

Critical behavior of magnetic films in the Ising model

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The Curie temperature $T_C(l)$ for Ising spin films calculated analytically as a function of l , the number of monolayers in the film, is compared with available data. When the exchange coupling between nearest neighboring pairs is assumed to be uniform, the normalized $T_C(l)$ is generally higher than what is observed, especially in the dimensionality crossover region in which $T_C(l)$ increases with l much too fast. To reproduce the experimental data measured by various groups on ultrathin Ni films in (001) orientation, we calculate the normalized transition temperature $T_C(l)/T_C(\text{bulk})$ to fourth order in the variational cumulant expansion for a face-centered-cubic lattice with variable anisotropic exchange coupling J_{ij} . It is shown that the data can be very well described by adjusting J_{ij} for a few monolayers near the surfaces. The results are consistent with the finite-size scaling law outside the region of dimensionality crossover, providing a clue to the number of monolayers influenced by the presence of surfaces. [S1063-651X(97)12709-X]

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I. INTRODUCTION

Advances in material growth technique such as molecular-beam epitaxy and scanning tunneling microscopy have prompted much research in recent years on thin films, heterostructures, and superlattices of various crystalline solids. The study of physical properties of solids as the dimensionality reduces is in general not only of fundamental interest but also of technological importance. In particular, studies of ultrathin magnetic films have revealed a number of novel phenomena that would not have been expected in either the three-dimensional (3D) or two-dimensional (2D) case [1–6]. Of particular interest is the critical behavior of magnetic films, from which one can explore and test the universality hypothesis.

Experimentally, there has been a great deal of work on the measurement of Curie temperature T_C and critical exponents of thin magnetic films [7–17]. T_C as a function of the film thickness for magnetic materials is reported, and various exponents are investigated. Theoretically, magnetic films and heterostructures are mostly treated either by numerical studies [18,19] or in the mean-field approximation [6]. A new approach to the critical temperature of Ising films has been proposed recently [20]. For the first time, $T_C(l)$ was calculated analytically to any order of accuracy in the variational cumulant expansion (VCE), where l is the number of monolayers in the film.

The theory is general enough that Ising spin films with different lattice structures can be treated as well. It is found that T_C depends strongly on the lattice structure due mainly to the coordination number [21]. The larger the coordination number, the higher the Curie temperature is for the same film thickness, which is qualitatively consistent with the well-established statement that adding more coupling bonds leads to higher T_C [22]. The anisotropic exchange coupling and

next-nearest-neighbor (nnn) interaction can also be handled by this theory without difficulty [23]. It is demonstrated in Ref. [23] that a slightly weaker interlayer coupling strength J_{\perp} than the intralayer J_{\parallel} may result in a significant decrease in T_C . On the other hand, an inclusion of a rather weak nnn interaction can increase T_C appreciably.

When the theoretical transition temperature $T_C(l)$ for films of a face-centered-cubic (fcc) lattice from Ref. [21] is compared with experimental data measured on ultrathin films of Ni(001) grown on the Cu(001) substrate and Ni(111) on W(110) and Re(0001) [24], it is observed that the theoretical $T_C(l)$ is generally higher than the data. More importantly, it rises too fast with increasing l in the region of dimensionality crossover. This is mainly because exchange coupling was assumed in Ref. [21] to be uniform throughout the film. Since the exchange coupling near surfaces is expected to be weaker than in the bulk, and the intraplane coupling is generally stronger than interplane coupling in a thin film, we calculate $T_C(l)$ of fcc films by allowing variable anisotropic coupling strengths. It is shown that the experimental data for fcc Ni(001) films can be reproduced. The magnetization of Ni(111) is in-plane anisotropy, and hence is not described by the Ising model.

It has been pointed out in the literature that the cubic symmetry of the fcc lattice is broken near the film surface, resulting in uniaxial anisotropy, and this effect is stronger the thinner the film [25]. Thus it may be justified to treat the ultrathin fcc Ni films of $l \leq 6$ by the Ising model. In fact it was argued a long time ago by Néel that the surface-induced anisotropy is of the easy-axis type [26]. His conclusion was supported by numerical calculations on the ground state of iron film on copper substrates [27], implying that for Ni(001) the magnetization should be normal to the film.

We review the theory in Sec. II, in which the continuity of higher-order internal energy at the critical point is also dis-

cussed. In Sec. III we discuss briefly how the results obtained for uniform magnetic films can be improved to account for experimental data. The modification of the method for the treatment of films with variable exchange coupling is outlined in Sec. IV, where a comparison of our results with experiments is also presented.

