Interface growth in a two-spin facilitated kinetic Ising model

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The kinetic growth of an interface in a two-spin facilitated Ising model is analyzed by numerical simulations. In contrast to the usual Eden model, a new nonuniversal short-time behavior is detected. For larger times a crossover to the universality of the Eden model takes place. It is shown that the bulk density and the evolution of the roughness of the interface are mainly determined by a characteristic length ξ_{kin}, which is closely connected to the correlation length ξ of the glass phase. The velocity of the interface can be described by a Williams-Landel-Ferry-like behavior.

I. INTRODUCTION

Theoretical models for irreversible growth such as the Eden model[1] have been of increasing importance in recent years. Natural realizations of such irreversible kinetic growth processes arise in different areas (biological pattern formation,[2] epidemic processes,[3] evolution of tumors[1]).

A typical irreversible growth can be observed in solids in which both crystalline and amorphous phases exist. While the crystalline phase derives from the liquid through a first-order phase transition, in the liquid to a glass (or amorphous phase) transition one observes with decreasing temperature a characteristic large and continuous slow down of the dynamics without any clear identification of a phase transition (glass transition). Such a dynamic glass transition is characterized by a high cooperativity of the underlying elementary processes.

Of important interest is the behavior upon an interface between a crystalline phase and the undercooled liquid (glass). In principle, the energetically more favorable crystalline phase tends to grow into the glass phase. While the crystalline surface grows irreversibly and unlimited at the contact with a simple liquid (kinetic growth of crystals in solutions or melts), the irreversible growth of the interface into an undercooled liquid is strongly determined by the dynamics in the glass phase (for instance, a typical situation is given in polyethylene). A theoretical description of an irreversible growth is given by the phenomenological approach of Kardar, Parisi, and Zhang[4,5] to the evolution of the interface in the Eden model. The Eden model describes a kinetic growth process on a lattice, starting from one or more seed particles. The growth mechanism follows an iterative rule, in which a new particle is added on a nonoccupied site of the Eden cluster, by choosing this site at random among all the possibilities. Defining $s_i = 0, -1$ to be the state (nonoccupied, occupied) at site i the following reaction scheme is considered:

$$s_i = -1 \text{ and } s_j = 0 \longrightarrow s_i = -1 \text{ and } s_j = -1, \quad (1)$$

which describes the kinetic process on the lattice.

The well-known analytical descriptions of the dynamic glass transition use continuous generalized hydrodynamic models and different types of mode coupling approximations,[6–9] whereas numerical approaches are based on microscopical and/or mesoscopical models that are usually solved by Monte Carlo (MC) like simulations. A powerful numerical method for the simulation of the glass transition in dense polymer systems (typical glass former) is the bond fluctuation method.[10,11]

Another possibility used for the following investigation is the numerical investigation of spin-lattice models by MC methods, introduced by Fredrickson and Andersen.[12,13] The original Fredrickson model[12,13] (two-spin-facilitated kinetic Ising model in d dimensions, usually acronymed as 2SFM) gives a local topological explanation for some essential features of the cooperativity in glass phases. Here, each spin on a regular lattice of size N^d (d: dimension) has two states $s_i = +1$ (up) and $s_i = 0$ (down). A down state is associated with a small liquidlike region (high mobility, low density), an up state with a small solidlike one (low mobility, high density) in a real glass and we can assume that the energy level of $s_i = 1$ is lower than the energy level of $s_i = 0$. The transition between these two states is determined by an energy difference ϵ_0 between the liquid and solidlike state and the temperature T,

$$p_{\text{trans}}(\text{solid} \rightarrow \text{liquid}) \sim \exp\left(-\frac{\epsilon_0}{T}\right). \quad (2)$$

The cooperativity between the single-flip mechanisms at different lattice sites is given by additional dynamic topological restrictions: only a spin with

$$m \leq n \quad (n : \text{fixed integer}, \; n \leq z) \quad (3)$$

(z: coordination number of the lattice) neighboring spins in the up state (solid) can undergo transition jumps

$$s_i = 0 \rightleftharpoons s_i = 1. \quad (4)$$

II. MODEL

The description of the irreversible growth of an interface in an undercooled liquid should be possible in a lat-

tice model by continuing the kinetics of both the Eden model and the two-spin-facilitated Ising model. To that end we introduce a three-state spin $s_i = 0, \pm 1$. Here, the spin state $s_i = -1$ at the lattice site i corresponds to the crystalline phase, whereas the states $s_i = +1$ and $s_i = 0$ are associated with low and high mobility states, respectively, of the glass phases.

