RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 35, NUMBER 7 1 MARCH 1987

Monte Carlo simulation of the growth of wetting layers

K. K. Mon, K. Binder,* and D. P. Landau
Center for Simulational Physics, University of Georgia, Athens, Georgia 30602
(Received 2 September 1986)

We present the first Monte Carlo study of the dynamics of the formation of the wet phase far from the wetting transition for a short-range attractive wall potential. A nearest-neighbor, simple-cubic, ferromagnetic Ising lattice gas with single spin-flip dynamics in a slab geometry ($L \times L$ cross section) is used. We show that there are two distinct dynamical regimes with crossover from fluctuation-dominated logarithmic growth for large L and short time t to quasi-one-dimensional diffusive growth for small L and large t. For any finite system, the long-time behavior {i.e., $t \gg L^4[\ln(L)^2]$} is predicted to always be quasi-one-dimensional diffusive in nature.

The physics of interfaces between coexisting phases represents an important topic of broad fundamental and applied interest.[1-8] Considerable current research focuses on the influence of external perturbations, such as the interactions introduced by the presence of walls (or surfaces) which preferentially favor formation of one phase over the others.[2] Depending on the strength and range of these perturbations, a wetting phase transition has been predicted theoretically[3] and observed experimentally in systems involving fluids and fluid mixtures.[8] These wetting transitions are also related to the delocalization of interface or domain wall depinning[2] in magnetic materials.

The static and dynamic nature of these phase transitions has been subjected to extensive investigation including mean field theory,[2,4,5] renormalization group,[2,6] Monte Carlo simulations,[2,7] and experiments.[2,8] In this paper, we present, to our knowledge, the first Monte Carlo study of the wet phase far from the wetting transition for short-range attractive wall potential.

We consider a nearest-neighbor ferromagnetic Ising model on a simple-cubic lattice with a geometry of $L \times L \times 40$ for $L = 8-24$. There are two free (100) surfaces and periodic boundary conditions in the remaining two directions. The exchange constant is J in the bulk and J_s in the surface planes. The bulk magnetic field is H with a field $H+H_1$ and $H+H_{40}$ acting on the top and bottom surface planes, respectively. We have considered the case with $J/kT=0.25$, $J_s/J=0.5$, $H=0$, $H_1/J=-5.0$, and $H_{40}/H_1=-3.5$. The precise values of the surface fields are not crucial but are chosen to ensure the formation of a single interfacial profile which matches smoothly into the bulk values as the bottom surface is approached. These parameters also put the system in the wet phase far from the estimated transition value of $-H_1/J=(1.3-1.4)$. Under these conditions, the wetting transition is continuous (critical). Note also that the bulk system with $J/kT=0.25$ is above the roughening transition temperature[9] ($J/kT_r=0.4$). Thus, the interface in our study is rough and the dynamics is not expected to be dominated by the layering transitions.[2]

The initial configuration is always a uniformly ordered state, modeling a bare substrate. Standard single spin-flip updates with sites chosen at random are used.[10] As the simulation progresses, a wet layer begins to form and grows, producing a density profile which evolves with "time" t. The profile is recorded and ensemble averaged over many samples to obtain $m(Z,t)$, where Z is distance from the top. The number of samples ranges from 60 to 3000 for times up to 5760 MCS (Monte Carlo steps per site) with the smaller number of samples used for larger lattices and longer times, resulting in larger statistical errors. Our results also indicate the absence of self-averaging which means that we then require a large number of samples even for large system sizes.

