Monte Carlo lattice-gas simulations of stable and unstable interfaces

R. Harris, Loki Jørgenson, and Martin Grant

*Department of Physics and Centre for the Physics of Materials, McGill University, Rutherford Building,
3600 University Street, Montréal, Québec, Canada H3A 2T8*

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We describe numerical investigations of the dynamics of interfaces in two dimensions, using a Monte Carlo algorithm that combines elements of both the microcanonical and the canonical methods. We employ a lattice-gas Hamiltonian which corresponds to a system with a first-order phase transition, so that we can study interfaces between distinct thermodynamic phases. Results include the first numerical observation of roughening dynamics for Hohenberg and Halperin's model *C* of critical dynamics, and the first numerical observation of the Mullins-Sekerka instability which results from the presence of a thermal gradient.

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I. INTRODUCTION

The dynamics of interfaces between distinct thermodynamic phases is an important topic in contemporary statistical mechanics. In this paper we focus on two particular aspects, namely, the roughening dynamics of interfaces near equilibrium, and the unstable growth of interfaces in the presence of a thermal gradient. Both these topics have been extensively treated in the literature [1–3]. Our paper describes the application of a new lattice-gas Monte Carlo algorithm to interfacial dynamics. We demonstrate that the algorithm reproduces known results for roughening and for the instability problem, and argue that it is a powerful tool for the investigation of more complex questions. Our present results deal entirely with one-dimensional interfaces in two space dimensions, so that the interfaces are always rough, but the extension to three dimensions is entirely straightforward.

We first give a detailed account of our algorithm, and discuss its applicability to the simulation of kinetics, rather than equilibrium properties. We then review the properties of the simple Hamiltonian which we use. The Hamiltonian models a system with a first-order phase transition, and we show how the latent heat of this transition enters in a natural way, and how it can be used to control the dynamics. We then describe the results for roughening dynamics, and for the growth of an interface made unstable by a temperature gradient, paying particular attention to computational details. In the discussion, we compare and contrast results for the two situations, and focus on implications for work in three dimensions.

II. THE ALGORITHM

It is convenient to introduce basic ideas with reference to the conventional Ising model, keeping in mind that the spin-up and spin-down states can readily be mapped onto the liquid and solid states of a lattice gas. Particulars of the algorithm are found in Appendix A, and a description

of the Hamiltonian that is actually used will be given later.

The algorithm is an extension of the microcanonical method introduced by Creutz in 1984 [4, 5] to study lattice gauge theories. As originally formulated, a system of N Ising spins is brought into contact and subsequent thermal equilibrium with a small auxiliary system known as a demon. Thermal equilibrium comes about in such a way that the total energy of spins plus demon is conserved. Because the demon is small, it acquires only a small fraction, $\sim 1/N$, of the total available energy, so that the energy of the spin system remains effectively constant. Hence arises the microcanonical nature of the procedure. Since it is in thermal equilibrium with the spins, the demon has the same temperature as them, and acts in many ways as a thermometer.

The original algorithm defines a set of rules for single-spin-flip processes. Suppose, at a given stage of the simulation, that the energy of the demon is E_D . This energy is defined to be always non-negative, and it is convenient, though not necessary, to set it equal to zero at the beginning of the simulation. To proceed, a spin is chosen as described in Appendix A, and the energy ΔE required for it to flip is determined. The flip takes place if *either* ΔE is negative, *or* the demon energy E_D is greater than or equal to ΔE . To ensure conservation of the total energy, E_D is then decremented by the amount ΔE . If E_D is less than ΔE , then the flip is not permitted. In summary, a flip is only permitted when

$$\Delta E \leq E_D$$

in which case

$$E_D \rightarrow E_D - \Delta E.$$

This procedure, repeated many times, leads to thermodynamic equilibrium between the demon and the spins. Therefore, in equilibrium, as the simulation proceeds, the probability of finding the demon with a particular energy E_D must be of Boltzmann form:

$$P(E_D) \simeq \exp(-E_D/T)$$

and the temperature T , in units of energy, corresponding to the state of equilibrium can be determined from measurement of the probability distribution.

Our extension of this algorithm has two distinct features, both of which are anticipated in a paper of Creutz [6]. In the first place, we use not one demon, but N of them, each associated with one site of the spin lattice. The energy distribution of each demon then measures the temperature of its own site, permitting the monitoring of temperature fluctuations and/or temperature gradients in the lattice. The demons facilitate the transport of energy between the sites of the lattice, so that the algorithm simulates the diffusion of heat.

To understand why it is appropriate to assign a temperature to local regions of the lattice, it is convenient to view the demons as a second thermodynamic system, of a particularly simple kind, which is in thermal equilibrium with the lattice. In equilibrium, the exchange of energy between the two systems is small. It is therefore again appropriate to invoke the canonical ensemble to determine the temperature from the distribution of energy fluctuations in the demon system. Similarly, if large regions of the system are in *local* equilibrium, then the energy distributions in those regions determine a local temperature. This provides a useful and convenient connection to the coarse-grained macroscopic equations of motion.

