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Facilitated model for glasses

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An analytical approach to the spin facilitated kinetic Ising model [Phys. Rev. Lett. **53**, 1244 (1984); J. Chem. Phys. **84**, 5822 (1985)] is proposed using a Fock space representation of the master equation. The cooperativity inherent in glassy materials is included by dynamical restrictions, which allows a change of local regions with different mobilities depending on the neighboring configurations. Applying a dynamical mean-field approximation, we get a non-Arrhenius relaxation behavior in the case of a simple activation dynamics for the kinetic coefficients. Whereas the short-time behavior is dominated by the conventional kinetic Ising model, the long-time limit is determined by the restrictions. Including an additional static interaction strength favoring a solidlike state, the relaxation time becomes drastically enlarged due to the partial freezing of the system. Related to this phenomenon the nucleation rate is strongly decreased, preventing the nucleation of droplets of the condensed phase. Analyzing the influence of spatial fluctuations, the perimeter of regions with extremely low mobility can be estimated in the long-time limit, resulting in a logarithmic behavior.
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I. INTRODUCTION

Although the liquid-glass transition is studied with different methods, it remains one of the unsolved challenging problem in the theory of phase transitions [1,2]. A large number of glass-forming liquids offer a very pronounced relaxation dynamics when they are cooled fast enough from a high-temperature liquid state to a low-temperature state. The relaxation patterns are nonexponential in time and depend strongly on temperature. The slow dynamics is also a feature of conventional phase transitions [3]; however, frozen liquids do not evolve into an observable long-range correlated or ordered state that is persistent in time.

The liquid-glass transition is dynamic in origin and characterized inevitably by a high cooperativity of local processes [4]. To illustrate the behavior let us divide the system into small cells, say, on the nanometer scale, which are characterized by different local mobilities. Assuming that the dynamics is based on hopping processes, there is a cooperative rearrangement of certain cells in order to change more immobile cells into more mobile ones and vice versa. The cooperativity originates from the observation that a given cell embedded in an environment of cells of different mobilities can be trapped by its neighbors. Therefore, a change of a state depends strongly on the processes in the neighborhood. Obviously, the cooperativity increases for decreasing temperatures [5], leading to a non-Arrhenius behavior in the plot of the characteristic relaxation time τ_r of the glass transition and the inverse temperature T^{-1} in the low-temperature regime $T_g<T<T_s$, where T_g is the glass transition temperature [6,7]. Although a fit of this curved trajectory is performed by a Williams-Landel-Ferry curve [8] with a finite Vogel temperature, the experimental data are also compatible with a zero Vogel temperature [9]. So a better theoretical understanding of the phenomena is highly desirable.

A great effort in an analytical analysis is based on the mode-coupling theory elaborated in several papers [1,10–14]. As a main result of that approach the authors find a critical temperature T_c above the glass temperature T_g. It can be interpreted as a precursor of the glass transition [15]. At T_c the system exhibits a crossover between a liquidlike (mobile) and an immobile solidlike behavior. The above-mentioned cooperativity is included only indirectly. The motivation of the present paper is to understand in an analytical manner the influence of cooperativity and the local restrictions. To this aim we use the Fredrickson-Andersen model (FAM) [16,17]; compare also Refs. [18] and [19]. The FAM is of Ising type, however, with kinetic confinements originated from the mentioned restrictions. The two orientations of the spin are related to the particle density of lattice cells, where the spin-down state represents a low density (a high mobility, liquidlike region) and the spin-up state corresponds to a high density (a low mobility, solidlike region), respectively. Neglecting diffusive motion, which should be relevant in the glass phase, we assume that the dynamics is based on hopping processes between states of different mobilities. However, the topological restrictions are taken into account explicitly, resulting in the above-mentioned cooperativity: Local spin-flip processes are allowed only if the number of neighboring cells in the spin-up state is smaller than or equal to a certain number f (the f-facilitated FAM).

