Surface magnetism: A Monte Carlo study of surface critical behavior

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We study the critical behavior of an Ising model on a simple cubic lattice with two free surfaces using the Monte Carlo method. The effect of both the ratio of surface to bulk coupling J_s/J_b, and the length to width ratio, $r=L/D$, are considered. The spontaneous magnetization and the magnetic susceptibility near the critical point are calculated for both the surface and the bulk. For sufficiently large ratio J_s/J_b, we find that the surface magnetizes at a higher temperature than the bulk, as has been reported previously. However, we find that the critical value of J_x/J_b, where the surface and bulk magnetization vanish at the same temperature, depends on the aspect ratio r, and that for a range of values of r this can be less than the value 1.55 valid for a semi-infinite geometry. The results reported here may be applicable to thin magnetic films.

I. INTRODUCTION

The field of surface magnetism has gained new vigor due to the development of new tools and techniques for the study of surfaces. The conventional techniques of surface science involving electron spectroscopy have recently been improved so as to routinely allow the detection of spin ordering. These experiments have been mainly concerned with the microscopic description of phase transitions from the paramagnetic to the ferromagnetic state of $3d$ metals. Studies on NiO,[1] Ni,[2] Fe,[3] and Gd on W (Ref. 4) have been performed. Some of these authors have indicated the existence of a ferromagnetic layer on a paramagnetic bulk. In some cases a critical temperature difference $T_{cs}-T_{cb}$ between the surface (T_{cs}) and the bulk (T_{cb}) as large as 22 K has been reported.[4]

The same phenomenon has been predicted on theoretical grounds by Seltzer and Majlis,[5] Hohenberg and Binder,[6] Nakanishi and Fisher,[7] and others. Some of these authors have pointed out that in order for surface magnetism to occur in the absence of bulk magnetization, the ratio of the coupling between spins on the surface and those in bulk, J_s/J_b, must be larger than the critical value $J_s/J_b=1.55$.[8]

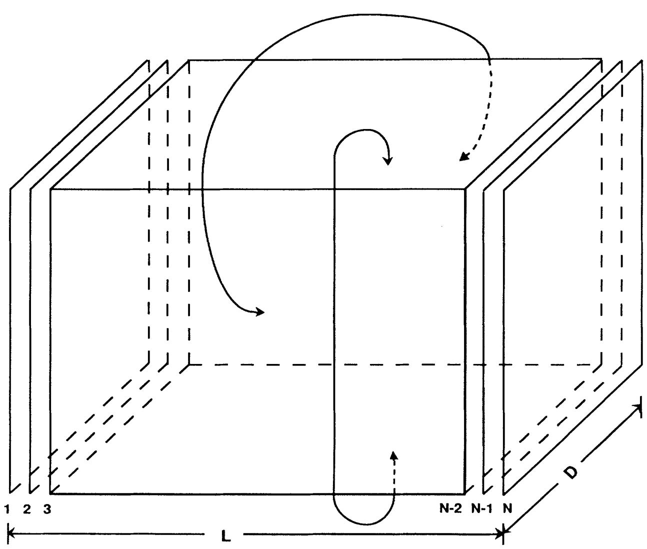

FIG. 1. Cubic lattice with two free surfaces separated by L lattice spacings with periodic boundary conditions in the transverse dimension. The first, second, $(N-1)$th, and Nth layers are considered as part of the surface.

We consider an Ising Model on a cubic lattice of length L, side D, and periodic boundary conditions in the transverse dimensions (see Fig. 1). The effect of different values of J_s/J_b and L/D are considered. Most of the calculations of this sort found in the literature were done with an applied field, and in those cases where no field was applied, only specific values of these ratios were considered. The excellent work of Binder, and Hohenberg and others[5-9] was focused on the critical properties of the model in the limit of a semi-infinite geometry ($L/D\rightarrow\infty$). In this study we examine in detail the effect of the aspect ratio on the transition.

It is well known that free boundary conditions can strongly affect the results of a Monte Carlo simulation, especially near the critical point. Therefore in order to distinguish the effects of surface magnetism from finite size effects it is necessary to study the effect of varying both the total number of spins and the ratio L/D.

The Ising model is directly applicable only to strongly uniaxial magnetic systems, but we expect that the general behavior of the surface and bulk magnetizations in more realistic models (e.g., the Heisenberg model) is similar.

