Structural Transition in the Ising-Model Interface

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We have studied the interface between regions of opposite spin in the simple cubic Ising model. Low-temperature expansions of moments of the gradient of the density profile and of the slope at its midpoint suggest that the interface width diverges at a temperature T_R about half the critical temperature. We discuss the physical significance of this transition and implications for general theories of interfacial properties.

Recently, there has been much progress in the understanding of the two-phase region in the Ising model. Dobrushin[1] has shown that at sufficiently low temperatures in the simple cubic Ising model with appropriate symmetry-breaking boundary conditions, there exists an interface of finite width between translationally nonequivalent regions of mostly "up" and mostly "down" spins. In contrast, Gallavotti[2] had proven for the square Ising model that there exist large fluctuations which cause the interface width to diverge at any finite temperature. In this Letter we present evidence suggesting that at a temperature T_R about half the bulk critical temperature T_c, interfacial fluctuations in the simple cubic Ising model become large enough to cause a divergence of the interface width and to produce singularities in the interface thermodynamic functions.

A transition at a new temperature $T_R < T_c$ may seem surprising since most known cases either of interface or free-surface thermodynamic functions are nonanalytic only at T_c. Examples include Onsager's[3] exact calculation of the square-lattice interface free energy (interface tension), σ, Fisher's and Ferdinand's[4] results for a number of related two-dimensional (2D) lattices, and the results for a free surface in two or three dimensions, most recently discussed by Binder and Hohenberg.[5]

However, the following simple argument, given in essence by Burton, Cabrera, and Frank in 1951,[6] suggests why the interface in the 3D Ising model should behave differently. At $T = 0$ the (100) interface between regions of up and down spins for a simple cubic lattice is perfectly flat. The spins in the lattice planes immediately above or below the interface feel no vertical mean field from the rest of the lattice, since the interactions from their neighboring lattice planes cancel. This suggests that a boundary lattice plane might behave like a 2D Ising model, with large spin fluctuations and thermodynamic singularities at the 2D critical temperature T_c^{2D} which is only $0.503T_c$. This argument helps rationalize the other cases: The interface in a 2D Ising model is 1D and hence has no critical point. Similarly, the surface layer of a free surface is in a nonzero stabilizing mean field from the plane below, and no additional singularities would be expected.

Similar conclusions follow if the 3D interface is treated by other approximate means such as the Bragg-Williams[7] or the Bethe approximations[8]; Kikuchi[9] has most recently predicted interface thermodynamic singularities using his cluster variation methods. However, the structural implications of these treatments are not very clear and mean-field and higher-order approximations are sometimes misleading when applied to critical-like phenomena.

We consider here the low-temperature expansion method, much used for critical phenomena, to examine quantities which yield information about the possible structural changes in the interface. Let the successive (100) lattice planes in a simple cubic Ising model of dimensions L^3 with isotropic coupling constant J be at $z = \pm\frac{1}{2}, \pm\frac{3}{2}, \pm\frac{5}{2}, \ldots$. An external magnetic field $H(z)$ uniform in the region $z < 0$ tends to point those spins up, and a similar reversed field points spins down for $z > 0$. The magnetic fields act rather like a stabilizing gravitational field in a real fluid system. We assume periodic boundary conditions in the x-y directions and antiperiodic boundary conditions between the positive and negative z boundaries. We require the limits $H \to 0^+$, giving us an infinitesimal symmetry-breaking field, and $L \to \infty$, the infinite-volume limit. Using lattice-gas language where an up spin is an occupied site and a down spin a vacancy, we define the layer densities $\rho(z)$ normalized so that at $T = 0$, $\rho(z) = 1$ for $z < 0$ and $\rho(z) = 0$ for $z > 0$.

Information about the density profile can be obtained from moments of the density gradient $d\rho(z)/dz$.[10] In a lattice system these are defined

as

$$\langle z^{2n}\rangle = \sum_{z=-\infty}^{\infty} [\rho(z-\tfrac{1}{2}) - \rho(z+\tfrac{1}{2})]z^{2n}.$$

$\langle z^2\rangle$ may be determined for a fluid by measurements of the interface reflectivity, and $l \equiv \langle z^2\rangle^{1/2}$ gives a measure of the interface width. At $T=0$, $l=0$, and Dobrushin's work[1] implies that l remains finite for low, nonzero, temperatures. However, at T_R we would expect the large fluctuations to cause l (and all the even moments) to diverge to infinity.

The layer densities $\rho(z)$ themselves give further structural information. At $T=0$, $\rho(\frac{1}{2})=0$, and $\rho(-\frac{1}{2})=1$ and these must change only slightly at very low temperatures. At T_R, however, one would expect the interfacial fluctuations to cause $\rho(\frac{1}{2})=\rho(-\frac{1}{2})=\frac{1}{2}$. Thus the quantity $M=1/[1-2\rho\frac{1}{2}]$ should diverge as $T \to T_R$. Note that M^{-1} gives the slope $d\rho(z)/dz$ of the density profile at $z=0$.