In the model the following transitions between these states are possible:

(1) Reversible (thermodynamic) jumps between low ($s_i = +1$) and high ($s_i = 0$) mobility states of the glass [under the topological restriction, that the number m of neighboring low mobility states ($s = +1$) and crystalline states ($s = -1$) is determined by $m \leq n$] with the reaction scheme

$$s_i = 0 \rightleftharpoons s_i = 1. \tag{5}$$

(2) Irreversible jumps from a liquidlike state ($s_i = 0$) to a crystalline state ($s_i = -1$) if one or more neighboring spins are in the $s_i = -1$ state and the number m of neighboring low mobility states ($s = +1$) and crystalline states ($s = -1$) is also given by $m \leq n$:

$$s_i = 0 \longrightarrow s_i = -1. \tag{6}$$

This model is controlled by the temperature which determines the dynamical structure of the glass, and the geometry as well as dimension of the initial interface.

In the following we investigate this model in a $(d+1)$-dimensional space $\mathbf{R}$, being formed by a d-dimensional surface $\mathbf{S}_d$ and a spreading direction $\mathbf{h}$, i.e., $\mathbf{R} = \mathbf{S}_d \times \mathbf{h}$. The extension in the spreading direction is unlimited, whereas the surface $\mathbf{S}_d$ has the area L^d.

In the original Eden model after a finite relaxation time an invariant kink front appears to move with a constant velocity which suggests a solitonlike behavior. The volume of bulk (region, in which the average of the local-spin state is nearly -1) grows as $V \sim t^d$, consequently the height of the bulk as $h \sim t$. The interface itself shows a fractallike structure. A characteristic measure of the roughness of the growing surface is the width

$$\sigma = \sqrt{\frac{\int_0^L d^d x [h(\mathbf{x},t) - \overline{h(\mathbf{x},t)}]^2}{L^d}}, \tag{7}$$

which depends on the length L and the height h by the universal scaling law

$$\sigma \simeq L^{\chi} g\left(\frac{t}{L^z}\right). \tag{8}$$

This model has a critical dimension $d_c = 2$ and universal critical exponents χ and z which are obtained by a renormalization-group approximation.[5,14] Especially for $d = 1$ follows $\chi = 1/2$ and $z = 3/2$. Numerical studies[15,16] on the Eden model show that $z \approx 1.55 \pm 0.15$ and $\chi/z \approx 0.30 \pm 0.02$ in good agreement with the renormalization-group approach.

Numerical investigations[17,18] of the 2SFM show the existence of a characteristic length ξ_{kin} for the glass phase. This length corresponds approximately to the averaged diameter of the active patches[17] and can be interpreted in the framework of a phenomenological cooperativity concept (see also the Appendix) as an approximate diameter of a cooperative region (being formed by particles which participate in a structural transition of the glass). These regions are always characterized by a finite length (in the temperature regime of the glass transition), no indication for any long-range correlation can be detected (compatible with the fact that a dynamic glass transition shows no phase-transitionlike properties). The existence of such a characteristic length induces the assumption that the scaling law (8) no longer holds. Rather, a more sophisticated behavior controlled both by the geometrical length L and the thermodynamic (kinetic) length ξ_{kin} can be expected. Only in the case when L and the roughness of the interface σ are sufficiently large in comparison to the thermodynamic length ξ_{kin}, the inhomogeneities in space (of the order of ξ_{kin} and caused by the glass dynamics) become irrelevant and the roughness of the interface can be described by the scaling law (8).