It should be remarked that a real liquid cannot be mapped completely onto an Ising model. Nevertheless, the FAM reflects the main features of glassy systems. For instance, the FAM yields a Kohlrausch-Williams-Watts behavior for the decay of the autocorrelation function [20]. Furthermore, it reveals a typical non-Arrhenius behavior of the relaxation time [21] that can be approximately fitted by $\ln\tau\propto A+BT^{-2}$. Obviously, the FAM does not offer a β process that should be interpreted as an indication that the underlying master equation is valid on a time scale larger than the time scale of the fast β process. However, the observation of a stretched exponential decay of the autocorrelation function as well as the non-Arrhenius behavior points out that the FAM should be considered as a reasonable approach to describe the slow α process of supercooled liquids well below the critical tem-

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perature T_c of the usual mode-coupling theory [1,10–13]. Moreover, the FAM can be extended in a straightforward manner including other degrees of freedom, for instance, vacancies [21,19].

Up to now the FAM had been studied numerically [20,17,21]. The results confirm the relevance of the model in describing real glasses. In particular, the FAM reflects the essential properties of supercooled liquids.

Recently, we solved the one-dimensional FAM [22], which does not show a phase transition but a significant enlarged relaxation time. The aim of the present paper consists in an analytical approach for an intermediate-temperature regime and for higher dimensions. Let us stress that we do not analyze the behavior in the vicinity of a possible critical temperature.

Within our analytical calculations the steric hindrances are automatically included by mapping the underlying dynamics satisfying a master equation onto a quantum problem in terms of Pauli operators. They allow only an empty or a single occupied state at a certain lattice cell. The basic idea of our approach is comparable to the one due to Jäckle and Krönig [23] applied in computer simulations.

II. ANALYTICAL FORMULATION OF THE FAM

As proposed above, the system is divided into cells that are characterized by the orientation of the spin

$$S_i = 1 - 2n_i. \tag{1}$$

If the lattice cell i is occupied by mobile particles, the state is assign to $n_i = 1$, whereas in the case of an immobile state we set $n_i = 0$. A certain configuration is characterized by $\vec{n} = (n_1, n_2 \dots)$. The dynamics is introduced via the master equation written in the symbolic form

$$\partial_t P(\vec{n},t) = L' P(\vec{n},t). \tag{2}$$

Here P is the probability that the configuration $\vec{n}$ is realized at time t. The evolution operator L' will be specified below.

Furthermore, let us introduce annihilation and creation operators to formulate a state in terms of occupation number operators where in the present paper those operators have the eigenvalues 0 and 1. Thus the problem is to formulate the dynamics in such a way that this restriction is taken into account [24–32]; for a recent review see Ref. [33]. The situation in mind can be analyzed in a seemingly compact form using master equation [34,35,28] introduced above [Eq. (2)]. Following Refs. [34–36,28,31], the probability distribution $P(\vec{n},t)$ is related to a state vector $|F(t)\rangle$ in Fock space according to $P(\vec{n},t) = \langle \vec{n} | F(t) \rangle$, with the basis vector $|\vec{n}\rangle$ composed of second quantized operators. The master equation (2) can be transformed to an equivalent equation in Fock space

$$\partial_t |F(t)\rangle = \hat{L} |F(t)\rangle. \tag{3}$$

The operator L' in Eq. (2) is mapped onto the operator $\hat{L}$. Up to now the procedure is independent of the operators used. Originally, the method had been applied for the Bose case [34–36]. Recently, an extension to restricted occupation numbers (two discrete orientations) was proposed [24,25,28–31] introducing Pauli operators. These operators commute at different cells and anticommute at the same lattice cell. A further extension to a p-fold occupation number is possible [37].

The relation between the quantumlike formalism and the probability approach is given by

$$|F(t)\rangle = \sum_{n_i} P(\vec{n},t) |\vec{n}\rangle. \tag{4}$$

As it was shown by Doi [34], the average of an arbitrary physical quantity $B(\vec{n})$ is defined by the average of the corresponding operator $\hat{B}(t)$,

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

with the state function $\langle s | = \Sigma \langle \vec{n} |$. Using the relation $\langle s | \hat{L} = 0$, the evolution equation for an operator $\hat{A}$ can be written