The second extension is a procedure for fixing the temperature(s) of selected sites in the lattice, typically those on one of the boundaries. In Creutz' original paper [4], the temperatures were fixed at either zero or infinity, but the idea is readily generalized to finite temperatures. We first described the details in a paper on the thermal conductivity of a kinetic Ising model [7], which served as the first test of many features of our current algorithm. The idea is to permit flips of the selected spins in accord with a Metropolis rule, namely, that the probability of flipping, P_i for site i is given by

$$P_i = 1; \Delta E_i \leq 0 \quad (1)$$

and

$$P_i = \exp(\Delta E_i/T); \Delta E_i > 0 \quad (2)$$

where ΔE_i is the energy required to flip the spin at site i , and T is the temperature.

To show that the two extensions to the basic algorithm yield meaningful results, we have carried out a series of simulations for which technical details are described in Appendix A. The rectangular geometrical arrangement is typical: Fig. 1 shows details. The system is defined on a square lattice, in the x - y plane, with an interface roughly parallel to the x axis. There are L sites along the x axis, and M along the y axis such that $L \times M = N$. Periodic-boundary conditions are imposed across the two boundaries AC and BD , but not across AB and CD . All sites other than those along AB and CD are in contact with their own demons, but the temperatures of the sites along the two opposite boundaries AB and CD are fixed at temperatures T_1 and T_2 , respectively. In the simplest case, we set $T_1 = T_2 = T$ and compute the equilibrium properties of the Ising model as a function of T . We are

FIG. 1. Schematic diagram showing the rectangular geometrical arrangement for the simulations of interface dynamics. There are L sites along the x axis and M along the y axis. The boundary conditions are periodic across the boundaries AC and BD , but not across AB and CD .

able to reproduce known results without difficulty.

A more interesting case is when the temperatures T_1 and T_2 are different. This is the analog of a thermal-conductivity experiment in which a temperature difference applied across a sample causes a steady temperature distribution. Thus, during a simulation, the sites in the bulk eventually reach a steady state, and the temperature distribution can be extracted from the demons with which they are in contact. In a previous paper [7], the existence of *local* thermodynamic equilibrium was demonstrated in several ways. Striking visual confirmation was obtained when both T_1 and T_2 were below T_c for the Ising model, with typical configurations showing that the local thermal-correlation length corresponded to the local temperature. In addition, typical runs for T_1 and T_2 above T_c gave well-defined temperatures, with fluctuations consistent with local thermodynamic equilibrium. Specifically, since the specific heat of the Ising model at high temperature varies as $1/T^2$, we were able to verify explicitly that $\delta(1/T)$ was independent of temperature in this regime. We were also able to compute the thermal conductivity of the Ising model, given our particular dynamics, and show that it was consistent with both high- and low-temperature series expansions.

The thermal-conductivity simulation thus makes clear that our Monte Carlo algorithm is useful when there is a steady state with local thermodynamic equilibrium. A larger question is its use to examine kinetic processes, either close to or far from equilibrium, where canonical Monte Carlo methods have had some success. Our approach is to demonstrate that our algorithm reproduces some well-known kinetic phenomena, so that we can justify its use to explore more complex situations. We will also comment on particular aspects which assist the modeling of dynamics.

III. THE HAMILTONIAN

As mentioned earlier, our work employs a Hamiltonian closely related to the Ising Hamiltonian

$$H = -J \sum_{\langle i,j \rangle} \sigma_i \sigma_j - \Delta \sum_i \sigma_i \quad (3)$$

where J is the spin coupling, the interactions are with the nearest neighbors, the spins are $\sigma_i = \pm 1$, and Δ represents a uniform external field. In the absence of Δ the second-order phase transition is at $T_c \approx 2.269J$.

In order to induce a first-order transition and a stable region of phase coexistence, we modify the Hamiltonian by giving a degeneracy δ_- to the upper state, $\sigma = -1$, of each spin, a degeneracy δ_+ to each lower state [8]. We require that δ_- be larger than δ_+ , and thus it is convenient to define an effective noninteger degeneracy of the upper states as $\delta = \delta_-/\delta_+$, so that $\delta > 1$. The effective degeneracy of the lower states is then unity. For the particular case of $\delta = 2$, we thus have a special case of the Blume-Emery-Griffiths Hamiltonian [9].

We maintain the interactions between the modified spins to be Ising-like, in the sense that *all* the degenerate upper states are assigned the value $\sigma = -1$. This causes a first-order phase transition at a melting temperature $T_m = 2\Delta/\ln \delta$, and introduces a latent heat Λ which is of order Δ . Consequently, Λ can be increased at constant T_m by modifying δ and Δ in the appropriate manner.

To understand the origin of this transition, it is helpful to consider the partition function for a set of *noninteracting* modified spins. This can be written as

$$Z = 1 + \delta \exp(-2\Delta/T) = 1 + \exp(-H_{\text{eff}}/T), \quad (4)$$

showing an effective temperature-dependent field $H_{\text{eff}} = 2\Delta - T \ln \delta$ which changes sign at $T_m = 2\Delta/\ln \delta$.

In the fully interacting system, the same effective field can be identified. It is convenient to classify configurations of the Ising model according to particular values of the number of spins N_- having $\sigma = -1$, and of the number of broken bonds B . Let the number of such configurations be $N_{N_-,B}$. In an external field 2Δ , the partition function then consists of all possible terms $Z_{N_-,B} = N_{N_-,B} \exp[-(E_B + 2\Delta N_-)/T]$, where the energy E_B is the energy of interaction, and $2\Delta N_-$, to within a constant, is the Zeeman energy. In the modified model, each of these terms carries an additional factor δ^{N_-} , which can be written as $\exp(N_- \ln \delta)$, leading as before to an effective field $H_{\text{eff}} = 2\Delta - T \ln \delta$.

