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Monte Carlo evaluations of interfacial tension and universal amplitude ratios of the three-dimensional Ising model

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The interfacial tension of the simple-cubic Ising model near the transition temperature has been calculated with use of an extension of a recently proposed novel Monte Carlo method. Finite-size scaling theory was used to analyze the results and to obtain the surface tension amplitude. Universal ratios involving the surface tension amplitude are evaluated. Previous ambiguities in the theoretial value of the ratio involving the surface tension and specific-heat amplitudes have been clarified.

The properties of surfaces and interfaces near the bulk critical point are of considerable current theoretical and experimental interest.[1] Until recently, there have been no efficient direct methods for calculating the interfacial tension near the critical point (T_c) that are applicable to large systems. Indirect methods using standard thermodynamic integration[2] techniques have been very useful below T_c, and a rather elegant method[3] based on studying the order parameter distribution was shown to be quite successful, but limited to systems of moderate size. In a recent paper,[4] we have proposed the application of a multistage sampling technique to the direct calculation of interfacial tension. Our conclusions, based on the use of extensive results using large system sizes and a wide range of temperatures, were in excellent agreement with exact results for the two-dimensional ferromagnetic Ising model. Here we extend this method to the three-dimensional ferromagnetic Ising model on a simple-cubic lattice using system size as large as $16\times16\times16$. (One may treat larger system sizes with this method, but for our present purposes it does not appear to be necessary.) The results have been analyzed using finite-size scaling theory to obtain the surface tension amplitude. An accurate computation of this amplitude allows a determination of estimates for a number of universal ratios involving the amplitude of surface tension and various other thermodynamic quantities which are experimentally accessible.

Specifically, in this Brief Report, the new estimate of the surface tension amplitude is used to update estimates for four universal ratios. For several reasons, the ratio U_1, involving the surface tension and correlation length, is of primary interest, but we shall also comment on three additional ratios which have been discussed. The first, $Y^{(\pm)}$, involves amplitudes for the surface tension and specific heat. The second, $\beta^2 c$, involves the surface tension, susceptibility, and correlation length, and, finally, X_0 involves surface tension, susceptibility, and the miscibility gap. These quantities will be defined below. Estimates of U_1 take on special significance because of a recent reanalysis of available experimental data by Moldover.[5] The agreement or, rather, lack of agreement, shall be discussed in the following. In general, comparison of these ratios is important in assessing the levels of present theoretical understanding. Accurate values for some of these ratios are also important in discussions of various phenomena such as critical wetting.[6]

We consider a ferromagnetic Ising model having nearest-neighbor exchange J on a simple-cubic lattice of sites labeled by integers (i,j,k) with $N_x\times N_y\times N_z$ ($N_x=N_y=N$, $N_z=N+2$) spins, $\sigma_{ijk}=\pm1$. The Ising Hamiltonian is, as usual, $H=-\sum_{\langle \mathrm{NN}\rangle} J\sigma_{ijk}\sigma_{lmn}$, the sum being over nearest-neighbor sites. The system is at temperature T below the critical temperature T_c in zero magnetic field. The interfacial tension is taken as the difference of free energy per unit area:

$$\tau=\lim_{N\to\infty}\left(\frac{-k_BT}{N^2}\ln\frac{Z_{+-}}{Z_{++}}\right) . \tag{1}$$

Here Z_{+-} and Z_{++} are the partition functions under two sets of boundary conditions $(+-)$ and $(++)$, respectively. The $(+-)$ boundary condition is defined by fixing the first and last layers of spins $[(i,j,1)$ and $(l,m,N_z)]$ with values $\sigma=+1$ and -1, respectively. Periodic boundary conditions are taken for the x and y directions. The $(++)$ boundary condition is similar except that the first and last layers of spins both have $\sigma=+1$. Equation (1) may be rewritten as

$$\begin{aligned}\tau&=-\frac{k_BT}{N^2}\ln\left\langle\exp\left[-\frac{1}{k_BT}(H_{+-}-H_{++})\right]\right\rangle_{++}\\&=-\frac{k_BT}{N^2}\ln\left\langle\exp\left[-\frac{2J}{k_BT}M_S\right]\right\rangle_{++} ,\end{aligned}\tag{2}$$

where $M_s=\sum_{ij=1}^{N}\sigma_{i,j,N+1}$ is the magnetization of the layer adjacent to the fixed (boundary) "all minus" layer, and the ensemble average $\langle\ \rangle_{++}$ is generated by the Hamiltonian H_{++}, in which the boundary spins satisfy the $(++)$ boundary condition.