III. NUMERICAL SIMULATIONS AND RESULTS

In the present numerical calculations we analyze the behavior in a $1+1$ dimensional space. Therefore, we use a square lattice with the coordination number $z = 2(d+1) = 4$ and the restriction $n = 2$ for the 2SFM with cyclic boundary conditions in the direction (x direction) orthogonal to the spreading direction (y direction) of the interface.

In a first step the generation of an equilibrium configuration of the glass phase was realized by using a MC Metropolis algorithm with the weight $\exp\{-\epsilon/T\}$ (ϵ is the energy difference between the states $s = 0$ and $s = \pm 1$). After reaching the equilibrium state, all spins with coordinates ($y = 0$, $x =$ arbitrary) are given the value $s = -1$ determining the initial interface for the kinetic growth. The following irreversible growth of the interface is treated by Monte Carlo (MC) simulations, whereas the Metropolis algorithm for the glass phase stays unchanged. This numerical algorithm corresponds to the time evolution of the probability distribution $P(\mathbf{s},t)$ by a one step master equation,[19] generated by the reaction scheme (5) and (6).

A. Velocity of the interface

To get a measure for the time evolution of the interface in the present model we define the actual coordinates of the surface by $[x,h(x,t)]$ with

$$h(x,t) = \max y \text{ with } s(x,y,t) = -1.$$

The averaged height $\overline{h}(t) = \langle h(x,t) \rangle$ characterizes the growth process and depends on the length L (length of the system in x direction) and the temperature.

Figure 1 shows the evolution of $h(t)$ for different temperatures and a fixed length $L = 256$. One can see that

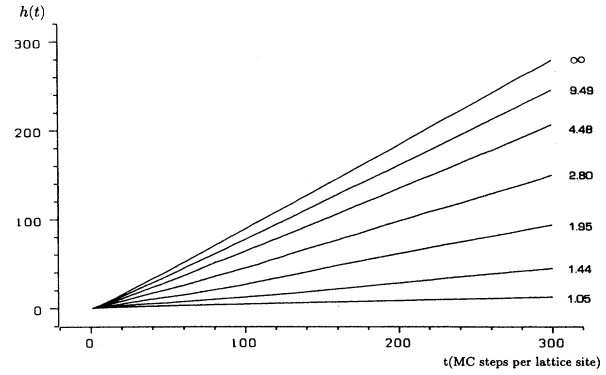

FIG. 1. Time evolution of the averaged height $h(t)$ for the length $L = 256$ and the reduced temperatures $\tau = T/\epsilon = \infty$, 9.49, 4.48, 2.80, 1.95, 1.44, 1.05, time in averaged steps per lattice site.

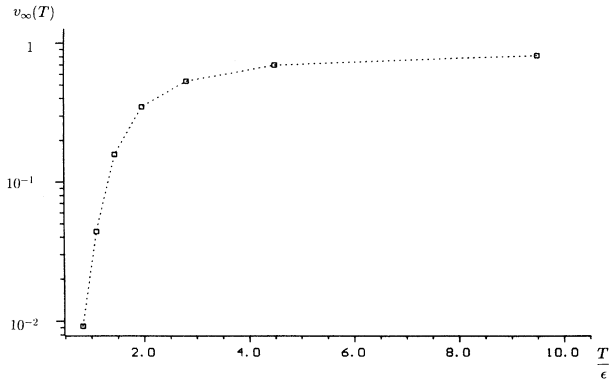

FIG. 3. Temperature dependence of $\log v_\infty(T)$.

the slopes of $h(t)$, after a short relaxation time, assume constant values. These stationary velocities of the interface $v(L,T)$ depend only on the length L and the temperature T (Fig. 2). For $L \to \infty$ the velocities $v(L,T)$ converge to finite values $v_\infty(T)$. These velocities are strongly temperature dependent and show the same behavior as the inverse relaxation times of the pure 2SFM (Refs. 20, 17, and 18) in the equilibrium state (Fig. 3). Therefore, we can expect, that the velocity of the interface becomes extremely small with decreasing temperature (WLF-like behavior), but the interface should not be pinned at any temperature. Such a pinning should only be possible if the underlying glass phase shows a transition to a nonergodic state, which, however, does not occur for the 2SFM.[20]