The fully interacting system therefore has the same behavior as the usual Ising model in a field H_{eff} . The effect of this field at temperatures below T_m is to select states with magnetization close to $m = 1$, but at temperatures above T_m , states with $m \simeq -1$ are favored. At T_m there is a sharp transition, involving a discontinuity in m : it is a first-order transition. Figure 2 illustrates the situation by superposing the plot of m vs T on the phase diagram of the Ising model. Clearly, the first-order transition only exists if $T_m \leq T_c$.

At the transition, the latent heat Λ is given by the discontinuity in the entropy, $\Lambda = T_m \Delta S$, so that when the magnetization is close to ± 1 for $T \sim T_m$ we obtain

$$\Lambda \simeq T_m \ln \delta$$

so that $\Lambda \simeq 2\Delta$.

FIG. 2. Schematic phase diagram of the modified Ising model in the temperature-magnetization plane. The solid line is the path of an Ising system having a second-order phase transition at T_c . The dashed line shows the path of an Ising system which has been modified to exhibit a first-order phase transition at $T_m < T_c$. At T_m , the magnetizations of both systems are identical.

IV. ROUGHENING KINETICS

To test the use of our algorithm for kinetics, we chose to investigate a situation which is well understood analytically, namely, an interface between two coexisting phases which initially coincides with the x axis of an x - y coordinate system. In two dimensions it is known that such an interface becomes rough [10]. The roughness is measured by the width w of the interface, defined at time t as $w(t)^2 = \langle (\xi - \bar{\xi})^2 \rangle$, where $\xi(x, t)$ describes the position of the interface relative to the x axis, and $\bar{\xi}(t)$ is the average position. It is often convenient to assume that $\xi(x, t)$ is single valued, but, of course, this does not have to be the case. In thermodynamic equilibrium, an interface in a system of transverse dimension $0 \leq y \leq L$ has a width $w \sim \sqrt{L}$, irrespective of its dynamics, but the approach to equilibrium depends strongly upon the circumstances. In general, however, we expect the width to be of the form $w \sim L^\chi f(t/L^z)$, where f is a scaling function [11] and where, from equilibrium [10], $2\chi = (3 - d) = 1$. Thus, when finite-size effects are unimportant, $w^2 \propto t^{2\chi/z}$.

To identify possible values of the dynamic exponent z , we proceed in terms of the Fourier modes conjugate to x . Standard analysis [10] gives

$$\langle \xi^2(k, t) \rangle = \frac{T}{\sigma k^2} \{1 - \exp[-t/\tau(k)]\}, \quad (5)$$

where σ is the surface tension and T the temperature. $\tau(k)$ is the relaxation time for mode k .

The most familiar situation is when the relaxation of the interface is controlled by the surface tension, so that temperature fluctuations associated—via the Gibbs-Thompson relation [12]—with local curvature of the interface are attenuated by the diffusion of heat. In this case, the Fourier mode $\xi(k, t)$ has a relaxation time proportional to $1/k^2$ [10], so that short-wavelength modes come rapidly into equilibrium, and

long-wavelength modes dominate the late-time regime. Appropriate substitutions give

$$\langle \xi^2(k, t) \rangle = \frac{T}{\sigma k^2} [1 - \exp(-2D\sigma k^2 t)], \quad (6)$$

where D is the thermal diffusion constant. In the thermodynamic limit $L \rightarrow \infty$, the time evolution of the width w is then given by

$$w^2(t) = \int \frac{dk}{2\pi} \langle \xi^2(k, t) \rangle \sim t^{\frac{1}{2}} \quad (7)$$

in two dimensions, so that $z = 2$.

A less familiar situation is when the relaxation is controlled by the production and absorption of latent heat at the interface. This will be the case when the temperature fluctuations associated with the production of latent heat exceed those associated with the curvature of the interface. Expressed in the language of Hohenberg and Halperin [13], this is model C , namely, a system with a nonconserved order parameter (the magnetization) coupled to a conserved field (the energy), as distinct from model A as described previously. In model C , it is usual to consider the case of symmetric coupling [14], however in the present case the coupling is asymmetric, since the energy density associated with the two phases differs because of the latent heat. It turns out that the relaxation time for mode $\xi(k, t)$ in this situation is proportional to $1/k^3$, so that the time evolution of the width w is obtained from

$$\langle \xi^2(k, t) \rangle = \frac{T}{\sigma k^2} [1 - \exp(-4Dd_0 k^3 t)], \quad (8)$$

where d_0 is the capillary length, defined by $d_0 = \sigma T_m C / \Lambda^2$. In the thermodynamic limit, this gives

$$w^2(t) \sim t^{\frac{1}{3}}$$

in two dimensions, so that $z = 3$.

