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Dynamical Monte Carlo study of crystal growth in a solid-on-solid model

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We have studied crystal growth using Monte Carlo simulations, with Glauber dynamics, of a two-dimensional solid-on-solid model with nearest-neighbor interactions. An applied field (chemical potential) produces a nonequilibrium state, leading to growth of one of the two phases. We construct a relation between the Monte Carlo "time" and the real time by comparing the simulated growth rate with the Wilson-Frenkel growth rate in the noninteracting limit. Simulations of the interacting system are then done for a broad range of temperatures and driving forces, with emphasis on the temperature regime close to the roughening transition. The results are compared with various theories of crystal growth and with experiments on the growth of ^{4}He crystals.

I. INTRODUCTION

The Metropolis Monte Carlo (MC) method has been widely used[1] to study equilibrium properties, especially phase diagrams, structures, and transitions, of thermodynamic systems. It is also very useful[1] for the simulation of kinetic phenomena such as phase separation, wetting film growth, etc. However, because of the unknown relation between the Monte Carlo "time" and the real time, the method is difficult to employ reliably in a study of the temperature dependence of dynamical quantities.

Crystal growth has been studied extensively by theorists,[2,3] and some quantitative experiments have been done recently.[4] Much of the theoretical understanding is based on the kinetic solid-on-solid (SOS) model.[2] The growth is usually divided into three regimes: (1) the two-dimensional nucleation regime below the roughening temperature (T_R); (2) the rough growth regime above the roughening temperature; and (3) a crossover regime for $T \approx T_R$. The roughening transition is blurred as a consequence of dynamical effects.

In this paper we report results from Monte Carlo simulations of crystal growth in the SOS model, including the variation of growth rate with temperature. One of our primary goals is to explore the possibility of applying standard MC methods to investigate dynamical quantities. It should be pointed out that extensive MC simulations of the SOS model, and various kinetic theories based on it, have been done previously.[2,5-7] What is new in our work is the attempt to find the relation between the real time and the Monte Carlo "time" in direct simulations of the model and the application of this relation to a systematic study of the crystal-growth rates. In the latter, we especially emphasize comparison with nucleation theory below the roughening temperature.

By comparing the simulated growth rate at zero interparticle coupling with the Wilson-Frenkel[8] growth rate under the same conditions, we are able to determine how an interval of the real time t and an interval of the Monte Carlo time τ, measured in Monte Carlo steps (MCs's) per site, are related at each value of the temperature T and the chemical potential μ. To the extent that the relation between t and τ is not significantly altered when the interparticle coupling is introduced, and this is the essential assumption in our work, we are then able to calculate from MC simulations such dynamical quantities as the T and μ dependence of the crystal-growth rate. The simulations are simple to perform, and we are able to study the growth in various regimes systematically.

The results are compared to various existing theories, simulations, and experiments. For example, we compare with the two-rate growth theory of Weeks, Gilmer, and Jackson,[5] and with their kinetic MC simulations. Also, below T_R, we determine the behavior of the step energy by fitting the growth rate to the predictions of two-dimensional nucleation theory.[7,9] Finally, we can make a comparison with recent experimental work[4] on ^{4}He and with the related renormalization analysis of Nozières and Gallet.[10]

Section II describes our model and the manner in which the MC time is related to the real time; Sec. III contains results and comparison of theory and experiment; and Sec. IV is a summary.

II. MODEL AND MONTE CARLO METHODS

We employ a solid-on-solid model with nearest-neighbor ferromagnetic coupling J on a two-dimensional square lattice of size $L \times L$, lying parallel to the crystal surface; h_i measures the height of the crystal at site i, and h_i can be any integer. The Hamiltonian is

$$H = J \sum_{\langle i,j \rangle} |h_i - h_j| - \sum_i \mu h_i \ , \tag{1}$$

where $\langle i,j \rangle$ denotes that the sum is over nearest-neighbor pairs of sites. The chemical potential provides the driving force that causes the h_i's to increase for $\mu > 0$; in equilibrium, $\mu = 0$. The roughening temperature for this model[11] is $T_R \simeq 1.24 J/k$; k is Boltzmann's constant.