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

It seems necessary to note that all the dynamical equations covering the classical problem are determined by the commutation rules of the underlying operators and the structure of the evolution operator $\hat{L}$. In our case the dynamics of the model is given by spin-flip processes indicating a change of the local mobilities and densities, respectively. In addition to conventional thermodynamically controlled flip rates, one has to consider the topological restriction that a flip is possible only if the following condition is satisfied:

$$\frac{1}{2} \sum_{j(i)} (1 + S_j) \leqslant f, \tag{7}$$

where $j(i)$ means all neighbors of lattice cell i and f is the restriction number (the f-spin-facilitated kinetic Ising model, the FAM). Thus a flip $S_i = -1 \rightleftharpoons S_i = +1$ is allowed only if the number of neighboring cells in the low-mobility state does not exceed f. Note that this topological restriction guarantees the highly cooperative dynamics of the FAM, the influence of which had been already demonstrated by numerical simulations [17,21]. We should stress that the FAM is a model on a mesoscopic scale that does not take into account detailed atomic motion. The occupation numbers play a role of block variables as the block spin in a scaling theory of phase transitions. So we conclude that the crossover from shear diffusion to shear waves also characteristic of supercooling cannot be expected within the Fredrickson-Andersen model.

The evolution operator for a simple flip process reads [38]

$$L_i = \lambda (d_i^\dagger - d_i d_i^\dagger) + \gamma (d_i - d_i^\dagger d_i), \tag{8}$$

where λ and γ are flip rates. The operators d_i and $d_i^\dagger$ fulfill the commutation rule of Pauli operators

$$[d_i, d_j^\dagger]_- = \delta_{ij} (1 - 2 d_i^\dagger d_i). \tag{9}$$

The occupation number operator $n_i = d_i^\dagger d_i$ is related to the spin due to Eq. (1).

Using Eq. (6), the average occupation number operator obeys

$$\partial_t \langle n_i \rangle = \lambda \langle 1 - n_i \rangle - \gamma \langle n_i \rangle . \tag{10}$$

The confinement manifested by Eq. (7) can be exactly taken into account by assuming flip rates that depend on the configuration of the local environment of a given cell; compare Eq. (11). Here we consider the f-facilitated FAM with an arbitrary number f, which is the reasonable case in f-dimensional networks; see also Ref. [22].

To include the local restrictions (7) we replace $\lambda \to \lambda n_{j_1} \cdots n_{j_f}$ and $\gamma \to \gamma n_{j_1} \cdots n_{j_f}$. Hence the evolution operator reads

$$\hat{L} = \sum_{i, j_1 \cdots j_f} \chi_{ij_1 \cdots j_f} [\lambda (d_i^\dagger - d_i d_i^\dagger) + \gamma (d_i - d_i^\dagger d_i)] n_{j_1} \cdots n_{j_f}, \tag{11}$$

where $\chi_{ij_1 \cdots j_f}$ is nonzero when the lattice indices $i, j_1 \cdots j_f$ belong to plaquettes of nearest neighbors. A flip process in the cell i is allowed whenever the neighboring cells are occupied, i.e., such a flip process is favored in the case of a liquidlike neighborhood.

Further analysis can be performed for an arbitrary number f. In the case of a simple cubic lattice we set $f = d = z/2$, where d is the spatial dimension and z is the number of nearest neighbors.

Moreover, the method can be extended by including the mutual interaction between different cells and by considering finite temperatures. To this aim we have to replace the evolution operator by [38,39]

$$\begin{aligned} \hat{L} = \nu \sum \chi_{ij_1 \cdots j_f} & [(1 - d_i) \exp(-\beta H/2) d_i^\dagger \exp(\beta H/2)] \\ & + [(1 - d_i^\dagger) \exp(-\beta H/2) d_i \exp(\beta H/2)] n_{j_1} \cdots n_{j_f}. \end{aligned} \tag{12}$$

Here ν is a new hopping rate, $\beta = T^{-1}$ is the inverse temperature of the heat bath, and H is the Hamiltonian describing the static interaction between different cells that gives rise to a nonlocal dynamics different from Eq. (8). We assume that

$$H = -h \sum_i S_i - \frac{1}{2} \sum_{\langle ij \rangle} J_{ij} S_i S_j . \tag{13}$$

Using Eq. (1), the Hamiltonian can be rewritten (up to an unimportant constant) as

$$H = 2 \sum_i (h + Jz) n_i - 2J \sum_{\langle i,j \rangle} n_i n_j \tag{14}$$

This Hamiltonian contains both the energetic level of each cell and the mutual interaction between adjacent regions of different energy and mobility. It should be remarked that the original FAM corresponds to a Hamiltonian with $J = 0$ (h plays the role of an activation energy). The Hamiltonian determines only the energetic levels of the actual states, i.e., it is responsible for the thermodynamic transition rates. Although we are aware that a real liquid cannot be mapped completely onto such an Ising-type Hamiltonian, the FAM is well established and it seems to be a reasonable model to describe supercooled liquids.