Except for very small systems, the ensemble averages cannot in practice be evaluated directly by standard Monte Carlo sampling. We have shown that accurate results can be obtained even for rather large systems by application of *multistage sampling techniques.*[4] To do this, Eq. (1) is rewritten by considering a three-stage sampling,

$$\frac{Z_{+-}}{Z_{++}}=\frac{Z_{+-}}{Z_{H'}}\frac{Z_{H'}}{Z_{H''}}\frac{Z_{H''}}{Z_{++}} , \tag{3}$$

where each ratio can be evaluated as an ensemble average as in Eqs. (1) and (2). For example,

$$\frac{Z_{H'}}{Z_{H''}}=\left\langle \exp\left[-\frac{1}{k_B T}(H'-H'')\right]\right\rangle_{H''} . \tag{4}$$

H' and H'' are some suitable Hamiltonians chosen such that the ratios can be evaluated efficiently. In the application to the two-dimensional Ising model,[4] only two stages were found to be necessary, and H' was defined by a "hybrid boundary condition." (The spins on one edge are all "+" and on the other edge "+" and "−" in an alternating sequence.) We have found the two-stage formulation to be adequate for small systems in three dimensions, but it rapidly becomes inefficient for large systems.

We have used the three-stage formulation [Eq. (3)] and have chosen H' and H'' as follows: The spins on one z face ($\sigma_{i,j,1}$) are taken to be $\sigma_{i,j,1}=+1$, and are coupled to the spins in the adjacent layer ($\sigma_{i,j,2}$) by J as before, but on the other z face, the spins ($\sigma_{i,j,N+2}$) are fixed at $\sigma_{i,j,N+2}=-1$ and are coupled to the spins in the adjacent layer ($\sigma_{i,j,N+1}$) by J' and J'', respectively. J' and J'' were chosen for efficient sampling. In the actual calculation, a range of J' and J'' has been used and the results are independent of the choices of J' and J''. Values of $J'\simeq -0.4J$ and $J''\simeq +0.3J$ were found to be quite efficient.

We have been able to calculate τ (near $k_BT_c/J=4.5114$) (Ref. 7) for $N\leq 16$ with about 10^5–10^6 Monte Carlo steps per spin and obtain, for the first time, accurate results for such large systems using direct Monte Carlo samplings. Much larger systems and/or lower temperatures can be studied by this method by using a larger number of intermediate stages.

The results for different sizes and temperatures are analyzed using finite-size scaling theory.[8] The scaling ansatz for the interfacial tension is

$$\tau_N(t)=\tau_0 t^{\mu}\Sigma(x) , \tag{5}$$

with $x=c_L N^{1/\nu}t$ and $t=(T_c-T)/T_c$. Here μ and ν are the appropriate critical indices for the interfacial tension and correlation length, respectively, and τ_0 and c_L are nonuniversal amplitudes. The universal scaling function $\Sigma(x)$ has the asymptotic limits, $\Sigma(x=\infty)=1$ and $\Sigma(x\to 0)\sim x^{-\mu}$. We assume the hyperscaling form $\mu=(d-1)\nu=2\nu$ along with $\nu=0.63$.[9] To obtain τ_0, we have plotted $N^{\mu/\nu}\tau_N(t)$ vs $N^{\mu/\nu}t^{\mu}$, with τ_0 as the limiting slope for large x. (See Fig. 1.) (For a discussion of this method of analysis, see Ref. 4.) This approach is more direct, and simpler than the previous method used in obtaining estimates for the three-dimensional Ising model.[3]

For data with $t\leq 0.02$, the large x limit is reached for $N^{\mu/\nu}t^{\mu}>0.2$, and the estimated slope is 5.4 ± 0.4. (For data with $t>0.02$, deviations are observed.) We obtain $F_s\equiv\tau_0/k_BT_c=1.2\pm0.1$, in contrast to previous estimates of Binder[3] (1.05 ± 0.05), using a different method with smaller system size ($N\leq 12$). Using the value for F_s, we can obtain estimates for universal ratios involving the surface tension. The most direct ratio is