All simulations are done at fixed temperature and (positive) chemical potential. The initial condition is $h_i = 0$ for all i. To obtain smooth values for the rate of growth, we average over about 20 runs in each case. The lattice size L was typically 20–40. The standard Metropolis MC method with Glauber dynamics,[9,10] was used. A site i was selected at random, and then a decision was made, based on the value of a random number on [0,1] relative to $\frac{1}{2}$, whether to attempt an increase or a decrease in the height h_i by one unit. The attempted change was accepted with a probability (choice 1)

$$w=\tfrac{1}{2}[1-\tanh(\delta\varepsilon/2kT)]\ , \tag{2}$$

or (choice 2)

$$w=\begin{cases}\exp(-\delta\varepsilon/kT) & \text{if } \delta\varepsilon>0\\ 1 & \text{otherwise}\ ;\end{cases} \tag{3}$$

$\delta\varepsilon$ is the energy change produced by the attempted change in h_i. The MC procedure is not the real dynamical process, as is clear from the fact that the choice of w is not unique. The procedure will produce the true equilibrium thermodynamic state of the system if w obeys the detailed-balance condition[12] and allows all microstates to be sampled. Both recipes for w, i.e., Eqs. (2) and (3), obey these conditions and both are frequently employed in simulations; the first is most often used for kinetic properties and the second, for equilibrium properties.

In order to find the real temperature dependence of dynamical quantities one must know how the MC time is related to the real time. For both choices of w, we obtain these relations in the limit $J=0$ and then use them with $J\neq 0$. Thus, the crucial assumption is that when a nonzero interparticle coupling is introduced, the effect on the relation between t and τ is not sufficient (although there will certainly be some effect) to be of any consequence for our results. The validity of this assumption is supported by the agreement of our simulations with earlier work.[5] Also, if our procedure is a reasonable one, computed growth rates would be more or less independent of the choice of w, e.g., Eq. (2) or (3). We have tested this point by doing two sets of simulations, one for each choice of w, in selected cases. Specific examples are given in Sec. III.

The location of the interface is found from the height Γ of the crystal surface,

$$\Gamma(\tau)=(1/N)\sum_i h_i(\tau)\ , \tag{4}$$

where $N=L^2$ is the number of sites in the lattice. The crystal-growth rate in Monte Carlo time τ, R_τ, is simply the derivative of Γ with respect to τ,

$$R_\tau=d\Gamma/d\tau\ , \tag{5}$$

which is, after an initial transient regime, a constant for given T and μ.

To convert R_τ to the real growth rate in physical time R_t, defined by

$$R_t=d\Gamma/dt\ , \tag{6}$$

we have to understand the relation between τ and t. It is known[13] that for an Isinglike stochastic system

$$\tau=f(\mu,T)t\ , \tag{7}$$

although knowledge about $f(\mu,T)$ is very limited. At $J=0$ we compare the growth rate from simulations to the Wilson-Frenkel rate[8]

$$R_{\rm WF}=k_+(1-e^{-\mu/kT})\ . \tag{8}$$

The latter should be exactly R_t in this limit, and thus we may extract $f(\mu,T)$ from the comparison. In Eq. (8),

$$k_+=k_{\rm eq}\exp(\mu/kT)$$

is the deposition rate in the kinetic SOS model;

$$k_{\rm eq}=\nu\exp(-4J/kT)$$

is the equilibrium ($\mu=0$) deposition rate, with ν constant. Thus we find $f(\mu,T)$ in the $J=0$ limit, aside from a constant, from

$$R_{\rm WF}=f(\mu,T)R_\tau\ . \tag{9}$$

III. RESULTS

We begin with the relation between t and τ. For $J=0$, one can easily calculate R_τ for any choice of w. In particular, for choice 1 [Eq. (2)] of w it is