The evolution of the ensemble-averaged profile $m(Z,t)$ for $L=8$ is presented in Fig. 1 for $t=20-2400$ MCS. The broadening of the averaged profile is a consequence of the ensemble average of the interfacial motion. One simple measure of the dynamics of film growth is the time dependence of the position $Z_0(L,t)$ for which $m(Z_0(L,t))=0$. A wide range of system sizes ($L=8-24$) is considered and two distinct dynamical regimes with finite size-dependent crossovers are observed. For short time (t), a logarithmic time dependence is observed as shown in Fig. 2. This result is consistent with fluctuation-dominated growth.[2,5] The interfaces, being in the rough phase, contain fluctuations which will lower the free energy by moving away from the surfaces. The amplitudes of these excitations (due to roughening) are expected to be proportional to $\ln(L)$.[1] For $z_0(L,t) \gg A\ln(L)$ (the interface far from the

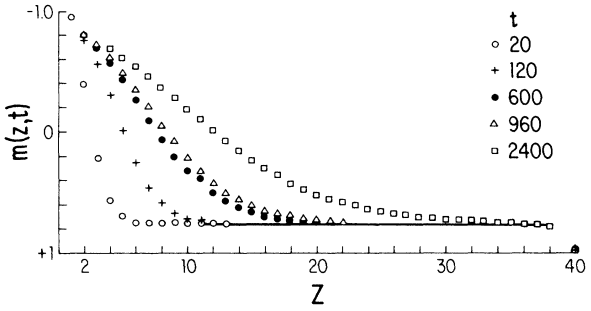

FIG. 1. Time evolution of the ensemble-averaged density profile $m(Z,t)$ for $8\times8\times40$ lattices for t: ○=20, +=120, ●=600, △=960, □=2400 in units of updates per site (Monte Carlo steps per site). The solid line shows the bulk magnetization.

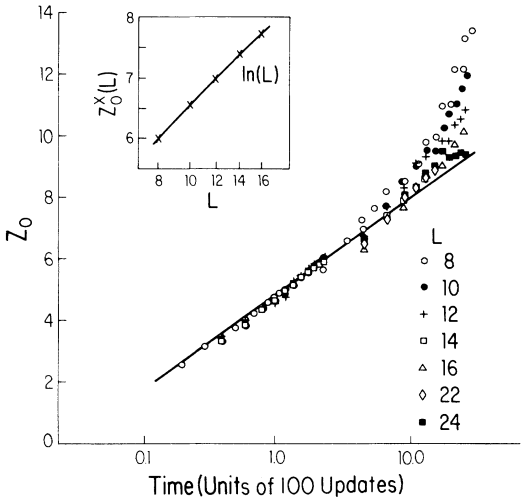

FIG. 2. Ensemble-averaged position of the interface $Z_0(L,t)$ (see text) vs $\ln(t)$ for $L\times L\times 40$ lattices. t is in units of 100 updates per site. The size dependence of the crossover $Z_x^0(L)$ (see text) is given in the inset.

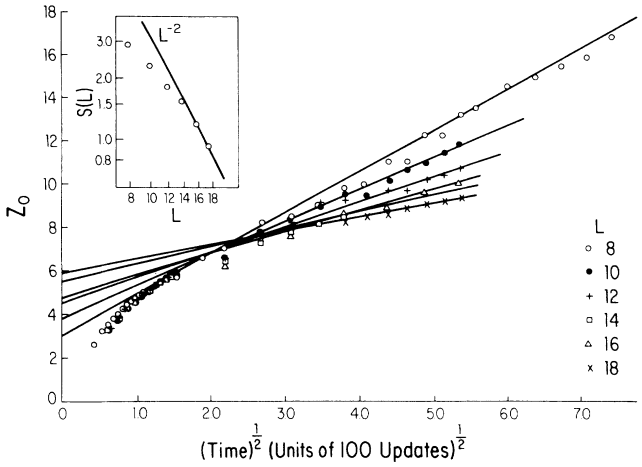

FIG. 3. Ensemble-averaged position of the interface $Z_0(L,t)$ vs $\sqrt{t}$ for $L\times L\times 40$ lattices. t is in unit of 100 updates per site. The size dependence of the slope $S(L)$ is given in the inset. The lines indicate a $1/L^2$ dependence.