An explicit derivation of this result is given by Zia and co-workers [15, 16], employing a description in terms of dynamic functionals. They also give an expression for the crossover behavior of the relaxation time from $1/k^2$ to $1/k^3$ behavior. The reader is referred to their papers for details. For present purposes, however, it is convenient to adopt a simple approach [2], which can describe both the model A and the model C situations, and can also predict the form of the crossover. Details are found in Appendix B. The explicit expression for the relaxation time is

$$\tau(k) = d_0^2 [(1 + \tilde{k}^2)^{1/2} + \tilde{k}] / 2D\tilde{k}^3 \quad (9)$$

with $\tilde{k} = d_0 k$. It is clear that when $\tilde{k} \ll 1$, or $k \ll 1/d_0$, the relaxation time has $1/k^3$ behavior, but when $k \gg 1/d_0$, conventional $1/k^2$ behavior is seen. The capillary length d_0 thus defines the crossover length scale, and the time scale at which crossover should be observed is given by

$$\tau_0 \sim d_0^2 / D.$$

The crossover behavior is a consequence of the competition between the rate of generation of latent heat at the

interface and the rate of heat diffusion to the bulk phases. If the rate of generation is very much larger than the rate of heat diffusion, then the relaxation will be retarded by the coupling to the energy field and the interface will grow with $z = 3$. If on the other hand the heat produced (absorbed) at the interface is efficiently diffused away, then the thermal fluctuations will grow unhindered with $z = 2$.

Thus, short-length scale fluctuations will grow freely at early times, giving a $z = 2$ behavior. This will eventually be supplanted by $z = 3$ when all of the fluctuations shorter than d_0 have grown into their equilibrium values and the long-length scale relaxation dominates. This will continue until the system saturates at its equilibrium width ($w_{eq}^2 \propto L$).

V. RESULTS FOR ROUGHENING KINETICS

To test our algorithm against these analytical results, we prepared the system as two bulk phases with an exactly flat interface between them [17]. It was convenient to place the interface parallel to the x axis (see Fig. 1) such that the total magnetization was zero. The temperatures at the two free edges were set at T_m , and the other demons were prepared by randomly distributing the energy estimated to place them in thermal equilibrium with the spins (i.e., at T_m), as described in Appendix A. We chose $T_m = 0.9T_c$ to permit a sufficiently fast relaxation for observation while avoiding the influence of critical fluctuations: the thermal-correlation length at this temperature is approximately five lattice units. Keeping T_m fixed, we carried out a series of simulations with the spin degeneracy varied over a range of $1.0 \leq \delta \leq 5.0$, such that Λ therefore varied between 0 and $0.53J$ [17]. For each value of Λ we averaged data over at least 50 runs, each of which lasted 5000 Monte Carlo steps (MCS) per site.

The particular case of $\Lambda = 0$, the conventional spin-1/2 Ising model, with no latent heat, was treated separately from the others. It was convenient to use a code specifically designed for the Ising case, thereby avoiding the overhead associated with the degeneracy. The fact that an interface between the two phases of the Ising model is not strictly stable did not cause numerical difficulties.

The interface was defined as a single-valued function, ignoring overhangs and bubbles in the bulk; in general, this was a more-than-reasonable approximation since the interface was always distinct and well behaved. After 5000 MCS, the mean width of the interface was ~ 10 lattice units, while the average interface position remained roughly constant (± 1 lattice unit) throughout the run. Saturation of the roughening was observed in systems smaller than $L = 512$, and consequently to observe power-law growth, the system size was set at $L = 1024$ and $M = 64$. Due to the lack of self-averaging in the roughening behavior, it was necessary to take an ensemble average of the growth of interface fluctuations, and therefore about 50 trials for each value of δ were made.

Figure 3 shows the growth of thermal fluctuations on a typical interface for typical values of the system parameters, and Fig. 4 shows the observed time dependence of

FIG. 3. The evolution of an initially planar interface towards its equilibrium roughened state as $t \rightarrow \infty$. The configurations shown are after 0, 1000, and 5000 MCS. Fluctuations in the bulk phases have been masked out for clarity.

the square of the interface width for $\Lambda = 0, 0.20, 0.33$, and $0.53J$. Error bars are somewhat smaller than the symbol size. The width was measured every 10 MCS, although for clarity not all data points are shown, and the other system quantities such as the magnetization, demon energy, spin energy, demon temperature, etc., were monitored every 100 MCS. The first 200 MCS were ignored to allow demons and spins to come to equilibrium. During

FIG. 4. Double logarithmic plot of the growth with time t of the square w^2 of the width of interfaces with different latent heats Λ . The values of Λ , from the top curve to the lowest, are $0, 0.20J, 0.33J$, and $0.53J$, respectively. The dotted lines represent the theoretical growth rates with $z = 2$ and $z = 3$. The vertical arrows show the values of τ_0 for the intermediate values of Λ . Data are averaged over at least 50 runs for each curve.

this time it proved impossible to describe the growth of the interface by an analytical expression: we ascribed the behavior to transients resulting from the initial mismatch of demon and system energies (see Appendix A).

Also shown in Fig. 4 are two lines drawn with slope $1/2$ and $1/3$, respectively. It is clear that systems with latent heats greater than $\Lambda \sim 0.5J$ grow uniformly with $z = 3$, while strong transitional behavior is observed for the smaller values of Λ . As Λ tends to zero, approaching the standard field-free Ising model, the growth rate is well described by $z = 2$. Power-law fits to the data confirm this visual impression: for the $\Lambda = 0$ Ising data we obtain $z = 2.02 \pm 0.03$, and for the $\Lambda = 0.53J$ data we obtain $z = 3.00 \pm 0.04$. For this latter case, the crossover length d_0 is of order of the lattice spacing: consequently, the transition from $z = 2$ to $z = 3$ occurs very early, well before the 200 MCS limit, and the interface is observed to grow with $z = 3$. The intermediate systems show less well-defined but consistent behavior. In both cases, there is apparently a transition region between $z = 2$ and $z = 3$ behavior, occurring near the time $\tau_0 \sim d_0^2/D$, but the growth rates before and after the transition are not distinctly defined. The data between $t=200$ MCS and the time of transition shows $z \sim 2$, but the amount of data involved is not sufficient for much precision. After the transition, the growth rate approaches but does not reach $z \sim 3$. These results are in excellent agreement with theory.

