

## Anisotropic Surface Tension, Step Free Energy, and Interfacial Roughening in the Three-Dimensional Ising Model

K. K. Mon, S. Wansleben,<sup>(a)</sup> and D. P. Landau

*Center for Simulational Physics, University of Georgia, Athens, Georgia 30602*

and

K. Binder

*Institut für Physik, Universität Mainz, D-6500 Mainz, Postfach 3980, West Germany*

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The anisotropic interfacial tension of a simple-cubic Ising model is calculated for the first time by use of extensive Monte Carlo studies with a tilted interface for a wide range of lattice size  $L$  and temperature  $T$ . The size dependence of the step free energy is used to probe the correlation length near the roughening transition and confirms the expected square-root  $T$  dependence. Finite-size scaling implies that the step free energy varies as  $1/L$  in the rough phase, and thus the applicability of a capillary-wave Hamiltonian to describe interfaces of lattice models may be limited.

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Surface and interfacial phenomena are ubiquitous and constitute a topic of current interest to many research fields, ranging from phenomena such as nucleation, wetting, crystal-faceting transitions to lattice gauge theory.<sup>1</sup> Below an ordering-phase transition, different phases can coexist and the nature of the coexistence is governed by the thermodynamic properties of the interface. For many systems such as crystalline solids and theoretical lattice models, orientation of the interface, i.e., anisotropy of the interfacial excess free energy, is relevant. For bulk dimensions larger than two, the interface separating ordered domains undergoes a finite temperature roughening phase transition, from a smooth to a rough interface<sup>2</sup> whose width diverges. The excess free energy of forming a single step on an otherwise flat interface also vanishes at the transition. This effect is responsible for the vanishing of facets in crystal-facet transition.<sup>2</sup> Properties of the rough interface also enter in the discussion of wetting, where one of two phases preferentially wets a substrate or a third phase.<sup>3</sup> Although these and similar physical phenomena have been studied extensively by both experiments and theories,<sup>3</sup> a detailed calculation of the anisotropic interfacial tension in a realistic model in three dimensions is absent. We report here a very large-scale Monte Carlo study of a simple-cubic Ising model with a tilted interface for a wide range of lattice sizes, temperatures, and tilt angles. The anisotropic interfacial tensions are obtained and finite-size dependence of the step free energy is used to probe the temperature dependence of the correlation length near the roughening transition. Our analysis yields the roughening temperature in agreement with previous estimates based on other methods, and demonstrates the predicted square-root temperature dependence of the correlation length for the first time. We also point out implications

of the step-free-energy finite-size scaling which may significantly affect the application of capillary-wave Hamiltonian approximations to wetting phenomena in lattice models.

The excess interfacial free energy is the free-energy difference between two systems with and without an interface, and when normalized by the cross-sectional area, it yields the related interfacial tension  $\tau$ . For systems lacking full rotational symmetry such as lattice models, an anisotropic interfacial tension  $\tau(\theta, T)$  is introduced to describe interfaces at an angle of  $\theta$  with respect to a given symmetry direction. We consider here the Ising model with an interface at temperatures  $T$  below the critical  $T_c$ . At  $T=0$ , the interface is flat and with increasing  $T$  excitations of step formation appear with probability determined by the related excess step free energy per unit length  $f_s(T)$ . At  $T=T_R$ , a roughening transition occurs at which  $f_s(T)$  vanishes. The proliferation of steps then leads to the divergence of the interface width. This is also the mechanism for the disappearance of crystal facets at the facet transition.<sup>2</sup>  $\tau(\theta, T)$  enters via the Wulff construction<sup>2</sup> in determining the crystal shape. Note that  $f_s$  is simply the limiting interfacial tension due to an infinitesimally small  $\theta$ , i.e.,  $f_s \sim \partial\tau(\theta, T)/\partial\theta|_{\theta=0}$  which thus also vanishes for  $T \geq T_R$ .

Except for  $T=0$ ,  $f_s(T)$  and  $\tau(\theta, T)$  are not known for the three-dimensional Ising model. In contrast, more is known about the roughening transitions through analytical and numerical studies of various solid-on-solid (SOS) models of interfaces (which neglect overhangs and bubbles).<sup>2</sup> Exact solution<sup>4</sup> and duality transformation<sup>5</sup> predict that these transitions are of the Kosterlitz-Thouless type<sup>6</sup> with an infinite-order transition and infinite correlation length<sup>2</sup> in the rough phase. Numerical evidence