II. THEORY

The action S for an Ising spin system is

$$\begin{aligned} S &= -\frac{1}{s^2} \sum_{\langle i,j \rangle} \frac{J_{ij}}{k_B T} s_i^z s_j^z \\ &= \frac{1}{k_B T} X, \end{aligned} \quad (1)$$

where the sum is over nearest-neighbor pairs only. The trial action is assumed to be

$$S_0 = \frac{1}{s} \sum_i \xi_i s_i^z, \quad (2)$$

where ξ is introduced as a variational parameter. The partition function for the system is

$$\begin{aligned} Z &= e^{-W} \\ &= \sum e^S \\ &= \sum e^{S-S_0} e^{S_0} \\ &= Z_0 \langle e^{S-S_0} \rangle_0, \end{aligned} \quad (3a)$$

where W stands for the free energy, and the Boltzmann ensemble average is defined as

$$\langle \cdot \rangle_0 = \frac{1}{Z_0} \sum_{t_i} (\cdot) \exp(\xi_i t_i). \quad (3b)$$

For a noninteracting system, the free energy is $W_0 = N \ln(\cosh \xi)$, where N denotes the number of lattice points. The corresponding partition function is then

$$Z_0 = e^{-W_0}. \quad (3c)$$

From Eqs. (3), we find the free energy

$$\begin{aligned} W &= W_0 - \ln \langle e^{S-S_0} \rangle_0 \\ &= W_0 - \ln \left[1 + \sum_{n=1}^{\infty} \frac{1}{n!} \langle (S-S_0)^n \rangle_0 \right] \\ &= W_0 + \sum_{\nu=1}^{\infty} \frac{(-1)^\nu}{\nu} \left[\sum_{n=1}^{\infty} \frac{1}{n!} \langle (S-S_0)^n \rangle_0 \right]^\nu \end{aligned} \quad (4)$$

by expanding the exponential and logarithmic functions. Equation (4) takes a much simpler form by introducing the VCE. Up to the order m , we have

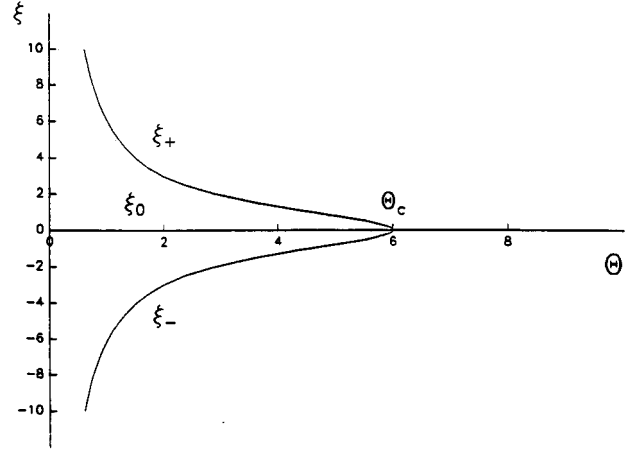


FIG. 1. Analytic behavior of solutions to Eq. (7). For $\Theta > \Theta_c^{(1)}$, there is only one solution corresponding to the minimum of the free energy $W_{\text{eff},1}$. For $\Theta < \Theta_c^{(1)}$, there are three solutions ξ_{\pm} and ξ_0 corresponding to minimum and maximum of the free energy, respectively. The critical temperature is therefore given by the bifurcation point $\Theta = \Theta_c^{(1)}$.

$$\begin{aligned} W &\approx W_0 - \sum_{n=1}^m \frac{1}{n!} \langle (S-S_0)^n \rangle_c \\ &= W_{\text{eff},m}, \end{aligned} \quad (5)$$

where the subscript $\langle \cdot \rangle_c$ denotes the cumulant average over the Boltzmann weight e^{S_0} . The relation between the two averages of a quantity can most easily be established by a comparison of the corresponding terms in expansions of Eqs. (4) and (5).

For simplicity, we consider a uniform spin- $\frac{1}{2}$ film with $J_{ij} = J$ and $\xi_i = \xi$. It is noted that the formalism is completely general, and can be applied to Ising systems with different spin values and with spatial dependent J_{ij} 's. The first-order free energy is then

$$\frac{1}{N} W_{\text{eff},1} = -\ln(2 \cosh \xi) - \frac{d}{\Theta} y^2 + \xi y, \quad (6a)$$

$$y = \tanh \xi, \quad (6b)$$

where $\Theta = k_B T$, and d stands for the dimensionality. By minimizing the free energy, we find

$$\tanh \xi = \frac{\Theta}{2d} \xi, \quad (7)$$

which determines the variational parameter.

