the second-order phase transition take place. To study the mixed critical behavior of two phase transitions is an interesting extension of the present work.

Finally, it should be remarked that the empirical stretching exponent γ is rather close to 0.5. This result is in agreement with recent analytical calculations.²¹

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