The Hamiltonian (14) completes the mapping of a real liquid onto an Ising model in comparison with the original FAM. In the case of a nonzero coupling J a solid-solid interaction is favored whenever the relations $h > 0$ and $h - Jz > 0$ are fulfilled. This result follows by replacing the operator n by $m = 1 - n$. With the help of the algebraic properties of Pauli operators we get

$$\begin{aligned} & \exp(-\beta H/2) d_i^\dagger \exp(\beta H/2) \\ & \quad = d_i^\dagger \exp\left[\beta \left(h + Jz - 2 \sum_{l(i)} J_{il} n_l \right) \right], \end{aligned} \tag{15}$$

where $l(i)$ means summation over all neighbors of cell i.

III. MEAN-FIELD APPROACH

Using Eqs. (12) and (15) we are able, as before [compare Eq. (10)], to derive an exact evolution equation for the average particle number operator [Eq. (6)]. A whole hierarchy of equations results. In the simplest approximation we decouple the equation within the mean-field approach. To be specific, we replace the Hamiltonian (13) by its mean-field expression

$$H_m \equiv \sum_i \tilde{h} n_i ,$$

with

$$\tilde{h} = 2h + 2Jz \langle S \rangle, \quad \langle S \rangle = 1 - 2 \langle n \rangle . \tag{16}$$

The decoupling procedure is performed in the spirit of the coarsening of the free energy such as in the conventional Ising model. Whereas the exact equation for the averaged density is valid on a microscopic scale, say, of the size of cells l, the equation resulting after decoupling and the subsequent continuum limit should describe the system on a scale $x \gg l$. However, the main feature of the model, the inclusion of hard-core dynamics at the same lattice site manifested in the anticommutation relations, is taken into account. As in the continuum Ising model, the correlations are included in spatially varying terms, the influence of which is discussed later. The evolution equation for the averaged density $\langle n_i(t) \rangle$ can be obtained in the same manner as Eq. (10). However, the corresponding equation is based on the modified evolution operator (12), which gives rise to an additional factor proportional to $[\langle n_i(t) \rangle]^f$. Furthermore, Eq. (15) is taken into account, leading to exponential terms.

Performing the continuum limit for $\langle n_i(t) \rangle = n(\vec{x}, t)$ (for simplicity we set $l = 1$), we find

$$\partial_t n = \gamma n^f [\exp(-\beta \tilde{h}/2)(1 - n) - n \exp(\beta \tilde{h}/2) n], \tag{17}$$

where $\tilde{h}$ is defined in Eq. (16). The steady state yields

$$n_0 = \frac{1}{\exp(\beta\tilde{h}) + 1}. \tag{18}$$

The relaxation time can be calculated by making an ansatz $n(t) = n_0 + n_1(t)$, which leads to

$$\partial_t n_1(t) = \tau^{-1} n_1(t),$$

with

$$\tau = \frac{1}{2\gamma n_0^f \{\cosh(\beta\tilde{h}_0/2) - 2(T^\star/T)\, n_0 \exp(\beta\tilde{h}_0/2)\}}, \tag{19}$$

where $\tilde{h}_0 = 2[h + T^\star(1 - 2n_0)]$ is introduced [see Eq. (16)] with the characteristic temperature $T^\star = Jz$ discussed in Sec. III B.

