Lattice-Gas Interface Structure: A Monte Carlo Simulation

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(Received 10 January 1973)

The (100), simple-cubic, lattice-gas interface structure has been simulated by Monte Carlo methods. For the interface width L we find $L = (1.05 \pm 0.04)(1 - T/T_c)^{-0.69}$, in the range $T \gtrsim 0.5T_c$. This is larger than the bulk correlation length ξ by a factor of ~ 3.

We have recently described the results of a Monte Carlo simulation of the (100) interface between a simple-cubic Ising solid and its vapor.[1,2] In that work, all atoms were required to be connected to the bulk phase through the underlying (100) interfacial layer. This requirement served to eliminate the critical point by maintaining the "solid" and "vapor" densities at 1 and 0, respectively. In this Letter we describe a simulation in which the "solid" phase is not required to be fully dense; i.e., a simulation of the classical Ising or lattice-gas system. The results of this study allow us to assess the effect of variable solid and vapor phase densities on interface structure, and constitute, to our knowledge, the first such results obtained by direct simulation of the interface in a two-phase system near its critical point.

The simulation was accomplished via the classical importance sampling method of Metropolis *et al.*[3] The system consisted of 28 (100) layers of 20×20 dimensions. Periodic boundary conditions were employed within each layer and the top (layer 28) and bottom (layer 1) were maintained with 0% and 100% occupancy, respectively. An initial configuration was produced by filling half of the remaining sites with atoms. A Markov[4] chain of configurations was produced from this initial state by Guttman's[4] pair exchange technique. In this scheme, a virtual exchange of randomly selected "atoms" and "vacancies" is considered. The exchange is executed whenever $\exp(-\Delta n\epsilon/kT)$ is greater than a random number between 0 and 1, where Δn is the change in the number of solid-solid bonds produced by the exchange, and ϵ is the bond energy. In general, 5×10^5 configuration chains were generated for a given ϵ/kT, and equilibrium values were obtained by averaging the results of the final 2×10^5 configurations. A smaller value of ϵ/kT was then selected and the entire process repeated, beginning with the final configuration of the preceding chain. The entire simulation was repeated for a system identical to the first, except that the interface was eliminated by reversing the interactions across a dividing surface between the fourteenth and fifteenth layers. Atoms adjacent to this surface must bond with vacancies on the other side. This scheme yielded two phases corresponding to the bulk solid and vapor which were uniform up to the dividing surface. Interfacial excess quantities were obtained directly by subtracting the values of this system from those of the system containing the interface.

Typical interface-density and energy-density profiles for $\epsilon/kT = 1.0$ ($\epsilon/kT_c = 0.887$)[5] are shown in Fig. 1. As is apparent, the local number density of atoms, X_i, agrees well with the Padé approximate value, X_s, of Essam and Fisher[5] in the region just beyond the completely solid bound-

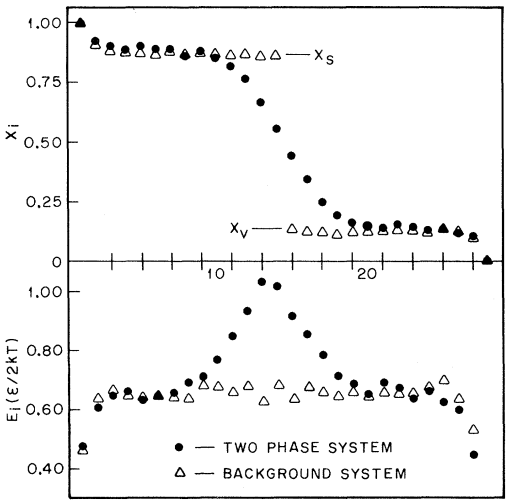

FIG. 1. Density (X_i) and energy density (E_i) profiles for a simple-cubic (100) lattice-gas interface at $\epsilon/kT = 1$. The data points are approximately 2σ in height.

ary layer, and decreases smoothly to the homogeneous vapor phase value $X_V = 1 - X_s$. The local energy density E_i, obtained by counting the number of solid-vacancy nearest-neighbor pairs in each (100) layer, exhibits a peak at the interface position and is symmetrical about $i = 14.5$. Figure 1 also shows that, although the density of the "background" reference system changes abruptly between layers 14 and 15, the energy density is uniform throughout the system.