from Monte Carlo simulations<sup>7</sup> of the interface width and other numerical results<sup>8-10</sup> for the Ising model are consistent with the predictions based on the SOS model and universality. In the SOS models,<sup>2</sup>  $f_s(T)$  is predicted to be related to the correlation length  $\xi(t)$  for the height-height correlation function

$$\xi(t) \sim B \exp(At^{-1/2}), \quad t = (1 - T/T_R), \quad (1)$$

and

$$f_s(t) \sim c\xi(t)^{-1} \text{ for } t > 0, \quad (2)$$

while  $\xi(t) = \infty$  and  $f_s(t) = 0$ , for  $t < 0$ . Invoking these relations for the Ising model, we probe the temperature dependence of  $\xi(t)$  by calculating  $f_s(t)$  only. We then avoid the need to define the location of the Ising interface and subsequent measurement of the height-height correlation function.

For temperature ( $T^* = T/T_c$ ) below  $T_c$  ( $J/k_B T_c = 0.221655^{11}$ ), we considered  $L \times L \times L$  simple-cubic lattices under two sets of boundary conditions to simulate systems with and without a tilted interface. By imposing antiperiodic boundary conditions (APBCs) in the  $z$  direction, periodic boundary conditions (PBCs) in the  $y$  direction, and a screw boundary condition in the  $x$  direction with a shift of  $N_\theta$  lattice constants, a tilted interface with the angle of  $\theta = \tan^{-1}(N_\theta/L)$  with respect to the  $xy$  plane is obtained. By comparing the energy of this system to that of an identical system, but replacing the APBC by a PBC, we obtain the excess interfacial energy:

$$\Delta U_{\text{ex}}(\theta, T^*, L) \equiv U_{\text{APBC}}(\theta, T^*, L) - U_{\text{PBC}}(\theta, T^*, L).$$

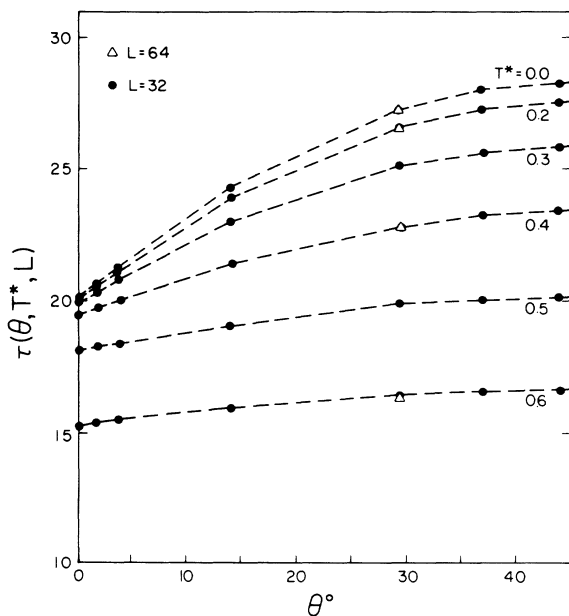


FIG. 1. Angle dependence of the (anisotropic) interfacial tension in units of  $J/k_B T_c$ . The reduced temperature  $T^* = T/T_c$ .

Standard thermodynamic integration (with  $\Delta T^* = 0.01$  or  $0.02$ )<sup>12</sup> starting from  $T^* = 0$  yields  $\tau(\theta, T^*, L)$ . An ultrafast multispin vectorized code<sup>13</sup> implemented on the Cyber 205 is used with speeds up to  $\approx 30 \times 10^6$  updates/s allowing us to study  $L$  as large as 96 using  $\approx 10^5$  Monte Carlo steps per datum point. The finite-size scaling<sup>14</sup> prediction for the step free energy on a finite lattice  $f_s(L, t)$  is

$$f_s(L, t) = c\xi^{-1}(t)X(L/\xi(t)), \quad (3)$$

where  $c$  is some positive constant.  $X(L/\xi(t))$  is the scaling function with the appropriate limits of  $X(\infty) = 1$  and  $X(L/\xi \rightarrow 0) \sim \xi/L$ . In a finite system  $f_s(L, t)$  is nonzero and vanishes only as  $L^{-1}$  at  $T \geq T_R$ . As noted above, this implies that  $\tau' = \partial\tau/\partial\theta|_{\theta=0}$  does not vanish and also scales as  $L^{-1}$ . For larger  $\theta$ , one expects little finite-size dependence and rapid convergence towards the thermodynamic limit.