$$U_1=F_s[f_1^{(+)}]^{d-1} , \tag{6}$$

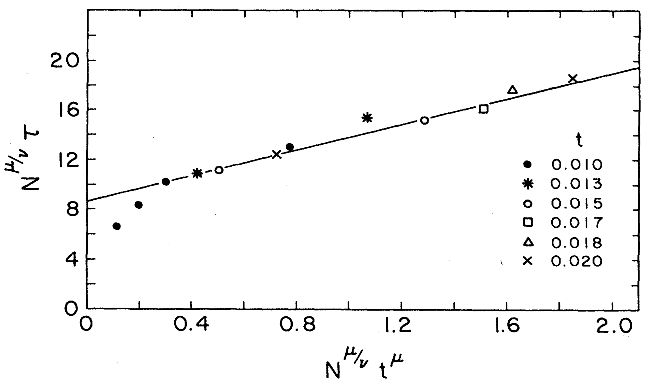

FIG. 1. Finite-size scaling analysis for $T<T_c$ with $N\leq 16$. $t=(T_c-T)/T_c$.

where the correlation length above (+) and below (−) T_c is defined as $\xi/a=f_1^{(\pm)}|t|^{-\nu}$, where a is the lattice spacing and ν is the standard exponent. The amplitudes $f_1^{\pm}$ have been estimated by Tarko and Fisher[10] as $f_1^{(+)}=0.478\,26\pm0.0004$ and $f_1^{(-)}=0.244\pm0.001$. Our estimate for U_1 is then $U_1=0.27\pm0.023$. (Here and in what follows our errors reflect only that in τ_0.) This result will be discussed further in the conclusions and is included in the summary (see Table I).

A related ratio involves the specific heat, whose amplitude $A^{(\pm)}$ above and below T_c is defined for convenience by

$$C^{(\pm)}(t)=\frac{k_BA^{(\pm)}}{\alpha}|t|^{-\alpha},\quad (T_c\to T\pm) . \tag{7}$$

The ratio which is presumed to be universal is given by[11]

$$Y^{(\pm)}\equiv\lim_{t\to 0}\left[\frac{\tau(t)/k_BT_c}{[\alpha t^2C^{(\pm)}(t)/k_B]^{(d-1)/d}}\right]=\frac{F_s}{[A^{(\pm)}]^{(d-1)/d}} . \tag{8}$$

In the first analysis of experimental data by Stauffer, Ferer, and Wortis,[11] estimates were obtained using specific-heat data for $T>T_c$.[12] Of course, $Y^{(+)}$ is related to $Y^{(-)}$ by use of the ratio $A^{(+)}/A^{(-)}$, which is also presumed to be universal. The values of Y given by Stauffer *et al.*[11] are hence for $Y^{(+)}$ and *not* $Y^{(-)}$. This rather subtle but important point was apparently not noticed in the first comparison[3] between theory and experiment and is responsible *in part*[13] for the rather large discrepancy between the two (12.3 vs ~6). The comparison involving $A^{(+)}$ is also preferred from the theoretical point of view. Estimates[14] for $A^{(+)}$ from series expansions are considered to be more reliable than $A^{(-)}$, and there is some uncertainty in the ratio $A^{(+)}/A^{(-)}$. We have used the present estimate for F_s and $A^{(+)}=0.142$ from series expansions[14] to obtain $Y^{(+)}=4.4\pm0.4$. (See Table I.) The discrepancy between theory and experiment is much reduced, and the two can be considered to be in reasonable agreement considering the rather large uncertainty in the experimental estimates.

Another universal ratio, first suggested by Fisk and Widom,[15] involves the correlation length and, in addition, the order parameter and susceptibility of the Ising model below

TABLE I. Comparison of theory and experiments for the universal ratios.

	Theory		Experiments	
	Present work	Ref. 3		
U_1	0.27 ± 0.023			0.39^{a}
$Y^{(+)}$	4.4 ± 0.4	. . .	Xe:	6.2 ± 0.06^{b}
			CO_2:	6.4 ± 0.4^{b}
			$C_6H_{12}-CH_4O$:	41.0 ± 1.0^{b}
$\beta^2 c$	0.104 ± 0.01	0.092 ± 0.005	SF_6:	0.093 ± 0.011^{c}
			Xe:	0.1 ± 0.12^{c}
			CO_2:	0.11^{c}
				0.123^{c}
				0.146^{c}
X_0	1.45 ± 0.2	1.2 ± 0.1	CO_2:	1.24 ± 0.1^{d}
			$C_7H_{14}-C_7F_{14}$:	1.3 ± 0.1^{d}

[a]From Ref. 5.
[b]Experimental values given in Ref. 11.
[c]Experimental values quoted in Ref. 20.
[d]Experimental values quoted in Ref. 19.