B. Density of the bulk

The probability that the spin at an arbitrary site i is $s_i = -1$ tends to unity in the original Eden model. In contrast to this behavior, in the present model this probability tends to a stationary value smaller than unity. The density of the state $s = -1$ in the bulk at the time t is defined by

$$\rho(t,L,T) = \frac{\sum_i \delta_{s_i,-1}}{\sum_x h(x,t)}. \tag{9}$$

Note, that this representation neglects the possible existence of overhangs at the interface. Because the overhangs are only present at the surface of the linearly with the time increasing bulk, this contributions vanishes for $t \to \infty$. Figure 4 shows the time evolution of the density (9) for different temperatures. Similarly to the velocity, the time evolution of the density approaches (in analogy to the velocity) stationary weakly L-dependent values. The extrapolation for $L \to \infty$ leads to a stationary limit of the density $\rho_\infty(T) = \rho(t,L,T)\,|_{L,t\to\infty}$, which is shown in Fig. 5. It should be remarked that all the lattice sites with spin state $s = -1$ form a single connected structure. Consequently, such structures are not determined by a pure random distribution, i.e., we can expect a distribution with a dominant length scale in the order of the characteristic length scale of the glass transition. Because of the temperature dependence of the density, we

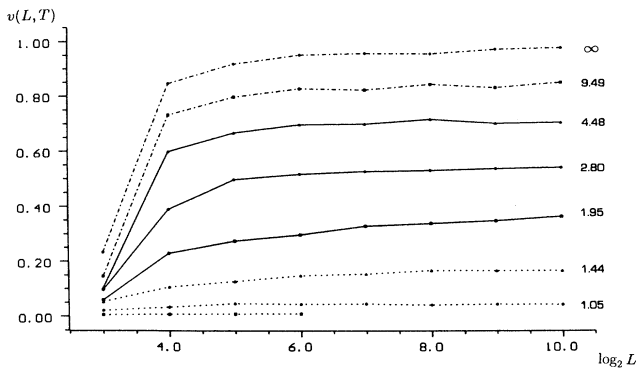

FIG. 2. Stationary values $v(L,T)$ as a function of L and for different reduced temperatures $\tau = T/\epsilon = \infty$, 9.49, 4.48, 2.80, 1.95, 1.44, 1.05, and 0.83.

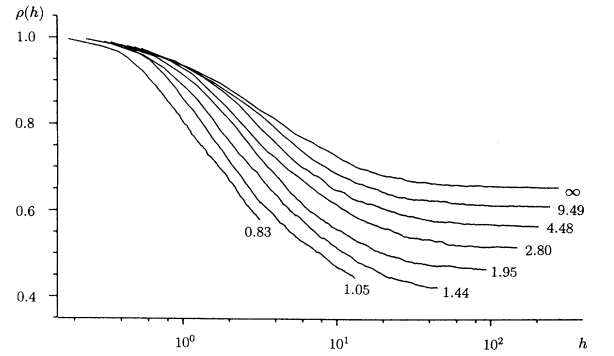

FIG. 4. Time evolution of the averaged density $\rho(t)$ for $L = 256$ and reduced temperatures $\tau = T/\epsilon = \infty$, 9.49, 4.48, 2.80, 1.95, 1.44, 1.05, and 0.83. Note, that in this reduced plot the time is replaced by the averaged height h.

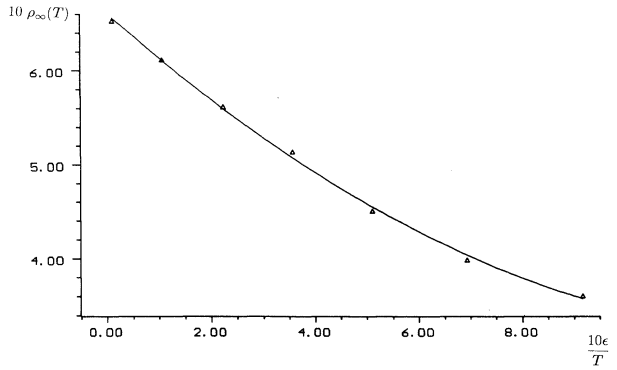

FIG. 5. Averaged density $\rho_\infty(T)$ of the bulk as a function of the inverse temperature T^{-1}.