$$R_\tau=\frac{1}{2}\left[\frac{e^{\mu/kT}-1}{e^{\mu/kT}+1}\right]\ , \tag{10}$$

and for the second choice,

$$R_\tau=\tfrac{1}{2}(1-e^{-\mu/kT})\ . \tag{11}$$

Use of Eqs. (8) and (9) immediately yields, for choice 1,

$$\tau=2k_+(1+e^{-\mu/kT})t\ . \tag{12}$$

The factor of 2 is a consequence of the fact that in the simulations we attempt an increase (as opposed to a decrease) of a column height in one-half of the attempted moves. For choice 2 we find

$$\tau=2k_+t\ . \tag{13}$$

We turn now to presentation of the growth rates R_t with $J\neq 0$, determined from simulations and the appropriate t-τ relation, i.e., Eq. (12) or (13). We drop the subscript on R; hereafter R always denotes the real growth rate R_t. Our first concern is with the dependence of R on the choice of w. In Fig. 1 we show R/k_+ versus kT/J for $\mu/J=2.0$ and 0.7, using w given by both Eqs. (2) and (3). The general behavior of R for a given μ is the same in both cases, although there are differences for the larger μ, in particular when T is somewhat above the roughening temperature. In fact, in all comparisons we have done, this is the regime of μ and T where the difference between the two predictions is the largest, about 25%.

The only qualitative difference we have found also appears for $\mu/J\approx 2$, but at temperatures well below T_R; here, there appears to be a cusp in the curve showing $R/R_{\rm WF}$ as a function of μ/J for the second choice of w,

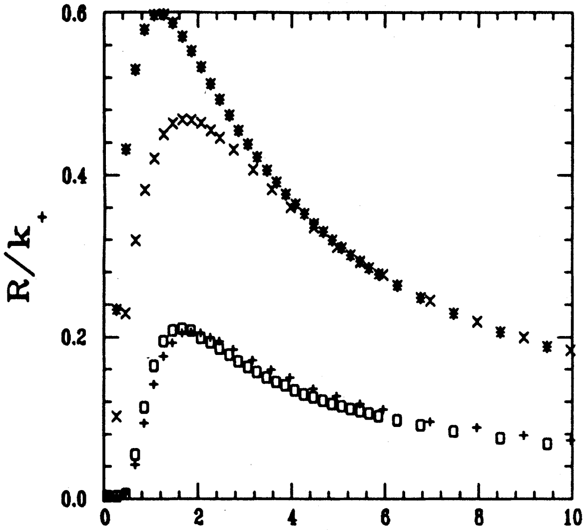

FIG. 1. The simulated growth rate R/k_+ is plotted vs kT/J for $\mu/J=2.0$ (×), and 0.7 (+) using choice 1 of w, and for $\mu/J=2.0$ (∗), 0.7 (□) employing choice 2 for w.

but not for the first. We suspect that this is a consequence of the nonanalytic behavior of this particular w at $\delta\varepsilon=0$. Our belief is that of the two choices, the first is more physically appropriate, at least in that it gives a nonzero probability of not accepting a configuration change with $\delta\varepsilon=0$; nevertheless, our simulations show no significant qualitative difference between the results from the two choices, aside from the one just described. All of the results that follow were obtained using Eq. (2) for w and, of course, Eq. (12) for f.