II. THEORY AND DEFINITIONS

Our approach is to consider the Ising spins in the surface layer and first layer below the surface to constitute "the surface," and the exchange coupling, J_s, between spins in these planes may or may not be different from the coupling, J_b, between spins in the bulk. The coupling between spins on the surface and spins in the bulk is taken to be equal to J_b. Only nearest-neighbor interactions are considered so the Hamiltonian for the system is

$$H=-\sum_{\langle ij\rangle}J_{ij}\sigma_i^z\sigma_j^z\ , \tag{1}$$

where

$$J_{ij}=\begin{cases} J_s & \text{for } i,j \text{ on the surface} \\ J_b & \text{otherwise} \end{cases} \tag{2}$$

and $\langle ij \rangle$ indicate nearest neighbors.

The susceptibility χ, the specific heat C, and the magnetization m are defined as

$$\chi_s=N_s(\langle m_s^2\rangle-\langle m_s\rangle^2)/\tau\ ,$$
$$\chi_b=N_b(\langle m_b^2\rangle-\langle m_b\rangle^2)/\tau\ , \tag{3}$$

$$C_s=N_s(\langle E_s^2\rangle-\langle E_s\rangle^2)/\tau^2\ ,$$
$$C_b=N_b(\langle E_b^2\rangle-\langle E_b\rangle^2)/\tau^2\ , \tag{4}$$

and for a single Monte Carlo run

$$m_s=\frac{1}{N_s}\sum_{i-1}^{N_s}\sigma_i,\quad m_b=\frac{1}{N_b}\sum_{i=1}^{N_b}\sigma_i\ , \tag{5}$$

where E is the calculated energy per spin, $\tau=k_BT/J_b$, T being the temperature and k_B the Boltzmann constant, N_s is the number of spins on the surface, and N_b is the number of spins in the bulk. In a long Monte Carlo calculation the values of the magnetization per spin obtained from (5) are used to generate a distribution function, $P(m)$, for the magnetization.

In a finite system the magnetization per spin will assume all possible values between -1 and $+1$ during a sufficiently long Monte Carlo simulation. The probability distribution for m, $P(m)$, will exhibit a single maximum for $T>T_c$ at $m=0$, while for $T<T_c$ $P(m)$ will have two peaks about $m=\pm m_0$.

It is well known that systems exhibit spontaneous symmetry breaking only in the thermodynamic limit. In finite systems the order parameter is identically zero. The same is true of sufficiently long Monte Carlo simulations of finite systems. The question then arises as to how one can determine the order parameter from Monte Carlo data.

Let $P(m)$ be the calculated distribution function for the magnetization per spin obtained from a Monte Carlo simulation. The distribution is normalized

$$\sum P(m_i)=1 \tag{6}$$

and is characterized by the moments $\langle m^n\rangle$,

$$\langle m^n\rangle=\sum P(m_i)m_i^n\ . \tag{7}$$

For a sufficiently long simulation we will find that $P(m)=P(-m)$, so that the first moment $\langle m\rangle=0$. For $T>T_c$ $P(m)$ is symmetric about the single maximum at $m=0$ and the width of the distribution, $(\langle m^2\rangle)^{1/2}$, is related to the susceptibility per site χ by

$$\chi=\beta N\langle m^2\rangle,\quad T>T_c\ , \tag{8}$$

where N is the number of sites and $\beta=1/k_BT$. For $T<T_c$ the situation is not so clear. The distribution develops peaks centered at $\pm m_0$ as shown in Fig. 2, and we may write

$$P(m)=\tfrac{1}{2}[P_1(m+m_0)+P_1(m-m_0)]\ . \tag{9}$$

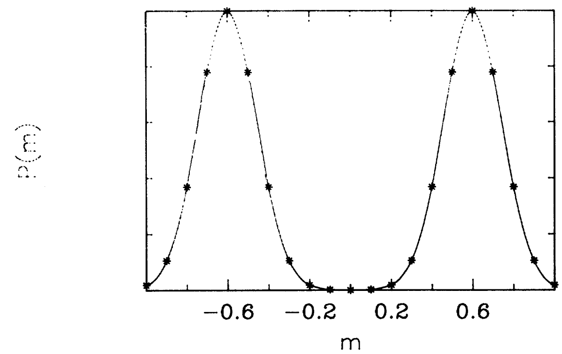

FIG. 2. Probability distribution for the magnetization m fitted to the sum of two Gaussians centered at $m_0=\pm 0.6$. The data points are calculated by Monte Carlo and symmetrized.

