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Critical temperature of an Ising magnetic film

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In this paper, we studied the critical temperatures T_{cf} of the magnetic-nonmagnetic phase transition of an Ising magnetic film with spin $\frac{1}{2}$ by the mean-field method. We found that, depending on the values of the exchange integrals near the surface region, the film critical temperature may be lower, higher than, or equal to that of the bulk. We also examined the dependence of T_{cf} on the number of film layers in these three cases, and found that the critical temperature of the semi-infinite system is always approached when the film becomes thicker. Some analytical expressions are also presented.

I. INTRODUCTION

Recently, the development of the technique of molecular-beam expitaxy has allowed us to fabricate high-quality thin films.[1] Among them, the magnetic film consisting of a few parallel atomic layers is of particular importance, for it has been used as a convenient model for theoretical and experimental analysis of a variety of magnetic phenomena. For example, it is an ideal model to study the magnetic size effect[2] and surface and interface magnetism.[3] Because it loses symmetry in the direction perpendicular to the surface, it cannot be studied within the conventional framework used for the bulk system. Many approaches have been employed recently in investigating the magnetic behaviors of the model thin-film system, e.g., the finite-size scaling method,[4] the renormalization-group method,[5,6] etc.[7] Though each method has its own advantage over others, there are certain limitations in any of them in treating the film system.

One of the more interesting problems to investigate in a magnetic system is the critical temperature T_c at which the system undergoes a magnetic-nonmagnetic phase transition. Of particular interest is the change of this temperature when the magnetic system becomes finite in a given direction. We know[8] that a semi-infinite system (e.g., a surface system) may present different behaviors from that of the bulk. Briefly, depending on the values of magnetic coupling constants near the surface region, two different behaviors at the surface can be demonstrated: (i) The surface critical temperature is the same as the bulk one when the coupling constant on the surface is smaller than a critical value. (ii) The surface critical temperature may be higher than the corresponding bulk one when the surface coupling is larger than the critical value.

As for the film system, which is finite in one direction, it is well known that its magnetic properties can differ greatly from those of the corresponding bulk.[9–12] A recent experiment[13] showed that the critical temperature of a vanadium film depends on the film thickness and its critical behavior is like that of the two-dimensional system rather than that of the three-dimensional bulk. Due to the finite size and the different environments between atoms in the surface region and in the bulk, we expect that the film transition temperature may be different from the bulk one. In this paper, we study the critical temperature of an Ising magnetic film with spin $\frac{1}{2}$ in the mean-field theory. The results we got are compared with the bulk transition temperature[14] obtained in the same way.

It should be noted that this paper is devoted to a theoretical study of the transition temperatures of magnetic films. We have no specific magnetic materials in mind and our intent is to give a basic understanding of this class of phenomena that one may encounter in such systems.

The method in this paper was previously proposed to investigate the semi-infinite magnetic system by Aguilera-Granja and Moran-Lopes.[14] Here we extend it to fit the film case.

In Sec. II, we give a brief description of the method. The results and discussions are contained in Sec. III.

II. METHOD

Below, we study an Ising magnetic film with spin $\frac{1}{2}$ per atom. We expect the results that we got will also hold for any value of spin. We consider a ferromagnetic (antiferromagnetic) single-crystal film with two free surfaces, which is schematically shown in Fig. 1. The system is subdivided into planes parallel to the surface planes. The coupling constant between spins in the ith and jth layers is denoted by J_{ij}. We use Z_0 and Z_1 to represent the number of nearest neighbors of an atomic site on a single

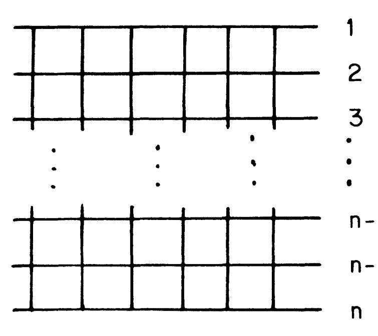

FIG. 1. Magnetic film with parallel planes numbered $1,2,\ldots,n$.

plane and between planes, respectively. In principle, the treatment below is suitable for any number of J_{ij} differing from the bulk value J. For simplicity we assume, in this paper, only J_{00} (the surface coupling constant) and J_{01} (the coupling between the surface and the first layer) to be different from the bulk J.