Figures 5 and 6 show data for the power spectra plotted at different times as a function of the wave vector k . Figure 5 is for the case of no latent heat: the conventional Ising model. Clearly visible is the $1/k^2$ behavior at large values of k , and the time evolution displays the equilibration of successively larger wavelengths. The functional

FIG. 5. The power spectra at different times for an interface with no latent heat. Data are shown after 100, 500, 1500, and 5000 MCS. The dashed lines show fits to the predicted form of the power spectra; at large k , the $1/k^2$ term dominates. The data are averaged over 50 runs.

FIG. 6. The power spectra at different times for an interface with latent heat $\Lambda = 0.53J$. Data are shown after 1000, 3500, and 5000 MCS. The dashed lines show fits to the predicted form of the power spectra; at large k , the $1/k^2$ term dominates, and at small k the spectra are linear in k . The data are averaged over at least 50 runs.

form of the power spectra at small k is consistent with the analytical expression: $P(k) \rightarrow 2DTt$ as $k^2t \rightarrow 0$. For finite latent heat, the data in Figure 6 also shows the characteristic $1/k^2$ behavior at large k . Significantly, the form at small k is different, being consistent with $P(k) \rightarrow 4Dd_0kt$ as $k^3t \rightarrow 0$, as expected. Again, therefore, our data are in excellent agreement with analytical predictions.

VI. KINETICS OF AN UNSTABLE INTERFACE

We now proceed to study an interface in a thermal gradient. The liquid phase is set, initially, at a temperature somewhat below the equilibrium melting temperature T_m , so that it is supercooled, but the solid phase is set at or above T_m . The kinetics therefore describe the unstable evolution of the interface, and provide a lattice-gas analog of the Mullins-Sekerka instability [18, 2].

In qualitative terms, this instability arises from a competition between two effects. The first of these is the preference of the unstable supercooled liquid phase to condense onto the surface of the solid. This happens in an inhomogeneous way, such that local modifications to the curvature of the interface are consistent, via the Gibbs-Thomson relation, with the local temperature. The second is the tendency for the relaxation mechanisms, the same as those describing the equilibrium kinetics, to restore the interface to its equilibrium configuration.

In the very early time regime, linearizing the equations of motion gives a number of results which seem consistent with the experimental situation. The derivation [2] follows the procedure given in Appendix B, except that there is an additional term in Eq. (B2) which depends

upon the diffusion length ℓ . This length is defined as $2D/v$, where D is the diffusion constant, as before, and v is the velocity of the almost planar interface in the direction of the initial thermal gradient. As described in the Appendix, omitting the damping due to surface tension gives a “dispersion relation” for the damped modes of the form

$$\omega(k) \equiv -[\tau(k)]^{-1} = \frac{2D}{\ell d_0} \tilde{k}(1 - \tilde{k}^2 \ell / d_0), \quad (10)$$

where as before, $\tilde{k} = kd_0$, and where the k^3 term is precisely the same as for the kinetics of the equilibrium interface. Within this linear approximation, therefore, there is a range of wave vectors for which ω is positive and which exhibit unstable behavior. The limit of instability is at $k_c = (d_0 \ell)^{-1/2}$, and the most unstable wave vector is $k_0 = (3d_0 \ell)^{-1/2}$. The characteristic length scale of the instability is therefore the geometric mean of the small and large length scales, d_0 and ℓ , respectively.

In the late-time regime, where fingers have already taken shape, there are also a number of results which relate the steady-state velocity of the tip of a growing finger to its radius of curvature. Such relationships must take the form [2]

$$v = \frac{2D}{d_0} V(\tilde{u}, \tilde{\rho}), \quad (11)$$

where $\tilde{u}$ is the undercooling in units of Λ/C , and $\tilde{\rho}$ is the curvature, ρ , in units of d_0 . In the limit where $\tilde{\rho} \gg 1$, the prediction is that $v\rho \sim \text{constant}$ for a given undercooling.

Predictions of more general validity, relating for example to the mechanism of mode selection, or the selection of sidebranches, are much more difficult. It is generally accepted that the most successful approach in recent years has been the “microscopic-solvability” technique [3] which has made new predictions susceptible to experimental verification. Of these, the most important, and surprising, is that the stability of a growing finger is controlled by the anisotropy of the surface energy [19].

VII. RESULTS FOR UNSTABLE INTERFACES

To investigate the unstable situation numerically [20, 21], we modified the generic configuration of Fig. 1 so that the two boundary temperatures were set at T_m and $0.2T_m$, respectively, corresponding to a solid at its melting temperature in contact with a supercooled liquid. We again considered lattices of size 1024×128 , large enough that finite-size effects were unimportant for the structures we observed, and employed 100 independent runs of duration 10^4 MCS. As before, runs were begun with a flat interface parallel to the x axis, but this time with 10% of the rows set solid. The initial demon-energy distributions were chosen to be consistent with uniform temperatures of T_m and $0.2T_m$ in the solid and liquid, respectively.