In Fig. 1, we present results for  $\tau(\theta, T^*, L = 32)$  for a wide range of  $\theta$  and  $T^*$ . The surface tension shows clear anisotropy. For very low temperature, there is pronounced curvature indicating that the second or higher derivatives of  $\tau$  with respect to  $\theta$  are substantial. A rapid decrease of the slope near  $\theta = 0$  with increasing  $T^*$  is evident and the curvature also decreases in the vicinity of  $T_R^* \approx 0.54$ .<sup>7</sup> For larger angles, the convergence with increasing size is very rapid as indicated by  $\theta = 29.3^\circ$ , where the estimates for  $L = 64$  are already indistinguishable from  $L = 32$  within the estimated errors of about 2%.

$f_s(L, t)$  is shown in Fig. 2 as a function of  $T^*$ . The estimated error is 0.01 for  $L = 16$  and increases to 0.06 for  $L = 96$  near the transition. To apply finite-size scaling [Eq. (3)] we need the temperature dependence of  $\xi(t)$ . To this end, we extrapolate  $f_s(L, t)$  to  $L \rightarrow \infty$  by assuming an  $L^{-1}$  dependence [Fig. 3(a)]. Using the extrapolated value of  $f_s(\infty, t)$  together with Eq. (2), we test the predicted behavior of  $\xi(t)$  by plotting [Fig. 3(b)]  $\ln f_s(\infty, t)$  vs  $\sqrt{t}$  with different trial values for  $T_R^*$ . A good linear fit is obtained for  $T_R^* = 0.54 \pm 0.02$ , con-

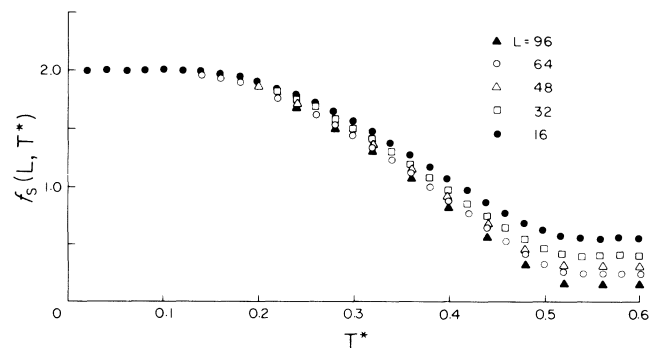


FIG. 2. Temperature dependence of the step free energy  $f_s(L, T)$  in units of  $J/k_B T_c$ .

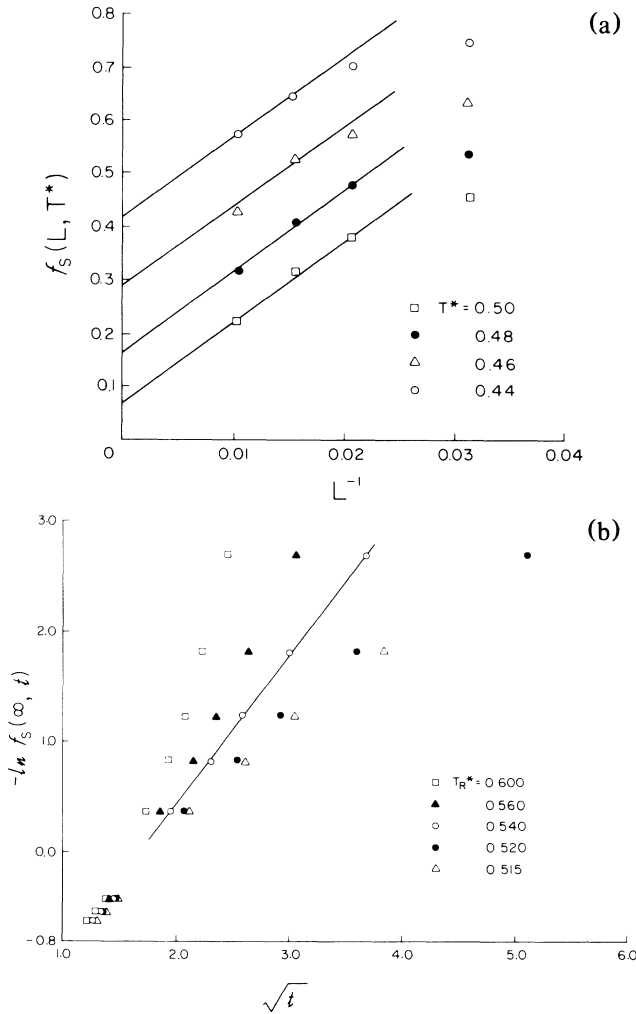


FIG. 3. (a) Size dependence of the step free energy  $f_s(L, T)$  in units of  $J/k_B T_c$ . (b) Log of step free energy per unit length of steps vs  $\sqrt{t}$  for  $t = 1 - T^*/T_R^*$  with different trial values of  $T_R^*$ . The solid line is the best linear fit obtained.

sistent with previous estimates<sup>7</sup> of  $0.56 \pm 0.03$  thus confirming the square-root dependence in Eq. (1). The fit gives  $A = 1.36 \pm 0.06$  and  $B = 9.84 \pm 2.0$  for  $\xi(t)$  [Eq. (1)]. Using these results for  $\xi(t)$ , we check finite-size scaling [Eq. (3)] by plotting  $[f_s(L, t)\xi(t)] - 1$  vs  $L/\xi(t)$  (Fig. 4). The data indeed scale quite well.