In the single-site approximation, for each plane there are two different probabilities, i.e., the probabilities of a site being occupied by a spin-up or a spin-down atom. These probabilities are denoted by $P_{i,\sigma}$, where $i=1, 2, \ldots, n$ are the labels of layers and $\sigma=\uparrow,\downarrow$ is the spin index.

We define the magnetic long-range order parameter at the ith plane

$$\eta_i=P_{i,\uparrow}-P_{i,\downarrow},\quad i=1,2,\ldots,n\,. \tag{2.1}$$

For ith plane, we can write the interaction energy U_i between spins in the plane and spins in other planes as

$$U_i=N_{\parallel}(-\tfrac{1}{2}Z_0J_{ii}\eta_i^2-Z_1J_{i,i+1}\eta_i\eta_{i+1})\,, \tag{2.2}$$

where $N_{\parallel}$ is the total number of spins per plane. The entropy of the ith plane is

$$\begin{aligned}S_i&=-k_BN_{\parallel}[P_{i,\uparrow}\ln P_{i,\uparrow}+(1-P_{i,\uparrow})\ln(1-P_{i,\uparrow})]\,,\\&=-k_BN_{\parallel}\left[\frac{1+\eta_i}{2}\ln\left(\frac{1+\eta_i}{2}\right)+\frac{1-\eta_i}{2}\ln\left(\frac{1-\eta_i}{2}\right)\right].\end{aligned} \tag{2.3}$$

The free energy of the film system can be written as

$$F=U-TS=N_{\parallel}\left\{\sum_{i=1}^{n}(-\tfrac{1}{2}Z_0J_{ii}\eta_i^2-Z_1J_{i,i+1}\eta_i\eta_{i+1})+k_BT\sum_{i=1}^{n}\left[\frac{1+\eta_i}{2}\ln\left(\frac{1+\eta_i}{2}\right)+\frac{1-\eta_i}{2}\ln\left(\frac{1-\eta_i}{2}\right)\right]\right\}. \tag{2.4}$$

To obtain the equilibrium values of the order parameters, we must minimize the free energy,

$$\frac{\partial F}{\partial\eta_i}=0,\quad i=1,2,\ldots,n\,, \tag{2.5}$$

which leads to the following coupled equations for $i=1,2,\ldots,n$:

$$\begin{aligned}&-2(Z_0J_{00}\eta_1+Z_1J_{01}\eta_2)+k_BT\ln\left(\frac{1+\eta_1}{1-\eta_1}\right)=0\,,\\&-2(Z_1J_{i-1,i}\eta_{i-1}+Z_0J_{ii}\eta_i+Z_1J_{i,i+1}\eta_{i+1})\\&\qquad\qquad+k_BT\ln\left(\frac{1+\eta_i}{1-\eta_i}\right)=0\,.\end{aligned} \tag{2.6}$$

The number of variables in (2.6) is n.

For an ideal film system, there exists symmetry in the direction perpendicular to the surface, which allows us to write

$$\eta_1=\eta_n,\quad \eta_2=\eta_{n-1},\ldots\,. \tag{2.7}$$

Substituting (2.7) into (2.6), the number of variables in (2.6) is reduced to $n-n/2$.

Near the critical temperature T_{cf}, we can expand the logarithmic terms in Eq. (2.6) as a series powers of η_i. Omitting the higher terms of η_i and keeping only the linear ones, we obtain the set of equations

$$\underline{A}\,\underline{\eta}=0\,, \tag{2.8}$$

where $\underline{A}$ is a matrix of order $n-(n/2)$ with elements

$$\begin{aligned}A_{mn}&=(k_BT_{cf}-Z_0J_{mm})\delta_{mm}\\&\quad-Z_1J_{mn}(\delta_{m+1,n}+\delta_{m,n+1})\end{aligned} \tag{2.9}$$

and $\underline{\eta}$ is a column,

$$\underline{\eta}=\begin{bmatrix}\eta_1\\\eta_2\\\vdots\end{bmatrix}. \tag{2.10}$$