Figure 2 shows R/k_+ as a function of μ/kT for several different fixed temperatures $kT/J=2.0$, 1.3, 1.0, and 0.7. Also shown is the Wilson-Frenkel growth rate, which should obtain in the high-temperature limit. The other solid lines are fits of the two-rate model[5] to the simulations. This model predicts

$$R/k_+=\frac{2\sinh(\mu/kT)}{e^{\mu/kT}+\cosh(2\alpha J/kT)} . \tag{14}$$

Although α should generally be a function of T and μ, we have chosen $\alpha=1.57$ throughout. This value is such that the simulated value at $\mu/kT=2.0$ and $kT/J=1.3$ is the same as that given by Eq. (14). Our results are in good agreement with simulations of the kinetic SOS model in Ref. 5, where a similar fit to the two-rate model was also done. The quantitative discrepancy in the low-temperature and low-driving force region is not surprising, since Eq. (14) is derived using the continuum approximation.

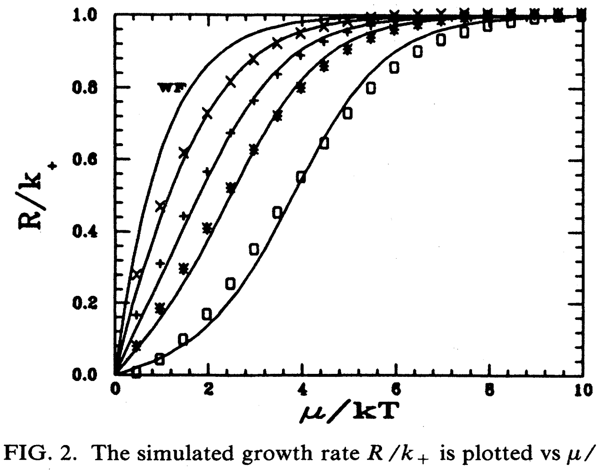

FIG. 2. The simulated growth rate R/k_+ is plotted vs μ/kT for $kT/J=2.0$ (×), 1.3 (+), 1.0 (∗), and 0.7 (□). Also shown (solid lines) are fits of the two-rate model of Ref. 5 to the simulations and the Wilson-Frenkel growth rate.

The reduced rate R/R_{WF} is shown in Fig. 3 for $\mu/J=2.0$, 0.7, and 0.05. For the smallest μ, the reduced rate increases sharply around the roughening temperature $T_R=1.24J/k$ from nearly zero to a large value. As the driving force μ increases, this consequence of the roughening transition is blurred, and the regime with a nearly zero reduced rate shrinks. At high temperatures, the reduced rates for different μ tend to fall on the same curve and finally go to unity, which implies that the crystal surface is completely rough. This behavior has been observed[4] in crystal-growth experiments on ^{4}He and has been studied using renormalization analysis;[10] see Fig. 5 of Ref. 4. Regrettably, detailed comparison of experiment and simulations is not possible. The excess chemical potential in the experiments, expressed in our units, is around $10^{-5}J$. We are able to do simulations only for chemical potentials larger than about $0.05J$; for smaller ones, the growth rate is so small as to require unacceptably long runs.

Figure 4 displays the growth rate R/k_+ as a function of μ/J for relatively small μ at several temperatures ranging from above to below T_R. As is well known, the growth rate behaves linearly, with driving force above T_R and nonlinearly at $T<T_R$, where the growth mechanism is described by 2D nucleation.[3]

The low-temperature growth rate may be fit to two-

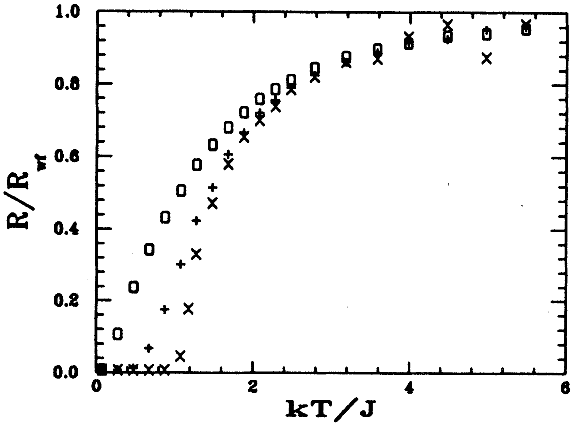

FIG. 3. The simulated reduced growth rate R/R_{WF} is plotted vs kT/J for $\mu/J=2.0$ (□), 0.7 (+), and 0.05 (×).