The solution of Eq. (7) depends on the slope $\Theta/2d$ of the straight line $\zeta = \xi\Theta/2d$, and the analytic behavior of these solutions is illustrated in Fig. 1. When $\Theta \geq \Theta_c$, there is only one solution $\xi_0 = 0$ corresponding to the minimum of $W_{\text{eff},1}$. When $\Theta < \Theta_c$, there are three solutions ξ_0 and ξ_{\pm} corresponding to the maximum and minima of $W_{\text{eff},1}$, respectively. Thus the critical value Θ_c is determined by the bifurcation point of the function $W_{\text{eff},1}(\Theta, \xi)$ for $\xi = 0$. The parameter ξ has the properties of the order parameter in Lan-

dau theory. On the basis of this analysis, a conjecture was proposed [20] that, to any order of the VCE, the critical temperature is determined by locating the bifurcation point of the free energy $W_{\text{eff},m}$, namely, by the condition

$$\frac{\partial^2}{\partial \xi^2} W_{\text{eff},m}(\Theta_c^{(m)}, \xi) \Big|_{\xi=0} = 0. \quad (8)$$

An important remark is in order at this point. Without this conjecture one can only be sure that in the first-order approximation, $\xi=0$ corresponds to a minimum of the function $W_{\text{eff},1}(\Theta, \xi)$ for $\Theta \geq \Theta_c^{(1)}$. This presents a difficulty in higher-order calculations of the internal energy. That $\xi=0$ corresponds to an extremum of $W_{\text{eff},m}(\Theta, \xi)$ for any order m in the high-temperature regime is understandable because the free energy obtained from the VCE is identical to that from the well-known linked-cluster expansion to every order of approximation in the high-temperature limit [28]. For $\Theta < \Theta_c$; however, the value of ξ_+ (or ξ_-) from the first-order approximation is also employed in higher-order calculations, and hence leads to a discontinuity in the internal energy. This is the origin of the so-called ‘‘unwanted first-order phase transition’’ [29,30]. With conjecture (8), it is easily shown that $W_{\text{eff},m}(\Theta, 0)$ remains analytic for $\Theta < \Theta_c^{(m)}$ which is always lower than $\Theta_c^{(1)}$. The extremum condition like Eq. (7) for the first order can be solved for every order. In fact we have approximately solved these equations for a 2D Ising model up to $m=8$ for Θ not far below $\Theta_c^{(m)}$ [31]. Once Θ becomes less than $\Theta_c^{(m)}$, the minimum free energy is characterized by $\xi_{\pm}^{(m)}$. As long as the variational parameter is so chosen that the free energy remains a minimum continuously across the critical point, there will not be any discontinuity in the internal energy, and hence no first-order phase transition will appear. On the other hand, if $\xi=0$ is assumed in the low-temperature region, it is found that the free energy does not show singular behavior and there is no phase transition at all. Therefore, the conjecture is justified.

As shown in Ref. [20], one can prove by mathematical induction that Eq. (8) reduces to

$$\left[\frac{\partial^2}{\partial \xi^2} \langle S^{m-1} \rangle_c - \frac{1}{m} \frac{\partial^2}{\partial \xi^2} \langle S^m \rangle_c \right]_{\xi=0} = 0, \quad m \geq 2, \quad (9)$$

which immediately yields the critical temperature

$$k_B T_c^{(m)} = \frac{1}{m} \left[\frac{\partial^2}{\partial \xi^2} \langle X^m \rangle_c \Big/ \frac{\partial^2}{\partial \xi^2} \langle X^{m+1} \rangle_c \right]_{\xi=0}, \quad m \geq 2. \quad (10)$$

Note that $X = k_B T S$ as defined in Eq. (1) and the energy is in the unit of J . The cumulants $\langle S^m \rangle_c$ are calculated from the first-order moment

$$\begin{aligned} \langle S_0 \rangle &= \frac{1}{2} \sum_{i,j} \beta_{ij} y_i y_j \\ &= \langle S \rangle_c, \end{aligned} \quad (11)$$