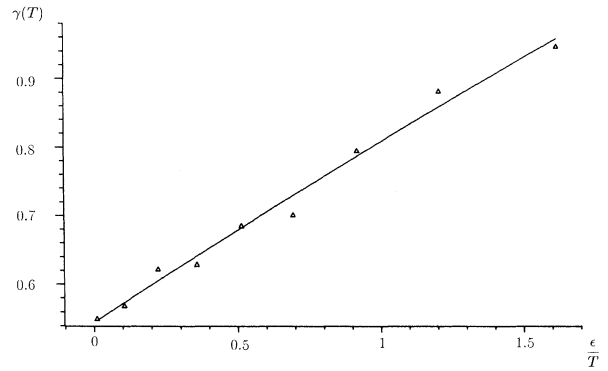

FIG. 7. Temperature dependence of the exponent $\gamma(T)$ for the first time evolution regime of the roughness.

can assume that this length (which determines for example the diameter of the bubbles or holes in the $s = -1$ structure) increases with decreasing temperature.

C. Roughness of the interface

The time evolution of the roughness (7) shows two and three characteristic regimes, respectively, (Fig. 6) and depends also on the length L and the temperature T. [Note, because of the monotonic relation between the height $h(t)$ and the time t (see Fig. 1), it is reasonable to use the usual representation $\sigma = \sigma(h, L, T)$.]

(1) The first regime is determined by an increase of the roughness with the height h by a power law $\sigma \sim h^{\gamma(T)}$ with a temperature-dependent exponent $\gamma(T)$ (Fig. 7).

(2) For large enough L a second power-law regime with a constant (universal) exponent $\sigma \sim h^{\beta}$ is observed. The prefactors of these two power laws depend only on the temperature, not on the length L. The value of β ($\beta = 0.3 \pm 0.02$) is the same exponent as for the original Eden model (see below).

(3) For large enough h, the roughness of the interface becomes a constant σ_∞, which depends on the length L by a power law $\sigma_\infty = A(T)L^{\chi}$ with a temperature-dependent prefactor (Fig. 8).

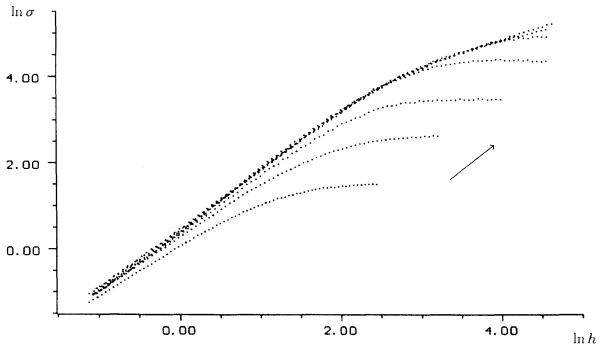

FIG. 6. Time evolution of the roughness σ for different lengths $L = 2^3 \ldots L = 2^{10}$ (L increases in the direction of the arrow) and the reduced temperature $\tau = T/\epsilon = 1.95$.

The crossover from the first to the second regime is independent on the length L and determined by the temperature exclusively. The corresponding thickness $\xi_{\text{kin}} = \sigma_{\text{crossover}}$ (Fig. 9) of the interface can be interpreted as a measure for the characteristic length of the cooperative regions in the glass phase. For a relative small thickness of the interface, the roughness is completely determined by the structure of the glass phase. With increasing evolution time and sufficiently large L also the thickness of the interface (characterized by the roughness measure σ) increases. For a thickness of the interface large in comparison to the length of the glass, the structure of the glass phase becomes more and more homogeneous as compared to the scale of the roughness of the interface and the growth process shows now the same universal behavior as the original Eden model. In other words, the local structure (this comprises all lattice properties, including the local-spin dynamic and the correlations of the glass phase with a finite correlation length), becomes irrelevant, because the kinetic growth approaches a critical behavior (critical dimension $d_c = 2$) with dominant long-range fluctuations.[4,14] Note, that this characteristic length of the cooperative regions is a result of the competition between the irreversible ki-