The proper thermodynamic averages $\langle m^n\rangle_1$ are taken with respect to $P_1(m)$, not $P(m)$. For example, by (7) we have

$$\langle m^2\rangle=\langle m^2\rangle_1+m_0^2\ , \tag{10}$$

where $\langle m^2\rangle_1$ is the second moment of P_1. This leads to a susceptibility

$$\chi=\beta N(\langle m^2\rangle_1+m_0^2)$$
$$=\chi_1+\beta N m_0^2\ . \tag{11}$$

The second term, being proportional to N, overwhelms the first and leads to an erroneous value for the susceptibility. In fact if the true susceptibility χ_1 is negligible, as it is for $T\to 0$, then m_0 can be found by

$$m_0\approx(\langle m^2\rangle)^{1/2}\ . \tag{12}$$

Unfortunately this procedure does not allow one to determine χ_1, which is the proper susceptibility. In general, calculating higher moments of $P(m)$ will not solve the problem since each new moment introduces a new unknown. However, in the limit of large (but finite) systems we can argue that $P_1(m)$ is Gaussian.

Let us divide the system under consideration into smaller volumes of linear dimension ξ, where ξ is the correlation length. Within each such volume the magnetization per spin will assume a value randomly distributed about m_0 (or $-m_0$). Whatever the distribution within each such volume, if $L/\xi\gg 1$ their cumulative distribution will be Gaussian by the law of large numbers.

Given that $P_1(m)$ is Gaussian, the fourth moment of $P(m)$ is

$$\langle m^4\rangle-3\langle m^2\rangle^2=-2m_0^4\ . \tag{13}$$

Solving for m_0, we have

$$m_0=[(3\langle m^2\rangle^2-\langle m^4\rangle)/2]^{1/4} \tag{14}$$

and for the susceptibility from (10) and (11)

$$\chi_1=\beta N[\langle m^2\rangle-m_0^2]\ . \tag{15}$$

III. RESULTS

The first step in the calculation was to see if we could obtain results similar to those reported in earlier works. We obtained results comparable to those of Hohenberg and Binder,[6] Binder and Landau,[8] Landau and Binder,[9] and Mon and Nightingale[10] for the case $L=D=20$, and $J_s/J_b=1.0$.

We next calculated the magnetization m_0 as a function of J_s/J_b in the range of $0.7\leq J_s/J_b\leq 1.8$ for $\tau=4.0$, $L=10$, and $D=20$ ($r=0.5$). The choice of $\tau<\tau_c=4.52$ ensures that the system is below the transition temperature τ_c and we avoid the problems associated with critical fluctuations. The results appear in Fig. 3, where the curve with the squares refers to the bulk and that with the dots refers to the surface. The same symbols will be used in the remaining graphs unless otherwise stated. As one can see from Fig. 3, below $J_s/J_b=1.14$ the bulk magnetization exceeds that of the surface while above the crossover point the surface magnetization is larger. The value of J_s/J_b at which the magnetization curves for the bulk and surface intersect was found to depend weakly on both the temperature $\tau<\tau_c$, and the aspect ratio r.

We then considered the effect of varying L on the bulk and surface transition temperatures, keeping D fixed at 16 and $J_s/J_b=1.2$. (Note that this ratio is below the critical value 1.55 of Binder and co-workers.) The temperature τ_c was obtained from the maximum in the susceptibility when plotted against T. From Fig. 4 it is evident that for small and large values of r the bulk and the surface have approximately the same critical temperature. However, there is an intermediate range of r values for which the bulk critical temperature lies below that of the surface. We can understand the behavior of the critical temperatures in terms of the longitudinal correlation length ξ_L. When $\xi_L/L\ll 1$ the two free surfaces are uncorrelated and the system behaves essentially as if it were semi-infinite. As we bring the free surfaces closer together, so that $\xi_L/L\approx 1$, the correlations between spins on the free surfaces enhance the surface magnetization, raising the critical temperature for the surfaces. Of course the magnetization of the surfaces tends to magnetize the bulk as well so that, as we see in Fig. 4, the bulk critical temperature is also increased.