T_c: $\langle \sigma \rangle = Bt^{\beta}$, $k_B T \chi = C^{(-)} t^{-\gamma}$. The ratio is given by[3]

$$\beta^2 c = \frac{C^{(-)} F_s}{4B^2 f_1^{(-)}} \quad . \tag{9}$$

The various amplitudes used are $B = 1.569$,[16] $C^{(-)} = 0.209$, and $f_1^{(-)} = 0.244$. The estimates for $C^{(-)}$ and $f_1^{(-)}$ are given by Tarko and Fisher.[10] We note at this point that, when comparing with experiments or calculations, there may be some ambiguity in identifying the amplitude for the correlation length.

The problem is the following. The Fisk-Widom approach is based on a functional method which has correlations at the level of modified Ornstein-Zernike (OZ) theory or "renormalized mean-field theory." At such a level, the critical exponent η plays a role and, for example, $\chi \sim \xi^{2-\eta}$ (not simply ξ^2). At that level, however, the "true" correlation length, based on the rate of exponential decay, is *equal* to the "second-moment" correlation length, based on the second spatial moment of the two-point function. Furthermore, the natural interface width at the Fisk-Widom level is also equal to these lengths. In general, however, the two correlation lengths have amplitudes differing by order unity. This is known from series expansions,[10,17] and seen at $O(\epsilon)$ in a renormalization-group $4-\epsilon$ expansion. At $O(\epsilon)$, however, the natural interface width turns out to be equal to the true bulk correlation length.[18] It is not known whether or not this result persists to higher order in ϵ. The problem, then, in comparing Eq. (9) with experiment is to get all involved to make a consistent choice of the correlation length and its amplitude. This may not be too serious in practice, since experiments are usually fit to an OZ form. However, the matter can be compounded experimentally since estimates of ξ may come from *dynamical* spectral width measurements, which may again have amplitudes differing slightly. The value $f_1^{(-)}$ of Tarko and Fisher[10] is a second-moment length.

The estimate for $\beta^2 c$ is 0.104 ± 0.01 in agreement (taking note of the cautions above) with estimates of Binder[3] and experiments. (See Table I.) The final universal ratio (universal only for $d=3$) was discussed by Langer and Schwartz in the context of nucleation phenomena in the critical region.[19] This ratio is defined by,[3,19] $X_0^2 = (16\pi/3\beta^2) F_s^3 [C^{(-)}]^2 / B^4$.

Using the present estimate for F_s with B and $C^{(-)}$, we obtain $X_0 = 1.45 \pm 0.2$. (See Table I.) This is also in agreement with estimates of Binder[3] and experiments (and not susceptible to the ambiguities noted above).

We have shown in this paper that the interfacial tension for the three-dimensional Ising model can be evaluated accurately and with system size larger than handled previously, by an extension of a novel multistage Monte Carlo sampling technique. The universal ratio U_1, defined in Eq. (6), is the most direct. From a theoretical point of view, estimates of correlation length amplitudes are usually more accurate via series analysis than specific heats. Our error estimate in U_1 reflects entirely the uncertainty in τ_0 ($\sim 10\%$), which is far larger than errors in $f^{(+)}$ suggested by Tarko and Fisher.[10] It has not been our intent to reanalyze series including, perhaps more systematically, corrections to scaling. Such systematic corrections are not likely to increase the uncertainty in $f_1^{(+)}$ by orders of magnitude. However, the theoretical estimate of U_1 seems to disagree substantially (perhaps 30% or 40%) with the confluence of experimental results $U_1 \simeq 0.39$ quoted by Moldover.[5] At the present time, the origin for the discrepancies is not certain.

Agreement with experimental values of the other universal ratios $Y^{(+)}$, $\beta^2 c$, and X_0 is reasonable, as shown in Table I, although residual differences with $Y^{(+)}$ on the order of 30% may remain. These should perhaps not be taken as seriously as differences in U_1 because of the difficulty of obtaining precise estimates of specific-heat amplitudes. Further experimentation here may yield significant improvement over results obtained over a decade ago.

One may wonder if the Ising model itself shows that U_1 is universal. Preliminary Monte Carlo estimates on the bcc lattice indicate that U_1 is indeed universal. These results will be presented elsewhere.

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