Following Cahn and Hilliard,[6] the interface thickness may be defined as $L = (X_s - X_V)/(X_{14} - X_{15})$, i.e., as the width given by a straight-line extrapolation of the density profile at mid-density to the solid and vapor homogeneous phase densities. As is well known, $L \propto (T_c - T)^{-f}$ in critical systems. The classical Van der Waals–Cahn-Hilliard[6,7] theory gives $f = \frac{1}{2}$, while Fisk and Widom's[8] nonclassical extension of it gives $f \sim 0.64$. Fisk and Widom's result arises because they equate the interface width, L', with the correlation length ξ in the one-phase nonclassical fluid, for which $f = \nu$, the critical exponent for ξ. (L' as defined by Fisk and Widom is somewhat larger, by a factor of ~ 5, than L as defined here.) Our simulation results are shown in Fig. 2 as a plot of $X_{14} - X_{15}$ versus kT/ϵ, where a linear relationship is clearly suggested. Assumption of linearity leads to $L/a = (1.05 \pm 0.04)(1 - T/T_c)^{-0.69}$, where Essam and Fisher's numerical value, $\frac{5}{16}$, for the $X_s - X_V$ exponent is employed, and a is the lattice parameter. The exponent -0.69 agrees reasonably well with Fisk and Widom's suggestion of -0.64, as well as with the experimental results of Gilmer *et al.*,[9] Huang and Webb,[10] and Zollweg, Hawkins, and Benedek[11] for a binary liquid mixture and pure xenon, respectively. The magnitude of L is found to be larger than $\xi \simeq 0.32 \times (1 - T/T_c)^{-0.64}$ as estimated by Fisher for the Ising model.[12] Moreover, the experimental determinations of L and the theory of Fisk and Widom agree in suggesting that L is somewhat larger than the bulk correlation length ξ. This simulation thus lends support to both. We do however, anticipate that L, as computed by Monte Carlo simulation, will be system-size dependent. In particular, large systems allow long-wavelength surface perturbations, which may cause a dramatic increase in L. We expect that our 20×20-system results are comparable with experiment since the long-wavelength perturbations in a real system are suppressed by gravity.[13] Further discussion of this topic will be included in forthcoming detailed reports of our simulation results for this and several interface orientations in the face-centered-cubic Ising system.

The interfacial excess energies, E, obtained here are compared in Fig. 3 with our previous

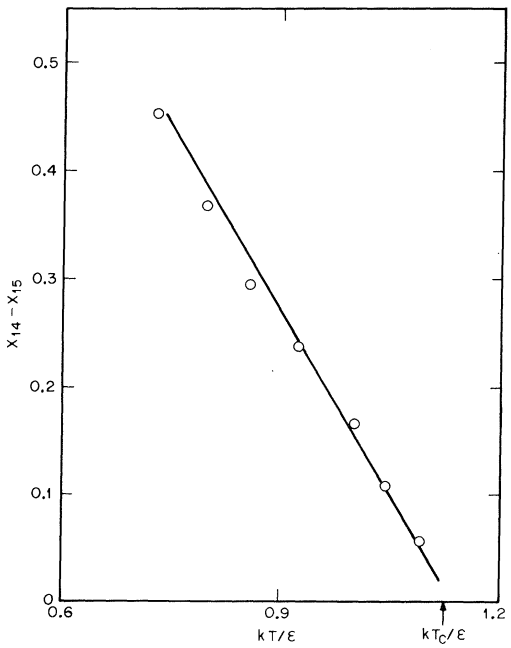

FIG. 2. The kT/ϵ dependence of the maximum density gradient, $X_{14} - X_{15}$, of the simple-cubic, lattice-gas interface. The data points are approximately 2σ in height.