surface) the interfacial fluctuations do not interact with the surface and are no longer relevant. The dynamics then crosses over into a quasi-one-dimensional diffusive regime with $Z_0(L,t)\sim\sqrt{t}/L^2$. The $\sqrt{t}$ dependence is a consequence of the interface executing quasi-one-dimensional random-walk behavior with the cross section of the $(L\times L)$ interface playing the role of an effective mass. For diffusion in an infinite one-dimensional system, the average interfacial displacements would be zero by forward and backward symmetry, but the root-mean-square displacements would increase as $\sqrt{t}$. For our system, the presence of the surface breaks the symmetry, resulting in the $\sqrt{t}$ dependence for the average interfacial displacement. Since in this motion L^2 sites move together like a rigidly locked object, we suggest the diffusion constant, which is inversely proportional to the mass of the diffusing object, for this coherent interface motion varies as $1/L^2$. These excitations can be considered to be the realization of the uniform Goldstone modes in the capillary wave spectrum. In this one-dimensional diffusive regime, the probability distribution for the position $Z_0(t)$ is independent of the size of the system. L dependence enters via the diffusion constant of the interface. This implies the absence of self-averaging as discussed recently[11] in the kinetics of domain growth. A large number of samples is thus needed to obtain accurate results even for large system sizes.

The position $Z_0^x(L)$, where the behavior crosses over from $\ln(t)$ to $\sqrt{t}$ is taken to be approximately the point of departure from the straight line in Fig. 2, and the finite size dependence of this point is shown in the inset of the same figure. Although the range of L for which the crossover is observed is limited ($L=8$–18), and $Z_0^x(L)$ can only be estimated within reasonable errors, the behavior observed is consistent with a logarithmic behavior. In Fig. 3, we have plotted the same data to exhibit the $\sqrt{t}$ regime. The size dependence of the amplitude is given in the inset and is consistent with a $1/L^2$ dependence in the large L limit.

One important implication of these numerical results is the role played by the finite size of the system. For any film with thickness $Z_0\gg\ln(L)$ (i.e., for the long-time behavior) quasi-one-dimensional diffusive dynamics is always expected. The logarithmic time dependence is relevant only for thin films or short time t. Analytical results such as mean-field theory always take the thermodynamic limit ($L\rightarrow\infty$) and thus exhibit only the logarithmic time dependence for all time.

Recently, it was shown[12] that a logarithmic growth law $[Z_0(t)\sim\ln(t)]$ can also be obtained from deterministic mean-field theory, i.e., without arguments based on interface fluctuations. The physical argument is as follows: In the wet phase, the interfacial profile at finite distance from the surface produces a mismatch of the order of $\exp[-Z_0(t)/\xi]$ to the boundary condition at the surface where ξ is the correlation length. This mismatch raises the free energy and acts as a driving force to the growth of the wetting layers producing a logarithmic growth law. This type of mechanism is an alternative to the fluctuation-dominated processes in explaining the logarithmic growth. For this process there should not be a dependence on the size of the system (L); however, the size dependence which was observed in the simulations for the range of sizes considered seems to indicate that this latter mechanism is not important.

These results are obtained for a wet phase which is far from the related wetting transition, which is continuous. It is expected to be valid also for a wet phase far from a related first-order wetting transition. The nature of the dynamics near the transition is an important related problem which will be considered elsewhere. There, the order of the transition is expected to play a crucial role in the dynamical properties.

The range of the surface potential has been shown to be relevant in the static[2] and dynamic[2,5] properties of the

wetting transitions. In this paper, only a short-range wall potential is considered, but extensions to long-range potential would be of physical interest. It has been suggested[5] that for long-range van der Waals forces, the logarithmic growth law is replaced by a power law $Z_p(t) \sim t^{1/4}$. We conclude that in this case, in a system of finite extent (linear dimension L parallel to the surface) a crossover to one-dimensional diffusive behavior of the interface would occur at a time of $t \sim L^8$, i.e., for a layer thickness of order L^2, while for the logarithmic growth case, the crossover occurs already for a thickness of order $\ln(L)$ [at a time of $t \sim L^4 \ln(L)^2$]. For the case of conserved order parameter, it was suggested[13] that $Z_0(t) \sim t^{1/8}$, which would already be overtaken by the diffusive one-dimensional motion $Z_0(t) \sim t^{1/2}/L^2$ at a time of $t \sim L^{16/3}$ and a layer thickness of $L^{2/3}$; i.e., then the interface has moved away from the wall a relatively small amount, only $\sim L^{-1/3}$. In general, if one has a growth law $Z_0(t) = t^x$ with $x < \frac{1}{2}$ for $L \to \infty$, crossover should occur at a thickness of $Z_0 \sim L^{4x/(1-2x)}$ (Ref. 14).