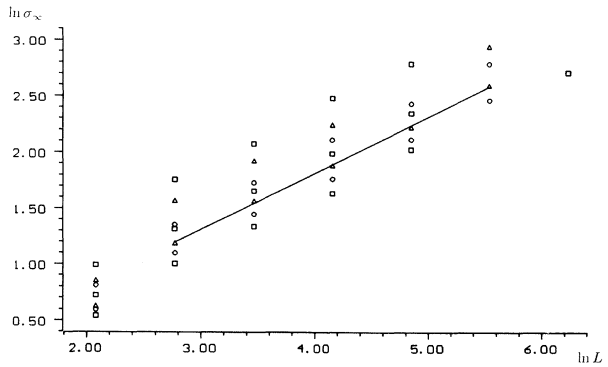

FIG. 8. σ_∞ as a function of L for different temperatures (from bottom to top $T = \infty$, 9.49, 4.48, 2.80, 1.95, 1.44, 1.09). The averaged slope (full line) for the region $L \geq 16$ is 0.48 ± 0.03, in good agreement with the results of the original Eden model.

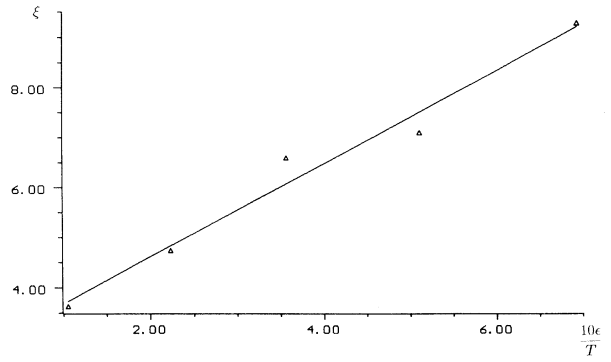

FIG. 9. Characteristic length $\xi_{\text{kin}} = \sigma_{\text{crossover}}$ as a function of the temperature T.

netic growth of the interface and the reversible dynamics of the glasslike bulk. Therefore, we expect a difference between the kinetic length ξ_{kin} and the usual correlation length ξ in a glass.[27,17,28] However, the temperature dependence of our kinetic characteristic length ξ_{kin} should scale in the same manner as the correlation length ξ in the glassy bulk up to an (irrelevant) prefactor.

IV. CONCLUSION

In our model the kinetic growth of an interface into a glass phase differs from other kinetic growth models of the Eden type in some essential points. Whereas in homogeneous space no restrictions hinder the kinetic growth of the interface, the growing process in 2SFM shows for each temperature ($T \to \infty$ inclosed) that all sites with $s = 1$ (low mobility states of the glass), neighboring the interface are unable to undergo a transition in the $s = -1$ state (i.e., to become a part of the growing cluster).

A similar growth shows the epidemic model.[21,22] Here, each site of the space with $s_i = 0$, neighbored to a site ($s_i = -1$) of the interface (active region) can realize either a transition to an elementary point of the growing cluster ($s_i = 0 \to s_i = -1$) with the probability p or a transition to a dead state ($s_i = 1$) with the probability q, which can never undergo a new transition in the $s_i = 0$ or $s_i = -1$ state [therefore, each site $s_i = 1$, neighbored to a site ($s_i = -1$) of the interface forms a passive region]. In other words, the growth of the interface is stopped in these points.

The growing process of the epidemic model is comparable to the kinetic growth in 2SFM in a restricted sense. The main difference between this model and the kinetic growth in the 2SFM is the existence of a critical probability q_c for the epidemic model. The growth of the interface in the epidemic model breaks down for each $q > q_c$, because the production rate of dead lattice sites is high enough to eliminate all active regions. This results in a configuration where all lattice sites with a spin value $s \neq 0$ neighbored to at least one spin with $s = 0$ are in the state $s = 1$, i.e., a further growing of the $s = -1$ cluster is impossible. Note, that this pinning effect exists also for the evolution of an interface in a stochastic force field. Here, the growth of the interface started only in the case, that an additional force $F > F_c$ (F_c is the critical force) is switched on in the spreading direction of the interface.[23] Really, such a pinning effect for the interface and therefore the existence of a critical temperature T_c is not observable for the kinetic growth in the 2SFM, because the always present dynamics between $s = 0$ and $s = 1$ states guarantees the possibility of a contact between an $s = 0$ state and the $s = -1$ cluster and therefore a permanent growing of the interface. The nonexistence of a critical temperature T_c corresponds to the fact, that the 2SFM has no nonergodic state[20,17,18] (in the sense of the mode coupling theory[6,7] or the Fredrickson-Anderson analysis[12,24]). Consequently, the lowering of the temperature leads only to an essential reduction of the growing velocity, not to a total freezing of the evolution.