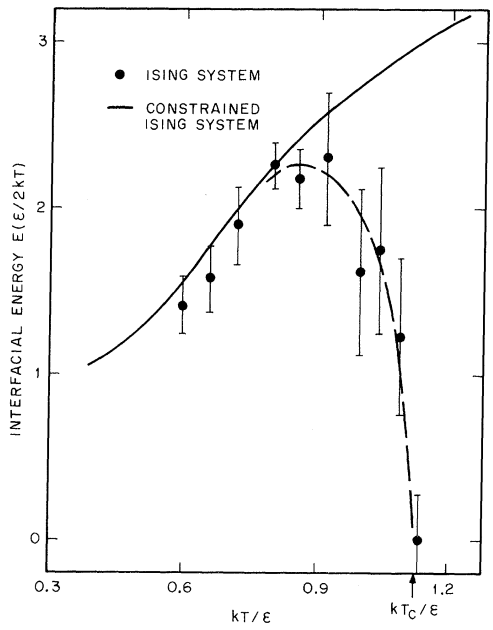

FIG. 3. The kT/ϵ dependence of the interfacial excess energy, E, of the simple-cubic, (100), lattice-gas interface. The error bars are 2σ in height.

results for a system constrained to maintain $X_s = 1$ and $X_v = 0$. As expected, these agree in the region $kT/\epsilon \lesssim 0.8$ where $X_s - X_v \simeq 1$ for the Ising system, but differ considerably for higher kT/ϵ. This behavior is expected since for the constrained system $E \to \infty$ as $\epsilon/kT \to 0$, while for the Ising system, $E \to 0$ as $T \to T_c$. The result of Fig. 3 may be contrasted with that for a two-dimensional lattice-gas interface, for which E decreases smoothly with temperature to a finite value at $T = T_c$.[14] Our results are similar in form to the classical-theory results[6,7]; i.e., E increases to a maximum at some $T < T_c$, and then decreases to 0 at $T = T_c$. The interfacial free energy σ is proportional to $(1 - T/T_c)^{\mu}$ in the Ising system where the exponent μ is given as $\frac{3}{2}$ by the classical Van der Waals–Cahn-Hilliard theory,[6,7] and as 1.22–1.33 by Fisk and Widom's nonclassical treatment. Since

$$E \propto \frac{d\sigma/kT}{d\epsilon/kT},$$

we obtain from the data in Fig. 3 a value for μ between 1.27 and 1.53, in general agreement with both experiment[9-11] and nonclassical theory.[8]

We are indebted to P. C. Hohenberg, F. H. Stillinger, and J. D. Weeks for helpful discussions.

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Critical Thicknesses in Superconducting Thin Films*

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Direct observations of the magnetic-field distribution in thin films of Pb, Sn, and In indicate that the critical thicknesses separating intermediate-state and mixed-state behavior in these systems are accurately given by theoretical calculations and are much lower than has been believed on the basis of critical-field measurements.

A fundamental characteristic of the equilibrium state of a superconductor is the magnetic structure which arises in an applied magnetic field H_0. Depending on the material and geometrical factors and on H_0, the equilibrium state can be perfectly diamagnetic or only paritally diamagnetic as in Abrikosov's "mixed state" of single quantum fluxoids (type-II superconductors) and in the coarser "intermediate state" found in type-I superconductors of finite dimension. The differences between the mixed and intermediate states is fundamental to the distinction between type-I and type-II superconductors, but, as Tinkham[1] pointed out, sufficiently thin films of any material should exist in the mixed state even if thicker specimens of the same material exhibit intermediate-state behavior. One expects, therefore, that thin films made of a type-I material and having a Ginzburg-Landau parameter $\kappa < 1/\sqrt{2}$ will be type-II superconductors for thicknesses below a critical value d_c. Experimental studies of this interesting change in magnetic behavior in thin-film systems have involved macroscopic properties of these systems and have been dif-