Finally, we note that our dynamical results have relevant implications for computer simulations of equilibrium properties of wetting. We have observed the rather long equilibration time needed to form a macroscopic wetting film, with intricate dependence on the system sizes and range of the surface potentials. The absence of self-averaging combined with slow relaxation implies that conclusions drawn concerning static equilibrium wetting properties using limited simulations must be considered with caution.

This research has been supported in part by the Advanced Computational Methods Center and National Science Foundation Grant No. DMR-8603605.

---

*Permanent address: Institut für Physik, Universität Mainz, 6500 Mainz, West Germany.

[1]For reviews, see K. Binder, in *Phase Transitions and Critical Phenomena,* edited by C. Domb and J. L. Lebowitz (Academic, New York, 1983); David Jasnow, Rep. Prog. Phys. **47**, 1059 (1984).

[2]For review on wetting, see P. G. de Gennes, Rev. Mod. Phys. **57**, 827 (1985); E. H. Hauge, in *Fundamental Problems in Statistical Mechanics VI,* edited by E. G. D. Cohen (North-Holland, Amsterdam, 1985), p. 65; D. E. Sullivan and M. M. Telo da Gama, in *Fluid Interfacial Phenomena,* edited by C. A. Croxton (Wiley, New York, 1985); R. Pandit, M. Schick, and M. Wortis, Phys. Rev. B **26**, 5112 (1982); M. E. Fisher, in Proceedings of the Twentieth Faraday Symposium, 1986 (unpublished).

[3]J. W. Cahn, J. Chem. Phys. **66**, 3667 (1977); C. Ebner and W. F. Saam, Phys. Rev. Lett. **38**, 1486 (1977).

[4]R. Lipowsky, D. Kroll, and R. Zia, Phys. Rev. B **27**, 4499 (1983); G. Forgacs, H. Orland, and M. Schick, *ibid.* **31**, 7434 (1985).

[5]R. Lipowsky, J. Phys. A **18**, L585 (1985).

[6]E. Brezin, B. I. Halperin, and S. Leibler, Phys. Rev. Lett. **50**, 1387 (1983); M. P. Nightingale, M. Schick, and W. Saam, Phys. Rev. B **30**, 3830 (1984).

[7]K. Binder, D. P. Landau, and D. Kroll, Phys. Rev. Lett. **56**, 2272 (1986).

[8]C. Franck and S. Schnatterly, Phys. Rev. Lett. **48**, 763 (1982); D. Beysens and S. Leibler, J. Phys. (Paris) Lett. **43**, L133 (1982); M. Moldover and J. Cahn, Science **207**, 1073 (1980); O'D. Kwon, D. Beaglehole, W. W. Webb, and B. Widom, Phys. Rev. Lett. **48**, 185 (1982); D. W. Pohl and W. Goldberg, *ibid.* **48**, 1111 (1982).

[9]E. Burkner and D. Stauffer, Z. Phys. B **53**, 241 (1983).

[10]For general discussion, see *Monte Carlo Methods in Statistical Physics,* edited by K. Binder (Springer, Berlin, 1979).

[11]A. Milchev, K. Binder, and D. W. Herrmann, Z. Phys. B (to be published).

[12]I. Schmidt and K. Binder (unpublished).

[13]R. Lipowsky and D. A. Huse (unpublished).

[14]Note that for retarded van der Waals forces, the growth law is $Z_0(t) \propto t^{1/5}$ (nonconserved case) and $Z_0(t) \propto t^{1/10}$ (conserved case), see Refs. 5 and 13.