Finally, we studied the dependence of τ_c on the ratio J_s/J_b for fixed r. We found that both the transition temperature of the surface and that of the bulk increase with J_s/J_b, and the critical temperature for the bulk tends to fall below that of the surface. For values of J_s/J_b larger than 1.4, it is evident that τ_{cb} of the bulk is different from that τ_{cs} of the surface. In the region of r values in the

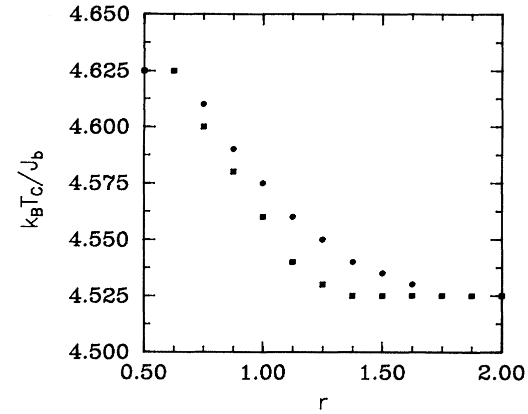

FIG. 4. Critical temperature τ_c vs $r=L/D$ where $D=16$ was kept constant and $J_s/J_b=1.2$. At those values of r where there is only one symbol, $\tau_c=k_BT_c/J_b$ for the surface and for the bulk are equal. Notice that over most of the range, $0.75<r<1.75$, the critical temperature of the surface is larger than that of the bulk.

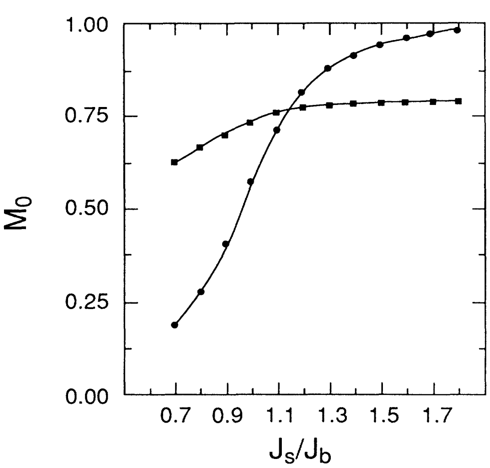

FIG. 3. Magnetization m_0 vs J_s/J_b with $J_b=1.0$ $\tau=4.0$, $L=10$, and $D=20$. The curve with the squares refers to the bulk and that with the dots refers to the surface. The crossing point corresponds to ratio $J_s/J_b=1.14$.

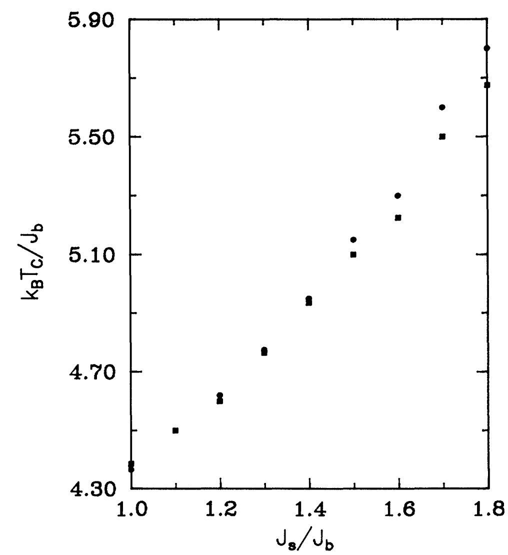

FIG. 5. The graph of $\tau_c=k_BT_c/J_b$ vs J_s/J_b shows the effect of changing coupling constants while leaving the size of the lattice constants [$L=10$, $D=20$ i.e., $r=0.5$]. As in all previous graphs the dots refer to the surface and the squares to the bulk.

range of $0.3 \le r \le 2.0$ we can see that the critical temperatures for the surface and bulk are different even for values as low as $J_s/J_b = 1.2$. In Fig. 5 we show the results obtained for the case $L = 10$, $D = 20$ (i.e., $r = 0.5$). The squares refer to the bulk and the dots refer to the surface.

IV. CONCLUSIONS

By extensive Monte Carlo calculations for different values of $r = L/D$ and J_s/J_b we have a complete picture of the effect of finite width on the surface magnetization. For sufficiently large J_s/J_b the surface magnetizes in the absence of bulk magnetization. This surface magnetization has the effect of shifting the bulk critical temperature to higher values. This shift is most pronounced when $L/\xi_L \sim O(1)$, and we find that the critical value of J_s/J_b approaches the value 1.55 only in the limit $r \to \infty$, i.e., the limit $L/\xi_L \gg 1$ where we have essentially semi-infinite geometry.

The calculations presented here are applicable to thin films, and it would be interesting to see if the behavior we find in the critical temperature for the surface and bulk can be reproduced experimentally by considering a series of films of varying thickness.

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