Performing the stability analysis for the kinetic Ising model without restrictions, we find that τ [Eq. (19)] can be put in the final form

$$\frac{\tau}{\tau_0} = \tau_{Is} n_0^{-f},$$

with

$$\tau_{Is} = \frac{1}{2[\cosh(\beta\tilde{h}_0/2) - 2(T^\star/T)\, n_0 \exp(\beta\tilde{h}_0/2)]}. \tag{20}$$

Here τ_0 stands for a microscopic time scale expressed by the inverse hopping rate ν and the coordination number. Equation (20) has a simple interpretation. Due to the kinetic restrictions introduced before, the relaxation time of the FAM is given by the corresponding relaxation time of the conventional kinetic Ising model scaled with a factor n_0^f, the consequences of which will be discussed in the following.

A. Activation dynamics

As in the numerical simulation (see Refs. [17],[21]), we discuss first a pure activation dynamics characterized by neglecting the static coupling between different cells. In our notation we set $J = 0$. Physically this means that a change of the mobility (spin-flip process) is possible with a probability proportional to $\exp(\pm\beta h)$ according to Eq. (15). The energy h is nothing but the difference between the liquidlike and the solidlike state per particle.

The relaxation time is given by

$$\ln\left(\frac{\tau}{\tau_0}\right) = \beta h + (f - 1)\ln[1 + \exp(\beta h)]. \tag{21}$$

From here we conclude a different behavior for low and high temperatures:

$$\begin{aligned} \ln\left(\frac{\tau}{\tau_0}\right) &\simeq (f-1)\ln 2 + \frac{\beta f h}{2} \quad \text{for} \quad \beta h \ll 1, \\ \ln\left(\frac{\tau}{\tau_0}\right) &\simeq \frac{\beta(2f-1)h}{2} \quad \text{for} \quad \beta h \gg 1. \end{aligned} \tag{22}$$

Asymptotically two Arrhenius trajectories appear. However, the low-temperature branch offers a slowing down compared to the high-temperature region manifested in a smaller slope. The crossover between both curves is roughly estimated to be $(\beta h)_{\mathrm{CO}} = 2\ \ln 2$.

The behavior of the relaxation time can be more pronounced for the function $F(t)$ introduced in Ref. [40]

$$F(T) = \left\{-\frac{d\ \ln(\tau/\tau_0)}{dT}\right\}^{-1/2}.$$

As a result, we find for our model

$$F(T) = T\sqrt{2/h}\left[1 + \frac{2(f-1)}{1 + \exp(-\beta h)}\right]^{-1/2}. \tag{23}$$

As mentioned above, the high- and low-temperature regimes are different:

$$\begin{aligned} F(T) &\simeq \sqrt{2/h}\frac{T}{\sqrt{f}} \quad \text{for} \quad \beta h \ll 1, \\ F(T) &\simeq \sqrt{2/h}\frac{T}{\sqrt{2f-1}} \quad \text{for} \quad \beta h \gg 1. \end{aligned} \tag{24}$$

B. Influence of interaction

The inclusion of a mutual interaction between regions of different mobility leads, in the mean-field approximation (15), to a temperature- and state-dependent activation energy $\tilde{h} = 2(h + T^\star\langle S\rangle)$, where $\langle S\rangle$ obeys the self-consistent equation

$$\langle S\rangle = \tanh\left(\frac{h}{T} + \frac{T^\star\langle S\rangle}{T}\right). \tag{25}$$

The characteristic temperature $T^\star = Jz$ does not signal a second-order phase transition because the activation energy has to be a nonzero parameter.

First, let us discuss the low-temperature limit $T \to 0$. Here we find that $\langle S\rangle_0 \simeq 1 - 2\exp[-2(h/k_B T) - 2(T^\star/T)]$. The relaxation time tends to infinity for $T \to 0$, according to

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

with

$$\tilde{h}_0 = 2\left\{h + T^\star\left[1 - 2\ \exp\left(-2\frac{h}{T} - 2\frac{T^\star}{T}\right)\right]\right\}. \tag{26}$$

In the case of vanishing activation energy h the relaxation time would greatly increase, approaching $T^\star$, indicating a second-order phase transition. Because the activation energy is always nonzero, the increase of τ is finite due to the finite strength of h. Furthermore, different from the conventional

Ising model where the relaxation time decreases below $T^\star$, the kinetic restriction prevents such a decay. The reason is the result revealed in Eq. (20), which relates the relaxation time of the FAM to that of the usual Ising model. Due to the factor n^f in the denominator of Eq. (20), which becomes smaller for low temperatures ($n_0 \to 0$ for $T \to 0$), the relaxation time increases drastically, characterizing the partial freezing of the mobility.