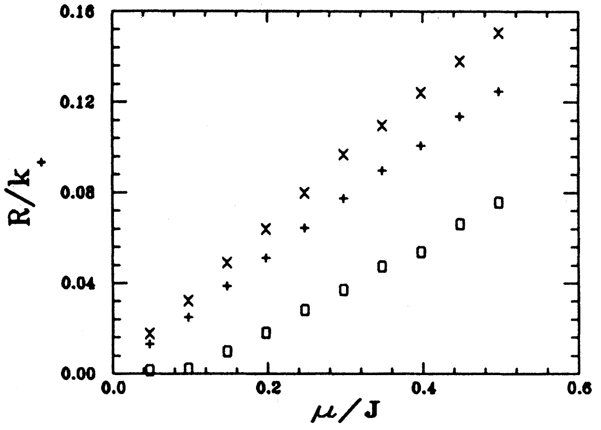

FIG. 4. The simulated growth rate R/k_+ is shown as a function of μ/J for $kT/J=2.0$ (×), 1.3 (+), and 1.0 (□) for relatively small driving force μ.

dimensional nucleation theory. For small μ, this theory predicts[7,9]

$$R/k_+ \sim g(\mu) e^{-\pi\beta^2/3kT\mu} , \qquad (15)$$

where β is the step energy and $g(\mu) \sim \mu^\theta$ with $\theta=\frac{5}{6}$. Figure 5 displays plots of $\ln[(R/k_+)(J/\mu)^\theta]$ versus J/μ at $T=1.05J/k$ for $\theta=\frac{5}{6}$, and also for $\theta=1$, a value obtained[3] by simply modifying the Wilson-Frenkel theory with the nucleation rate which is proportional to $\exp[-\pi/\beta^2/(3\mu kT)]$. On the basis of this figure, and similar ones at other temperatures, it appears that the simulations better fit $\theta=1$. Also, from the slopes of these lines and Eq. (15), we can determine the step energy. We find that if we assume $\theta=\frac{5}{6}$, the step energy does not approach zero as $T \rightarrow T_R$ but rather remains finite at considerably higher temperatures. For $\theta=1$, on the other hand, the step energy does go to zero close to T_R. Thus our results are clearly more consistent with $\theta=1$. The same is true of the experiments on the growth of ^{4}He crystals.[4] In Fig. 6 we plot, using $\theta=1$, $-\ln(\beta/J)$ versus $(1-T/T_R)^{-1/2}$ assuming $T_R=1.24J$. The points for T close to T_R can be reasonably fit to a straight line, in agreement with the prediction of Kosterlitz-Thouless theory,[14] which is appropriate for the two-dimensional SOS model,[11]

$$\beta \sim e^{-D(1-T/T_R)^{-1/2}} ; \qquad (16)$$

here, D is a constant independent of T.

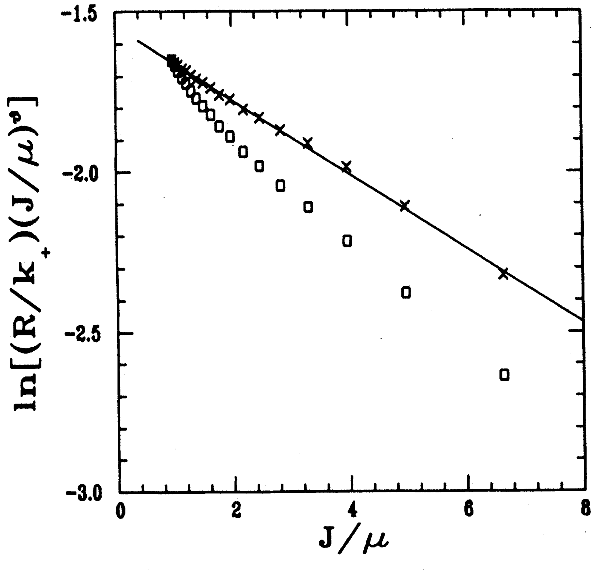

FIG. 5. We plot simulated values of $\ln[(R/k_+)(J/\mu)^\theta]$ for $\theta=\frac{5}{6}$ (□) and 1 (×), vs J/μ at $T=1.05J$. The straight line is a least squares fit to the simulations.