Typical results for the development of the interface with time are shown in Fig. 7 [20]. Despite the presence of roughening fluctuations reminiscent of the equilibrium configurations discussed earlier, it is easy to see that, even at very early times ($t \sim 10^3$ MCS), the interface

FIG. 7. The unstable evolution of an initially planar interface in a thermal gradient. The configurations shown are after 0, 2000, and 10 000 MCS. Fluctuations of the magnetization in the bulk phases show clearly that the temperature in the solid (black) phase is higher than that in the liquid phase. Note, however, that there is a marked temperature gradient in the liquid near the interface.

shows precursors of the dendritic structures which form more fully at a later stage. In particular, the characteristic spacing between dendrites, approximately 70 lattice spacings, is already evident at early times. This suggests that all of the data are characteristic of an asymptotic regime, where we expect simple scaling laws to apply. As a first step towards a scaling analysis, Fig. 8 demonstrates that after an initial period of rapid growth, the average length of the interfaces, s , increased at a constant rate. In view of the roughness of the interfaces, the precision with which this rate was defined is quite remarkable. By contrast, the tip velocity of typical dendrites was difficult to determine with any precision, although a value

$v \sim 6 \times 10^{-3}$ per MCS seemed typical.

It is useful to compare our data with the predictions of simple theory for the late-stage regime. Agreement is good. To see this, we estimate in Appendix C that the capillary length $d_0 \sim 1$, the diffusion length $l \sim 100$, and the undercooling $\Delta \sim 0.8$. Thus, if we calculate the characteristic wavelength using

$$\lambda = 2\pi/k_0 = 2\pi(d_0\ell)^{\frac{1}{2}} \quad (12)$$

we obtain $\lambda \sim 60$, compared with 70 for the simulation.

Similarly, if we calculate the tip velocity using the simple expression

FIG. 8. The total perimeter length s of an unstable interface as a function of time t . The dashed straight line is included to guide the eye. Note the datum at $t = 0$. The data are averaged over 100 runs, and statistical error bars are the size of the data points.

FIG. 9. The power spectra at different times for an unstable interface. Data are shown after 1000, 5000, and 10 000 MCS. At large k , the spectra show $1/k^2$ behavior, as indicated by the straight lines. For clarity, the spectra have been displaced vertically by factors of 1, 1.5, and 4 respectively. The data are averaged over 100 runs.

$$v = \frac{D}{R} \left(\Delta - \frac{2d_0}{R} \right), \quad (13)$$

taking the diffusion constant $D = v_0 \ell / 2$, we obtain $v/v_0 \sim 4$. Since v_0 is the velocity of those parts of the interface which show no instability, we were able to estimate it as around 0.5×10^{-3} per MCS. This value gives $v/v_0 \sim 10$, which is quite consistent.

Further insight comes from the power spectrum $P(k)$ of fluctuations of the interface, shown in Fig. 9 as a function of wave number k . For this analysis, overhangs of the interface were eliminated by projecting them onto the x axis prior to performing a one-dimensional transform. A complete analysis using a two-dimensional transform did not yield additional useful information at large length scales, because bubbles and overhangs appear on scales comparable to the lattice spacing. Thus, there were significant numerical advantages in the use of the more efficient one-dimensional algorithm. Other methods of spectral analysis, such as using perimeter-length and orientation variables, were also attempted, but again did not give useful additional information.

Figure 9 shows clearly that the unstable interfaces support short-wavelength fluctuations which can be interpreted as roughening: the large- k region of the spectrum exhibits characteristic $1/k^2$ behavior. As far as we can tell, these fluctuations play no direct role in controlling the instability that occurs at smaller wave vectors, where the spectrum shows marked enhancements above the roughening behavior. In contrast to the equilibrium data shown in Figs. 5 and 6, which never exhibit spectral weight beyond the roughening value of $1/k^2$, the unstable data always exceed this value for $k \leq 0.1\pi$. It is tempting to interpret this enhancement within the linear stability analysis discussed earlier, but this is not appropriate because the data, as we have established, correspond to the late-time regime. It is therefore not surprising that an attempt to identify exponents for the exponential growth of individual modes did not succeed. To obtain data for

an analysis of the linear regime is a difficult undertaking which we describe elsewhere [22].

However, precisely because the data corresponds to the late-time regime, an analysis in terms of scaled variables is most appropriate. In Fig. 10 we show that the $k \rightarrow 0$ part of the power spectrum scales with the growing length s of the interface. By factoring out the underlying $1/k^2$ behavior we identify the dominant mode at $k \sim 0.02\pi$, and establish that the “dendritic” structure is growing without significant change of shape and without measurable coarsening, at least during the time of observation. We also note that there is relatively little side branching, although such structure is difficult, in this two-dimensional situation, to distinguish from roughening.

VIII. DISCUSSION

The results presented in this paper establish our algorithm as a powerful new technique for the study of kinetic phenomena, both in equilibrium and nonequilibrium situations. Our results for both the stable and unstable interfaces confirm that it does lead to kinetic behavior characteristic of real physical processes. This is particularly evident in the studies of equilibrium roughening, where we obtain growth exponents and time-dependent power spectra in full accord with analytical expectations. The observation of crossover to model C behavior as the latent heat is increased is not only interesting in itself, but also represents an essential preliminary step for the modeling of the Mullins-Sekerka instability.