with the help of the recursion formula for moments

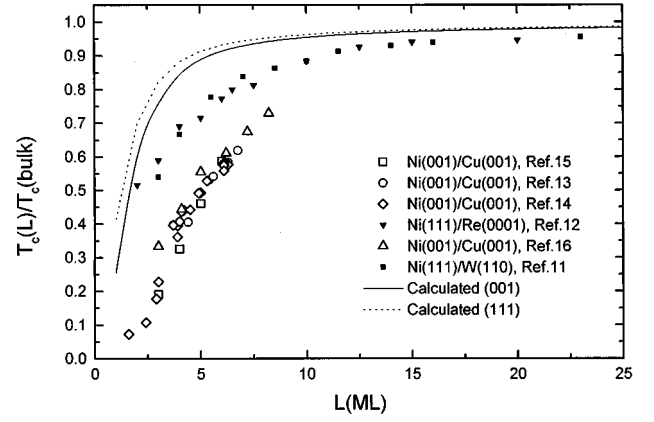


FIG. 2. Normalized Curie temperature calculated on the Ising model with uniform coupling for fcc films and compared with experimental data measured on ultrathin Ni films. The dashed and solid lines represent T_c for a film with (111) and (001) surfaces, respectively.

$$\langle S^{p+1} \rangle_0 = \left[\frac{1}{2} \sum_{\langle i,j \rangle} \beta_{ij} \frac{\partial^2}{\partial \xi_i \partial \xi_j} + \sum_{\langle i,j \rangle} \beta_{ij} y_i \frac{\partial}{\partial \xi_j} + \langle S \rangle_c \right] \langle S^p \rangle_0, \quad (12)$$

and relations between $\langle S^p \rangle_c$ and $\langle S^p \rangle_0$ where β_{ij} measures J_{ij} in the unit of $k_B T$, and p is an integer.

III. UNIFORM MAGNETIC FILMS

For a uniform spin system with $J_{ij}=J$ and $\xi_i=\xi$, the transition temperature has been calculated to fourth order for films with simple cubic (sc) and body-centered-cubic (bcc) lattices, and to third order for the fcc lattices [20,21]. It is found that the coordination number of the lattice is responsible for the strong dependence of T_c on the lattice structure. Generally speaking, the results are satisfactory for all three structures in the sense that they converge quickly to the exact 2D result and to the best known 3D values for all three cases that can be found in the literature. The variation of $T_c(l)$ with the film thickness is also qualitatively similar for all cases.

In Fig. 2 we show that $T_c(l)$ calculated for uniform films rises too fast as the thickness increases in the dimensionality crossover region, namely, when l changes from 1 (2D) to ~ 6 (nearly 3D) monolayers. This is true for all three lattice structures. The main reason is the fact that we have assumed the same exchange coupling J_{ij} between nearest neighbors throughout the uniform film. Near the surfaces of a film, however, the coupling is expected to be somewhat weaker than that in the bulk. As a matter of fact, the exchange coupling in a thin film may be very different from that in the bulk. It has been suggested [32] and confirmed [33] that the surface layer of a ferromagnet follows a spin wave $T^{3/2}$ law, but with a surface Bloch parameter $b_s = \alpha_s b_{\text{bulk}}$ enhanced by a factor $\alpha_s = 2$ if the exchange remains homogeneous up to the surface. It has also been shown later that a reduced exchange strength in the surface can cause a further enhancement, resulting in $\alpha_s > 2$ [34]. Such an enhancement of the surface spin wave parameter has been observed in recent experiments [35,36], indicating the reduced exchange cou-

pling in the surface. Furthermore, the intraplane coupling is also expected to be stronger than the interplane coupling in ultrathin films. It is therefore natural to assume that the discrepancies may be removed by allowing the exchange integral to vary.

Let the intraplane and interplane coupling integrals be $J_{\parallel} = a_i J$ and $J_{\perp} = b_i J$, respectively. The subscript i labels the monolayer with $i=1$ for the surface and $a_i = b_i = 1$ for $i \geq 4$. It is assumed further that $a_i < a_{i+1} < 1$ and $a_i > b_i$. As we shall discuss later, the normalized $T_C(l)$ calculated for uniform coupling is, in every case, consistent with the finite-size scaling law, until the number of monolayers in the film reduces to ~ 10 . This implies that the 3D correlation length ν starts to lose its meaning for $L \approx 10$. Thus it appears plausible that the exchange coupling in monolayers near the surface is influenced by the presence of surfaces. It turns out, however, that the data can be fitted with fewer monolayers affected by the surfaces than expected. This will become clear in Sec. IV.