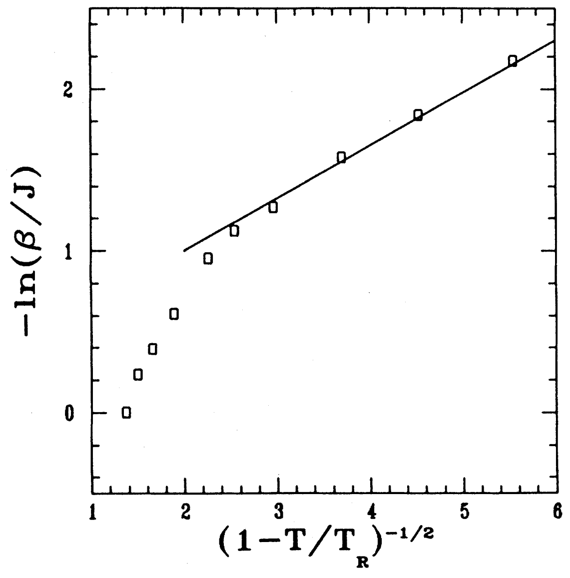

FIG. 6. The negative logarithm of the step energy in units of J, inferred from the simulations and Eq. (15) with $\theta=1$, is plotted vs $(1-T/T_R)^{-1/2}$; the straight line serves to guide the eye.

IV. SUMMARY

In this paper we have presented results from Monte Carlo simulations of crystal growth, both above and below the roughening temperature, using a two-dimensional solid-on-solid model with Glauber dynamics. The simulations may be compared with real crystal growth provided one knows the relation between the Monte Carlo time (Monte Carlo steps per site) and the real time. This relation will naturally depend on the recipe one uses to determine whether an attempted Monte Carlo step is accepted. We have considered two standard choices, Eqs. (2) and (3), and have extracted relations between the Monte Carlo and real times, given by Eqs. (12) and (13), by comparing the growth rate which would emerge from simulations of the SOS model, with zero intercolumn coupling with the Wilson-Frenkel growth rate. We then *assumed* that these relations remain valid also for $J \neq 0$. This idea was tested by comparing results for R

(with $J \neq 0$) using both choices of w (see Fig. 1). Although the results in the two instances are not the same, differences are in general neither large nor qualitative, suggesting that it is possible to study dynamical properties with MC simulations. Additional results reported here were obtained using the first choice of w, although the second choice was also employed in some cases as a check that the choice of w does not make a significant difference.

We have compared our simulated growth rates with the two-rate theory of Weeks *et al.*, and with their kinetic MC simulations,[5] finding good agreement. Our simulations also show that the roughening transition is increasingly blurred with increasing μ by dynamical effects. This behavior was also observed in experiments[4] on the growth of solid ^{4}He and found in the related renormalization analysis of Nozières and Gallet.[10] However, quantitative comparison with these experiments is not possible because of computer time limitations. The simulated growth rate at low temperatures is fit to two-dimensional nucleation theory.[7,9] We find (in agreement with the experiments[4]) that the dependence of R on the chemical potential is slightly different from the prediction. Finally, from the comparison with nucleation theory, we have extracted the step energy of the model as a function of temperature and found that for $T \rightarrow T_R$ from below, its logarithm is consistent with variation as $(1-T/T_R)^{-1/2}$, the prediction of Kosterlitz-Thouless theory,[14] which is appropriate for the SOS model in two dimensions.[11]

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