We remark that for a small activation energy $h < h_c$ and $T < T^\star$ there exist three solutions of the self-consistent equation (25) where the critical activation energy is given by

$$h_c = \frac{2T^2}{3(T^\star)^3}(T^\star - T)^{2/3}.$$

It is easy to see that in the case of $h > 0$ considered here, only the solution with positive $\langle S \rangle_0$ is relevant.

IV. NUCLEATION PROCESS

As a further consequence of the inclusion of the interaction manifested in Eq. (19) let us discuss the nucleation process. When a glass former is cooled down the formation of droplets of the solid phase is prevented. We would like to demonstrate within our formalism that the nucleation radius of droplets remains finite at low temperatures.

Due to the kinetic restrictions manifested in state-dependent kinetic coefficients [see Eq. (12)] and the inclusion of the mutual interaction between particles situated at nearest-neighbor sites, a change of the density is effectively allowed via diffusion. For instance, let us assume that there is a mobile particle within a certain region surrounded by immobile ones. Such a particles can change its position only by hopping processes, which corresponds, in our approach, to a diffusive motion. Therefore, the diffusive transport is responsible only for a nucleation process. For that reason we adopt the conventional Lifshitz-Slyozov-Wagner (LSW) theory [41] to describe the latest stage of the nucleation process, however, with the modification due to the time scaling manifested in Eq. (20). Because cooling is performed with a certain cooling rate, i.e., the temperature is time dependent via $T(t)$, one obtains a diffusion coefficient that also depends strongly on time. Thus one has to replace the combination Γt with the diffusivity Γ by $\int_0^t \Gamma(t')dt'$. One can be convinced that the final result of the LSW theory remains essentially unchanged by this replacement. In particular, the average radius of a droplet is now given by [42]

$$\overline{R} = \left(\frac{4\pi}{9} \int_0^t \Gamma(t')dt' \right)^{1/3}. \tag{27}$$

To estimate the behavior of Γ we take into account that the diffusion coefficient for a fixed spatial interval is proportional to the inverse of a characteristic time that is chosen to be the relaxation time given by Eq. (20). Furthermore, let us assume a constant cooling rate κ. Thus we get

$$\int \Gamma(t')dt' \simeq \int \tau(T)^{-1} \frac{dt'}{dT} dT \equiv -\frac{1}{\kappa} \int_{T^\star}^{T_0} \tau^{-1}(T) dT. \tag{28}$$

Here the temperature T_0 is the lowest temperature for which the system is cooled down. In particular, we consider the limit $T_0 \to 0$. Using the low-temperature limit for the relaxation time (22) results in

$$\overline{R} \simeq \kappa^{-1/3} \left\{ \mathrm{Ei}\left(\frac{(2f-1)h}{2T^\star} \right) + \frac{2T^\star}{(2f-1)h} \times \exp[-(2f-1)h/T^\star] \right\}^{1/3}, \tag{29}$$

where $\mathrm{Ei}(x)$ is the exponential integral. In contrast to the conventional LSW theory, the droplet radius remains finite. Droplets of finite sizes appear and disappear. Nucleation is strongly reduced, i.e., a sufficiently fast cooling procedure prevents crystallization. Obviously the same kind of calculation can be performed with a nonconstant cooling rate. The main result is unchanged.

V. SPATIAL FLUCTUATIONS

Up to now we have neglected spatial fluctuations originating from the static coupling strength J. Following the spirit of the mean-field approach, we can include lowest-order gradient terms by inserting Eq. (15) into Eq. (12) and using

$$\sum_{l(i)} J_{il} n_l = \sum_{r(i)} J_{ir}(n_r - n_i) + zJn_i \approx Jl^2 \nabla^2 n_i + zJn_i. \tag{30}$$