The analysis of both the bulk density and the roughness of the interface suggests the existence of a temperature-dependent characteristic length ξ_{kin}, which determines the dynamics of the glass phase. This length ξ_{kin} corresponds not necessarily to a correlation length of the structure (the static structure factor of the SFM suggest a randomly distributed disorder). Rather, ξ_{kin} must be interpreted as the average diameter of a region around an arbitrary lattice site i of the model, in which a certain fraction of spins must change their state at least once ($s = 0 \leftrightarrow s = 1$), before the spin at site i can undertake a transition to another state.

The analysis of the numerical results for the characteristic length ξ_{kin} (Fig. 9) in the investigated T interval leads to the following approximation formula:

$$\xi_{\text{kin}} = 2.7 + \frac{9.4}{T} = \xi_0 + \frac{9.4}{T} + O(T^{-2}). \tag{10}$$

As mentioned above, we have different measures of the correlation length in spin-facilitated models.[27,17,28] Our kinetic length ξ_{kin} is a special measure, which is related to the irreversible (Eden) growth process and the reversible spin dynamics in the glass region and differs from usual equilibrium definitions.[27,17,28] Using the concentration $c = [1 + \exp(T^{-1})]^{-1}$, such an equilibrium correlation length ξ behaves as

$$\xi = \alpha \exp \frac{\beta}{c}. \tag{11}$$

The high-temperature limit of (11) corresponds to the approximation (10), whereas a fit in the low-temperature regime of our approximation leads to $\alpha = 3.32$ and $\beta = 0.31$. As expected, the temperature behavior of the correlation length (given by β) is in a good agreement with the numerical results of equilibrium simulations $\beta = 0.27$ (Ref. 27) and $\beta = 0.21$,[28] respectively. The prefactor α differs from the equilibrium data [$\alpha = 0.48$ (Ref. 27) and $\alpha = 1.13$,[28] respectively] as a consequence of definition of the kinetic characteristic length ξ_{kin}.

On the other hand, the characteristic lengths ξ_{kin} as obtained by the numerical simulations are also consistent with the approximately predictions of a fluctuation

theory for the case $T_\infty = 0$ and $d = 2$ (see the Appendix and note the 2SFM has no Vogel temperature T_∞). ξ_0 is an elementary length of the order of the diameter of a region of neighboring points around one lattice site, which corresponds to the characteristic length of the 2SFM for $T \to \infty$.

If the typical lengths, which determine the growing process of the interface in the 2SFM (e.g., the thickness σ), are sufficiently small as compared to $\xi_{\rm kin}(T)$, the influence of the temperature on the growing process becomes very important. This fact is expressed in the deviation from the universality of the original Eden model (e.g., a temperature-dependent critical exponent $\gamma(T)$ for the first region of the growing process).

On the other hand, if the kinetic growth reaches length scales, which are sufficiently large in comparison to the characteristic length $\xi_{\rm kin}(T)$, the system obeys the well-known universal behavior (8) of the Eden model with universal exponents χ and z, whereas the influence of the temperature is confined exclusive to irrelevant prefactors.

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APPENDIX: THE PHENOMENOLOGICAL FLUCTUATION THEORY OF GLASS TRANSITION

This theory is based on the concept of cooperativity[25] and gives a phenomenological reasoning for the existence of characteristic length scale which control the glass transition. To that purpose one divides for a given temperature T the volume of the glass in subvolumes of the length $\xi(T)$. This length is determined by the restriction that each region is approximately statistically independent from the neighbored subvolumes. The minimum possible length corresponds to the average diameter of a cooperative transition region (i.e., this length corresponds in our version of the 2SFM to the averaged diameter of a region around the spin s_i, in which the spins must change their state, before the restrictions allow a change of s_i).