Our results for the unstable interfaces are, in our view, the main results of our paper. However, they are also the most difficult to interpret. In part this is because there are few clear cut analytical results with which to compare, but it is also true that the data have inherent limitations. Thus, for example, we are not able to identify a time regime in which linear stability analysis is valid. There is no trace in our data of the exponential growth of modes, even in the unstable region of the power spectra. In retrospect, this is not surprising, because the linear regime will be of very short duration when the interactions between sites are very short range [23]. Indeed, we observe exponential growth only by increasing the range of interaction [22].

A further limitation arises because the kinetic roughening which is observed for the equilibrium interfaces also affects the nonequilibrium data. Thus, it obscures the large-scale structure characteristic of the instability. Although it is possible to distinguish this structure and even to identify the corresponding length scale, as in Fig. 10, it is very difficult to measure tip velocities or to identify what might be side-branching behavior. The precise location of a “tip” or the value of its radius of curvature cannot be obtained in a reliable fashion as a direct consequence of the roughness of the interface. Thus we cannot with our existing data verify conclusively that the relation between tip radius and velocity, nor test the effect of the anisotropy of the surface energy. Similarly, structure such as that just to the left of the center in Fig. 7 cannot be interpreted as a side branch with any degree

FIG. 10. The scaled power spectra at different times for an unstable interface. Data is shown after 5000 MCS (plusses), 7000 MCS (triangles), 8000 MCS (crosses), and 10000 MCS (squares), averaged over 100 runs.

of certainty.

One direction for future work is to develop our computer codes to run in three dimensions. The effects of roughening are known to be much reduced in this case, scaling as the logarithm of the interface length instead of its square root at temperatures above the roughening temperature T_R [10]. It should thus be possible to measure tip velocities and radii, and to disentangle side-branching behavior from numerical noise. The computational effort, of course, is much increased, since systems with many more sites must be used in order that finite-size effects be minimized. Such calculations are underway, and will be reported shortly.

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APPENDIX A: IMPLEMENTATION OF THE ALGORITHM

As in any Monte Carlo algorithm, we proceed by accessing lattice sites one by one. Conventionally, the order in which sites are visited is random, but a more efficient scheme is a predetermined series of strides through the system: a simple example might be a checkerboard where strides of 2 are taken. Such a scheme has been employed for equilibrium studies, typically on vector type computers [24]. To avoid unphysical dynamical correlations, we use strides large enough that no flipped site will be within a specific range of any other on a given pass through the system. Choosing the stride series randomly provides sufficient mixing of the states to maintain a Markov-like process. On long-time scales, this introduces no significant correlations, and comparisons with completely random visitation show no detectable differences in any of the measured quantities.

Most of our simulations take place at temperatures well below T_c of the pure Ising model. Thus, the rate of spin flips and of the diffusion of energy is very slow. To enhance these rates, a spin which is attempting to flip will alternately access one of the demons of its nearest neighbors. This increases the rate of energy redistribution, in some cases by as much as a factor of 10, and also smooths the demon distribution, permitting improved temperature measurement.

Another important aspect of the algorithm is the implementation of the degeneracy δ , which, as described in the main text, can have any value, integer or noninteger, that is greater than unity. The simplest method computes a random number r in the range $[0, 1 + \delta]$, and permits a flip towards the nondegenerate $\sigma = +1$ state if $r < 1$. A flip in the other direction is permitted if $r > 1$. In practice, the procedure is optimized by always trying to flip a spin for x tries, and then only trying to flip to-

wards the degenerate state for $(\delta - 1)x$ tries. x is chosen to have a convenient value such as 100.

In view of our concern with dynamics, a critical feature of our algorithm is the manner in which we control initial nonphysical behavior, by adjusting the initialization of the spin and demon distributions. In particular, because the demons can represent a significant fraction of the total energy, it is important to use initial conditions where the spin and demon systems are in local equilibrium with each other. Such conditions minimize the duration of transient behavior not related to the dynamics of models A or C .

Initializing the spin system is usually done by randomly distributing spins using values for $\langle m \rangle(T)$ from mean-field expressions for the modified Ising model [8], or from the Onsager solution for the unmodified case [25]. Alternatively, the spin system can be initialized by operating on it with a Metropolis-type algorithm for a reasonable period of time, flipping each spin according to the usual rules for a given T .

The demon system is more difficult to initialize since it is directly coupled to the spin system, and should therefore reflect local temperatures as well as having an overall Boltzmann distribution. The simplest case is that of the unmodified Ising model. Here, the minimum unit for the exchange of energy between spins and demons is $2J$, so that if the demon energy is not initially distributed in units of $2J$, there will be a residue of energy which will never be used. By contrast, when $\Delta \neq 0$, the range of possible demon values can become nearly continuous. In practice, the initial demon energy is set by visiting each demon and choosing a random value for its energy (to the nearest unit exchange energy) statistically based upon its intended temperature. Any small discrepancies are smoothed out soon after the simulation commences, typically after the first 200 MCS.

APPENDIX B: SURFACE DYNAMICS FOR MODEL C

The continuum equations [2] that describe an almost planar interface between two phases are the Gibbs Thomson equation

$$u = -d_0\rho, \quad (\text{B1})$$

the bulk heat-diffusion equation (for which D could be different in the two phases)

$$D\nabla^2 u = \partial u / \partial t, \quad (\text{B2})$$

and the equation which describes the release of latent heat at the interface as the interface moves

$$v_n = D[\nabla u_{\text{solid}} - \nabla u_{\text{liquid}}] \cdot \hat{\mathbf{n}}. \quad (\text{B3})$$

The variable u (the dimensionless undercooling) represents the temperature field. It is defined as

$$(\Lambda/C)u = T - \Lambda/C, \quad (\text{B4})$$

so that it is measured in units of Λ/C (the ratio of latent heat to specific heat), and represents the deviation of the

temperature from the value Λ/C . The vector $\hat{n}$ represents the normal to the interface, and the variable v_n is the velocity of the interface in this normal direction. ρ is the curvature of the interface.