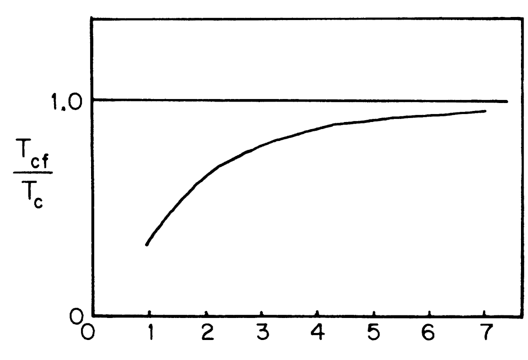

FIG. 2. The critical temperature as a function of the number of layers for a fcc crystal film with (100) surface when $J_{00}=J_{01}=J$.

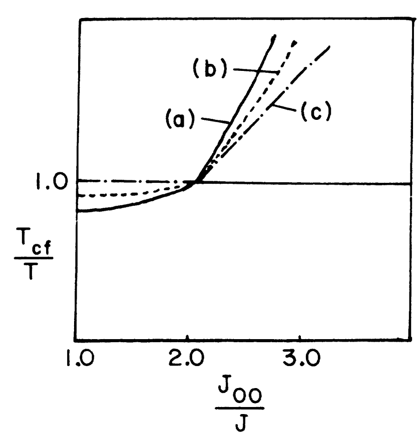

FIG. 3. The dependence of film critical temperature on J_{00}/J when $J=J$ for a, $n=4$; b, $n=6$; and c, $n\rightarrow\infty$.

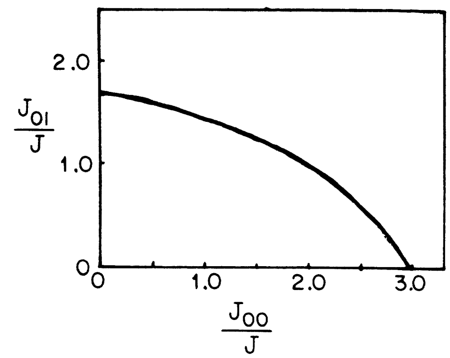

FIG. 4. The boundary for $T_{cf}=T_c$ for films more than two layers thick.

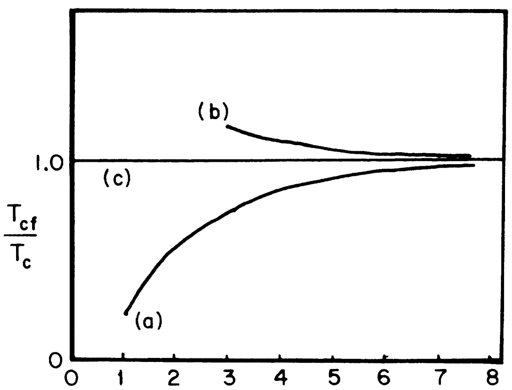

FIG. 5. The dependence of the film critical temperature on the number of layers for a, $J_{00}=0.8J$, $J_{01}=J$, corresponding to the case $T_{cf}<T_c$; b, $J_{00}=2.5J$, $J_{01}=J$, corresponding to the case $T_{cf}>T_c$. c, $J_{01}=J$ and J_{00} assume the values given by (3.1a), (3.1b), and (3.1c), corresponding to $T_{cf}=T_c$.

The critical temperature T_{cf} can be obtained from

$$\det\underline{A}=0\,. \tag{2.11}$$

III. RESULTS AND DISCUSSIONS

Using the method illustrated in the above section, we first study the critical temperatures of a film whose coupling constants in the surface region are the same as the bulk J. Figure 2 shows the results. It can be seen that in this case the film critical temperature is lower than the bulk one, i.e., the film disorders at a lower temperature than the bulk does. Figure 3 shows that as we enlarge the values of J_{00} and J_{01}, the film critical temperature T_{cf} will be raised, and when J_{00} and J_{01} are larger than the critical values $J_{00,c}$ and $J_{01,c}$, the film T_{cf} will be higher than bulk T_c. The exact expressions for the values $J_{00,c}$ and $J_{01,c}$ can be obtained through Eq. (2.11) by letting $T_{cf}=T_c=ZJ/k_B$:

$$J_{00,c}=\frac{Z}{Z_0}J \text{ for } n=1\,, \tag{3.1a}$$

$$J_{00,c}+J_{01,c}=\frac{Z}{Z_1}J \text{ for } n=2\,, \tag{3.1b}$$

$$\left[Z-Z_0\frac{J_{00}}{J}\right]-Z_1\left[\frac{J_{01}}{J}\right]^2=0 \text{ for } n\geq 3\,, \tag{3.1c}$$

where $Z=Z_0+2Z_1$ is the bulk coordination number. The case (3.1c) is schematically shown in Fig. 4. For a magnetic film more than three layers thick and with values (J_{00},J_{01}) laying inside (outside) the curve, it will disorder at temperatures lower (higher) than the bulk one. On the curve, the film disorders at the same temperature as the bulk T_c. One may note that the expressions for critical couplings are the same for a film with more than two layers. This implies at this situation T_{cf} is independent of the film thickness. It can also be seen that the expression (3.1c) for the critical values of $J_{00,c}$ and $J_{01,c}$ for the film is the same as that for the semi-infinite surface system.[14]

In Fig. 5, we present the behavior of T_{cf} as a function of the number of layers n. We first studied the dependence of T_{cf} upon n when the film T_{cf} is lower than bulk T_c. Curve a illustrates that T_{cf} increases as the number of film layers n increases. And when n becomes larger, the film T_{cf} approaches the bulk value rapidly. Next, we examined the relation between the film T_{cf} and its number of layers n, when the film T_{cf} is higher than bulk T_c. In this situation, we can see that contrary to the first case, the film T_{cf} decreases when n increases. This is because the surface region in this case is magnetically harder than the inner region, and as n increases, the strength of the molecular field in the film layers decreases, which leads to a decrease of T_{cf}. Curve c illustrates that when the film T_{cf} equals T_c, the film T_{cf} is independent of the number of film layers as is mentioned in the above paragraph.

We also analyzed the dependence of T_{cf}/T_c on J_{00}/J for several films with different thicknesses. The results are displayed in Fig. 3. The extreme case in these situations is that when the film is infinitely thick, its phase-transition diagram (curve c), as is expected, is the same as that of the semi-infinite system.[8]

Within the above method, we can calculate the magnetization of each layer of the magnetic film through the solution of the nonlinear set of Eqs. (2.6) and (2.7), but to do this would cost somewhat more computing time. This work is now in progress.

In summary, we conclude that the critical temperature of a magnetic thin film may be lower (higher) than or equal to the bulk one, depending on the values of magnetic coupling constants near the surface region. The critical temperature of the semi-infinite system is always approached when the thickness of the film increases.

[1]A. J. Freeman, J. Magn. Magn. Mater. **15-18**, 1070 (1980).

[2]M. N. Barber, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and J. L. Lebowits (Academic, New York, 1983), Vol. 8.

[3]H. Nakanishi and M. E. Fisher, J. Chem. Phys. **78**, 3279 (1983).

[4]M. E. Fisher and H. Nakanishi, J. Chem. Phys. **75**, 5857 (1981).

[5]R. R. dos Santos, L. Sneddon, and R. B. Stinchcombe, J. Phys. A **14**, 3329 (1981).
[6]A. M. Nemirovsky and K. F. Freed, J. Phys. A **18**, L319 (1985); *ibid.* **19**, 591 (1986).
[7]E. Eisenriegler, Z. Phys. B **61**, 299 (1985).
[8]K. Binder, in *Phase Transitions and Critical Phenomena* (Ref. 2).
[9]R. Richter, J. G. Gay, and J. R. Smith, Phys. Rev. Lett. **54**, 2704 (1985).
[10]R. H. Victora and L. M. Falicov, Phys. Rev. B **31**, 7335 (1985).
[11]C. Rau, C. Schneider, G. Xing, and K. Jamison, Phys. Rev. Lett. **57**, 3221 (1986).
[12]D. Pescia, G. Zampieri, G. L. Bona, R. F. Willis, and F. Meier, Phys. Rev. Lett. **58**, 9 (1987).
[13]C. Rau, G. Xing, and C. Liu, Phys. Lett. A **135**, 227 (1989).
[14]F. Aguilera-Granja and J. L. Moran-Lopes, Phys. Rev. B **31**, 7146 (1985).