In such an approximately statistically independent region of length ξ (i.e., volume $V_c = \xi^d$) the temperature fluctuations are given by[26]

$$\delta T^2 \simeq \langle (\Delta T)^2 \rangle = \frac{T^2}{C_V} \tag{A1}$$

with the specific heat proportional to the volume $C_V \sim V$, i.e.,

$$\delta T^2 \sim \xi^{-d}. \tag{A2}$$

In each of these subsystems a characteristic frequency $\omega(T)$ represents a measure for the velocity of the transitions between different thermodynamic states. Consequently, the temperature fluctuations δT correspond to a dispersion of the frequency $\delta\omega$. This means, that each observable, which is related to the glass transition, has a temperature-dependent width $\delta\omega(T)$ [Fig. 10(a)]. This width is determined by two characteristic frequencies $\omega_1(T)$ and $\omega_2(T)$, respectively [corresponding to the two turning points in Fig. 10(a)]. The temperature dependence of these frequencies can be fitted by a Williams-Landel-Ferry (WLF) curve[12] with a finite Vogel temperature T_∞

$$\ln \omega_\alpha - C = \frac{D_\alpha}{T - T_\infty}, \tag{A3}$$

($\alpha = 1, 2$). These two hyperbolas in the $\ln\omega$-T plot [Fig. 10(b)] have the same asymptotic behavior, i.e., only the constants D_α are different for the two hyperbolas. The distances between the hyperbolas along the $\ln\omega$ and T direction, respectively, corresponds to the width of the temperature fluctuations δT and the attached width $\delta \ln\omega$ in the frequency spectrum, respectively.

The hyperbolic structure implies the exact relation

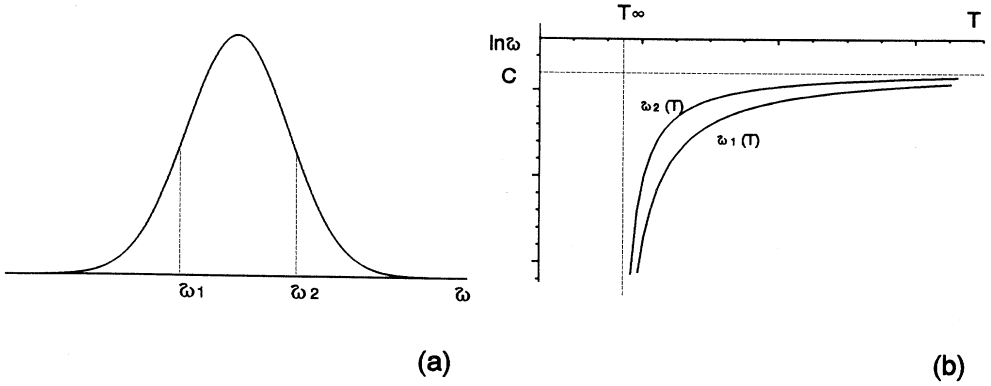

FIG. 10. (a) Frequency dependence of an observable, relevant to the glass transition, the width $\omega_2 - \omega_1$ reflects the dispersion of the frequency, (b) $\ln\omega$-T plot for $\omega_1(T)$ and $\omega_2(T)$, respectively.

$$\delta T \delta \ln \omega = \text{const.} \tag{A4}$$

On the other hand we have from (A3)

$$\frac{\delta \ln \omega}{\delta T} \simeq \frac{d \ln \omega}{dT} = -\frac{D_\alpha}{(T - T_\infty)^2}. \tag{A5}$$

Using (A4) and (A3), we get

$$\delta T \sim T - T_\infty \tag{A6}$$

and therefore with (A2)

$$\xi \sim (T - T_\infty)^{-2/d}. \tag{A7}$$

This scaling law gives an empirical connection between the characteristic length of the glass transition and the temperature.

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