In the linear regime, it is assumed that the interface is almost planar, almost along the x axis in the x - y plane, so that the Fourier transform of its position at time t can be written in terms of modes

$$\xi \sim \xi_k \exp(ikx + \omega t), \quad (\text{B5})$$

where ξ_k is small. In the same spirit, the temperature can be written as

$$u \sim u_k \exp(ikx \pm qy + \omega t), \quad (\text{B6})$$

where the plus sign refers to the solid phase and the minus sign to the liquid. Again, u_k is small.

Substituting into the 3 equations (B1)–(B3) yields, respectively,

$$u_k = -d_0 k^2 \xi_k, \quad (\text{B7})$$

$$D(-k^2 + q^2) = \omega, \quad (\text{B8})$$

and

$$\omega \xi_k = 2Dqu_k. \quad (\text{B9})$$

These three algebraic equations have a solution if

$$\omega = -1/\tau(\tilde{k}), \quad (\text{B10})$$

where

$$\tau(\tilde{k}) = d_0^2 [(1 + \tilde{k}^2)^{1/2} + \tilde{k}]/2D\tilde{k}^3 \quad (\text{B11})$$

and where the dimensionless wave vector $\tilde{k}$ is defined to be $k d_0$.

The expression (B11) should be compared with that derived by Zia *et al.* [16] by their method of dynamic functionals. In the limit when the densities of their two phases are equal (which corresponds in the lattice-gas situation to equal magnitude of the magnetization in the two phases at T_m) their expression is identical to ours. The only modification that we have made to Langer's derivation is the inclusion of the time derivative in (B2) and therefore the ω term in (B8). This term is important when the diffusion of heat away from the interface happens on a time scale comparable to that for the motion of the interface. It gives rise to the correct large- k behavior, where the dynamics are dominated by roughening fluctuations.

APPENDIX C: TYPICAL MODEL PARAMETERS

Melting temperatures T_m were chosen around $0.9T_c$ (of the Ising model) to enhance the dynamics, and undercooled temperatures in the unstable systems were nor-

mally around $0.1T_c$. Field strengths were of order of $\Delta \sim J$, so that degeneracies were in the range 2–5. With these values, typical values of the capillary length d_0 and the diffusion length ℓ can be derived.

As defined in the text, the capillary length is

$$d_0 = \sigma T_m C / \Lambda^2, \quad (\text{C1})$$

where σ is the surface tension, T_m the melting temperature, C the specific heat capacity of the bulk phases, and Λ the latent heat. Of these parameters, T_m and Λ are known, σ can be measured directly by simulation, and C can either be measured by simulation or computed using the mean-field expression

$$C = dE/dT = \ln[\delta(1+m)/(1-m)]^2 / [4/(1-m^2) - 4\alpha J/T]. \quad (\text{C2})$$

The values in the table are based upon measured values for σ and mean-field values for C :

T_m	Λ	δ	C	σ	d_0
0.4	0.5	1.33	0.066	0.98	0.235
0.9	0.5	1.17	0.87	0.42	2.99
0.4	5.0	17	0.36	0.98	0.0128
0.9	5.0	4.7	3.78	0.42	0.130

Qualitatively, these small values of d_0 are all of the order of one lattice spacing.

The diffusion length is defined to be $\ell = 2D/v$, where v is the measured velocity of the unstable interface and D is the diffusion constant. In the limit of $\ell \rightarrow \infty$, the time scale of thermal diffusion is much shorter than that for the motion of the interface, and one approaches the “quasistatic” limit. The diffusion length can be measured in a number of ways. For example, the thermal field in front of an advancing stable planar interface is predicted to have the form

$$T(z) \sim \exp(-2z/\ell),$$

so that we can measure the gradient in a near steady-state configuration to extract ℓ . Alternatively, simulation studies can be undertaken on bulk systems (no interface) to find a value for D . In one such study, we measure the diffusion of a “hotspot” in a bulk system which is otherwise in equilibrium. Straightforward analysis using the diffusion equation shows that the temperature near the perturbation has the form

$$T(r) \sim [\exp(r^2/4Dt)]/(Dt)^{1/2},$$

where $r = 0$ is its origin. In another approach, we measure the thermal conductivity κ [7] and calculate D from $D = \kappa/C$. Regardless of the method, ℓ is consistently found to be of the order of 100 lattice units in the systems we have studied.

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FIG. 1. Schematic diagram showing the rectangular geometrical arrangement for the simulations of interface dynamics. There are L sites along the x axis and M along the y axis. The boundary conditions are periodic across the boundaries AC and BD , but not across AB and CD .

FIG. 7. The unstable evolution of an initially planar interface in a thermal gradient. The configurations shown are after 0, 2000, and 10 000 MCS. Fluctuations of the magnetization in the bulk phases show clearly that the temperature in the solid (black) phase is higher than that in the liquid phase. Note, however, that there is a marked temperature gradient in the liquid near the interface.