For a discussion of capillary waves, the higher derivatives of  $\tau(\theta, T^*, L)$  with respect to the angle  $\theta$  are important.<sup>15</sup> We consider

$$\tau(\theta, T^*, L) = \tau(0, T^*, L) + \tau'(0, T^*, L) |\theta| + \frac{1}{2} \tau''(0, T^*, L) \theta^2. \quad (4)$$

We ignore higher-order terms and find that  $\tau''$  is negative for low  $T^*$  but, for large  $L$ , crosses to positive values near  $T_R^*$ . This behavior is consistent with the mechanical stability requirement by the interface. Further dis-

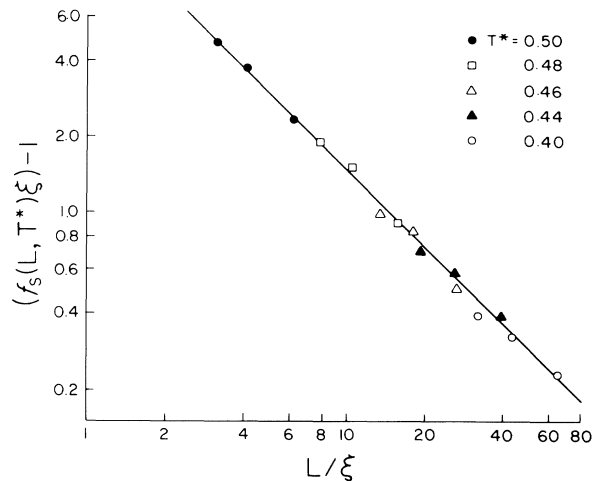


FIG. 4. Finite-size scaling plots of step free energy. See text.

cussion and results will be presented elsewhere.

In conclusion, our detailed and accurate results for the anisotropic surface tension  $\tau(\theta, T^*, L)$  and step free energy  $f_s(L, t)$  for the Ising model are consistent with predictions based on the theory of roughening transitions. Our results have important implications for numerous applications, e.g., critical wetting for which recent simulations<sup>16</sup> are inconsistent with a renormalization-group calculation based on a capillary-wave Hamiltonian.<sup>17</sup> This model is essentially a continuous Gaussian SOS model in which the coupling constant  $\sigma$  is related to the coarse-grained anisotropic interfacial tension of the Ising interface.<sup>15</sup> For example, in two-dimensional systems above  $T_R$  ( $T_R = 0$  in  $d=2$ ),  $\tau' = 0$  and  $\sigma = \tau_{cg}(0, T^*) + \tau_{cg}''(0, T^*)$ , with the assumption that Eq. (4) holds for  $\tau_{cg}(\theta, T^*)$ . One then defines  $\tau_{cg}(\theta, T^*)$  by integrating over short wavelength fluctuations up to a length scale, which is at least of the order of the bulk correlation length  $\xi_b$ . For  $d=3$  the expression for  $\sigma$  is more complicated (two angles are needed to describe arbitrary interface orientation), but terms in  $\tau'$  are still absent. Since we expect that the  $L$  dependences of  $\tau_{cg}(\theta, T^*)$  and  $\tau(\theta, T^*)$  are qualitatively similar, our results indicate  $\tau_{cg}'$  scales as  $L^{-1}$  and  $\tau_{cg}''$  is positive definite (but finite) in the rough phase. For critical wetting, the long wavelength excitations are important and  $\theta$  is of order of  $L^{-1}$ . This implies that for any finite  $L$ , the linear term cannot be neglected and may be comparable to the quadratic term. The use of the capillary-wave Hamiltonian is thus a questionable approximation, except when very near  $T_c$  where  $\xi_b \rightarrow \infty$  and  $\tau$  is isotropic. We hope that our work will stimulate research to clarify these effects of anisotropy on the application of capillary-wave Hamiltonians.

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<sup>(a)</sup>Present and permanent address: Kontron Elektronik, D-8057 Eching, West Germany.

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