We have derived low-temperature expansions[11] for these and related quantities. No qualitatively new computational problems arise when an interface is present, but the quantitative details are more difficult because of the symmetry reduction and the much larger number of low-energy configurations of overturned spins which can be excited near the $z=0$ nominal interface. For example, at $O(Y^9)$ in the expansion variable $Y=e^{-4J/kT}$, there are nearly 3000 different connected configurations touching the $z=0$ plane including large horizontal clusters of up to twenty overturned spins and vertical towers extending to the $z=\pm 4$ planes. We have generated by computer all the contributions through $O(Y^9)$ from single connected interface clusters or from groups of two or three separated clusters, leaving only a relatively small number to do by hand.[12]

Table I gives the low-temperature expansion coefficients for $\langle z^2\rangle$, $\langle z^4\rangle$, and $M-1$. The results of a Padé approximant analysis[13] of the logarithmic derivative of the series is given in Table II. This analysis is based on the assumption that near T_R the dominant singularity is of the form $(Y-Y_R)^{\theta}$, where Y_R is the value of Y at T_R, and θ is the exponent. The predicted transition temperature T_R, in units of the critical temperature T_c, and the exponent θ are given for various $[N,D]$ Padé approximants.

The results of Teble II show reasonably good agreement between the various Padé orders in predicting a transition temperature T_R at about $0.57T_c$. However, the results of any series analysis must be viewed with caution, and the series we have used are not very long, so the quantitative accuracy of these results is difficult to estimate. The important point is that such physically different quantities as $\langle z^2\rangle$ and M both are predicted to diverge at essentially the same nonzero temperature T_R, in agreement with the physical picture we have discussed above. As is typically the case, the apparent convergence of the exponents[14] is less satisfactory. The relation between the exponents in Table II, however, does suggest that the density profile cannot be scaled in terms of a single temperature-dependent characteristic length $l(T)$.

Further evidence that T_R is not related to the bulk T_c comes from the anisotropic limit of the Ising model in which the vertical coupling J_z between lattice planes approaches infinity, giving the solid-on-solid (SOS)[15] model familiar to crystal-growth theorists. In this limit no new vertical "broken bonds" can occur, so bulk excitations and overhanging configurations are suppressed and T_c tends to infinity. Low-temperature expansions for the SOS model give essentially identical singularities to those described here for the isotropic Ising model. This is to be expected since at T_R the bulk densities in the isotropic Ising model differ by less than 2% from the $T=0$ values. These SOS results, together with analy-

TABLE I. Low-temperature expansion coefficients A_n for $Q=\sum A_n Y^n$.

Q \ n	2	3	4	5	6	7	8	9
$\langle z^2\rangle$	2	6	40	144	826	3270	17 364	74 832
$\langle z^4\rangle$	2	6	64	192	1570	5190	35 460	135 648
$M-1$	2	8	38	176	854	4116	20 210	99 088

TABLE II. Estimates of the transition temperature T_R and exponent θ derived from the $[N,D]$ Padé approximants to the logarithmitic derivative of the series in Table I. Here N and D refer to the order of the polynomials in the numerator and denominator, respectively.

	For $\langle z^2\rangle$		For $\langle z^4\rangle$		For M	
	T_R/T_c	θ	T_R/T_c	θ	T_R/T_c	θ
[3,3]	0.576	0.999	0.581	1.35	0.546	0.782
[2,4]	0.577	1.01	0.586	1.43	0.546	0.782
[4,2]	0.560	0.785	0.534	0.644	0.546	0.792
[2,3]	0.579	1.02	0.589	1.50	0.546	0.784
[3,2]	0.579	1.02	0.602	1.73	0.547	0.802
[2,2]	0.575	0.997	0.570	1.92	0.549	0.818

sis of the interface thermodynamic functions for both models and Monte Carlo simulations of the SOS interface, all of which indicate singular behavior near T_R, will be presented in detail in another paper. Some indirect evidence for the existence of a transition at T_R is given by modern theories of crystal growth[16] which use the concept of surface roughening at the "roughening temperature" T_R to describe changes in equilibrium crystal morphology.

The divergences in interface width l and other singularities we have described should occur only in the limit $H \to 0^+$. They are analogous to the divergences in interface width which occur in the drumhead model[17] of the liquid-gas interface as a result of long-wavelength surface ripples when the gravitation constant $g \to 0^+$. However, the lattice-gas interface "roughens" at a finite T_R and not at $T = 0$ as for the (continuum) drumhead model. A complete theory of interface properties must include the effects of a stabilizing field, and theories which introduce only bulk thermodynamic concepts and which use the concept of an intrinsic (field-independent) interface width[18] cannot explain the present results. However, these approaches may still be useful for describing the behavior of the interface width in a finite field near T_c, since the dependence of the density profile on the external field H is probably very small when H is finite, rather than infinitesimal. The intrinsic-width theories may accurately approximate the actual interface width over a wide range of finite external fields.[19] More work is needed to resolve unambiguously the contributions to the interface width from a postulated intrinsic width and from long-wavelength surface ripples, and to assess their respective dependencies on an external field.

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