As a result we get an evolution equation for the average density field $n(\vec{x},t)$ in the form

$$\begin{aligned} \partial_t n = {} & \gamma n^f [\exp(-\beta \tilde{h}/2)(1-n) - n \exp(\beta \tilde{h}/2) n] \\ & + \gamma 2J/T l^2 n^f [\exp(-\beta \tilde{h}/2)(1-n) \\ & + n \exp(\beta \tilde{h}/2) n] \nabla^2 n. \end{aligned} \tag{31}$$

To study the influence of spatial fluctuations we make the ansatz $n(\vec{x},t) = n_e(t) + m(\vec{x},t)$, where $n_e(t)$ satisfies Eq. (17). Inserting this ansatz in Eq. (31) and linearizing the resulting equation with respect to $m(\vec{x},t)$ we find

$$\partial_t m(\vec{x},t) = C(t) m(\vec{x},t) + D(t) \nabla^2 m(\vec{x},t), \tag{32}$$

where the coefficients $C(t)$ and $D(t)$ [see Eq. (33)] are determined by the pure time-dependent density $n_e(t)$ obeying Eq. (17). Equation (32) is solved by

$$m(\vec{x},t) = m_1(\vec{x},t) \exp\left(\int_0^t A(t')dt' \right).$$

Here m_1 obeys a diffusive equation, however, with a time-dependent diffusivity $D(t)$,

$$\partial_t m_1(\vec{x},t) = D(t) \nabla^2 m_1(\vec{x},t),$$

with

$$D(t)=\gamma 2Jl^2/Tn_e^f[(1-n_e)\exp(-\tilde{h}_e/2T)+n_e\exp(\tilde{h}_e/2T)],$$
$$\tilde{h}_e=2[h+T^\star(1-2n_e)]. \qquad (33)$$

Further progress can be reached by analyzing the asymptotic behavior in the long-time limit. Moreover, we are interested in the solidlike region where $n_e \ll 1$, i.e., for low temperatures. Therefore, let us expand the corresponding equations for $n_e \to 0$. In this limit we find for a cubic lattice with $f=d=z/2$,

$$\partial_t n_e \simeq -rn_0^{f+1},$$
$$D(t)\simeq d_1 n_e^d - d_2 n^{d+1}, \qquad (34)$$

where the coefficients are given by

$$r=\nu[(1-2T^\star/T)\exp(-a)+\exp a],$$

$$d_1=\frac{\nu 2Jl^2}{T}\exp(-a),$$

$$d_2=\frac{\nu 2Jl^2}{T}[(1-2T^\star/T)\exp(-a)-\exp a],$$

with

$$a=\frac{h+T^\star}{T}.$$

To estimate the long-time behavior let us solve Eq. (34) and find the time-dependent diffusion coefficient $D(t)$. The resulting diffusion equation with an arbitrary time-dependent diffusivity [Eq. (33)] can be solved analytically, leading to a characteristic dynamical length scale $L^2=\int^t D(t')dt'$. Using Eqs. (33) and (34), it results in the long-time limit

$$L^2=\exp(-2a)\frac{2Jl^2}{fT}\ln(t/t_0). \qquad (35)$$

Physically, this time-dependent characteristic length scale L is interpreted as the perimeter of solidlike regions that are self-organized by the underlying restrictions. 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 glass phase. In this case we would expect a more pronounced increase related to the very strong decrease of mobility.

VI. CONCLUSIONS

The main motivation of our paper was to demonstrate the influence of a constraint to the kinetic Ising model in the sense of a facilitated kinetic Ising model (the FAM). Although the mean-field theory is a very crude approximation, it reveals some features of a glass transition such as no stable static solution and a crossover between Arrhenius and non-Arrhenius behavior. The model seems to be an alternative way towards an analytical approach of the dynamics of cooperatively rearranging regions. Such a behavior seems to be relevant for liquids in the vicinity of a glass transition.

The mutual restrictions of moving particles leading to a freezing of the mobility are included explicitly in the model by a formulation of the collective stochastic dynamics in terms of Pauli operators. In doing so we were successful in solving the mean-field equation that is still a nonlinear evolution equation. The approach is related to the conventional one using the Landau theory of phase transitions. As a further step we want to include fluctuation effects to get a better understanding of the glass transition. Furthermore, we consider the correlation function and should include defects that yield different relaxation processes, as already demonstrated numerically [21].

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