IV. FILMS WITH VARIABLE EXCHANGE COUPLING

Since the system is no longer homogeneous, topologically equivalent graphs cannot be simply grouped together in the graph counting as before. Every term has to be individually computed. To illustrate the procedure of the calculation, we consider the second-order calculation for a film of 3 ML. Following the notation of Ref. [21], we find

$$\begin{aligned} \langle X \rangle_c &= (6a_1 + 6b_1 + 3a_2)Jy^2 \\ &= D_1 J y^2, \end{aligned} \quad (13a)$$

$$\begin{aligned} \langle X^2 \rangle_c &= J^2 [(6a_1^2 + 6b_1^2 + 3a_2^2)(1-y^2)^2 + (72a_1^2 + 54b_1^2 \\ &\quad + 36a_2^2 + 72a_1 b_1 + 72a_2 b_1)y^2(1-y^2)] \\ &= J^2 [D_2(1-y^2)^2 + D_3 y^2(1-y^2)], \end{aligned} \quad (13b)$$

where $y = \tanh \xi$. The second step in Eqs. (13) defines the structure factors D_i for $i=1, 2$, and 3. Inserting Eqs. (13) into Eq. (10), we find immediately that

$$k_B T_C^{(2)}(3)/J = \frac{D_3 - D_2}{D_1}. \quad (14)$$

The procedure remains straightforward, but higher-order calculations now become considerably more involved. We calculated $T_C(l)$ for a fcc film to fourth order. The final expression is rather cumbersome, and hence is not reproduced here. By adjusting the coefficients a_i and b_i for a couple of monolayers near the surfaces, excellent agreement with data can be achieved by gradually decreasing the coupling strength towards the surface. As a matter of fact, we find that it is sufficient to allow weaker coupling in only the first two monolayers for (001) orientation. The least number of coefficients that fit the data are posted in Table I. It is interesting to point out that the surface coupling reduction we find in this fitting is qualitatively consistent with recent measurements of the enhancement of the spin-wave parameter [35,36].

TABLE I. Relative strength of the exchange integral in the neighboring monolayers near surfaces of the Ni(001) film.

Monolayer No.	1		2		3	
Relative strength	a_1	b_1	a_2	b_2	a_3	b_3
fcc(001)	0.17	0.14	0.66	0.45	1.00	0.76

Before we discuss the fitting, it is interesting to note from Fig. 2 that the data split into two branches depending on the growth surfaces (001) and (111) of the sample. At this point it is necessary to remark that the interlayer distances are different for the two orientations, and the difference is not exhibited in the figure in which the abscissa is the number of monolayers. It is also worth pointing out that the two sets of Ni(111) data are not distinguishable although the samples are grown on substrates of different symmetries. This may suggest that $T_C(l)$ is determined by the common feature of Ni film instead of the symmetry of substrates. That the Curie temperature for a (111) surface is generally higher than that for a (001) surface for ultrathin fcc films can easily be understood by considering the coordination number once more. In the case of (111) surfaces, every lattice site of a fcc film has six intraplane nearest neighbors (nn's) and three interplane nn's on either side. On the other hand, both the intraplane and interplane nn numbers are equal to 4 in a (001) fcc film. The fact that $T_C(111) > T_C(001)$ implies that $J_{\parallel} > J_{\perp}$ in thin films, since each coupling bond contributes to T_C on equal footing. When the thickness increases, the exchange coupling becomes isotropic in the central part of the film, and hence the film behaves like a bulk.

The magnetization of Ni(111) films is in-plane. It is therefore more appropriately described by the xy model. The present formalism is being extended to the xy model for a calculation of $T_C(l)$ which will be reported in the future. For ultrathin Ni(001) films, the magnetization is normal to the plane. The calculated results are plotted along with the data in Fig. 3. We emphasize that, to our knowledge, up to the present time, there have been only two data points for $l \geq 7$ available from the four sets of measurements. Because of the reorientation of magnetization observed in fcc Fe films [37], the Ising model may not apply to films of $l \geq 7$. However, on

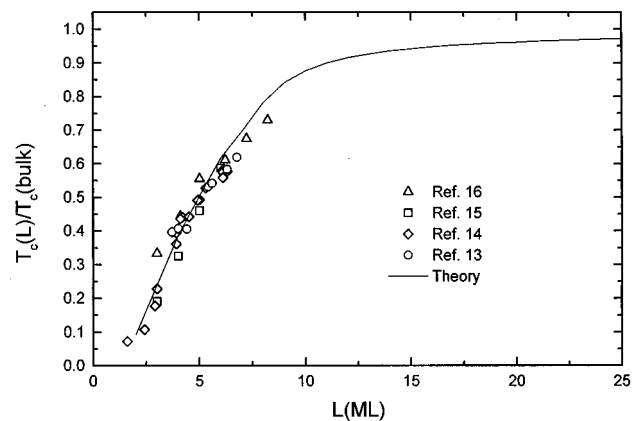


FIG. 3. Normalized Curie temperature calculated on the Ising model for fcc films with variable exchange coupling and compared with experimental data measured on ultrathin Ni(001) films.

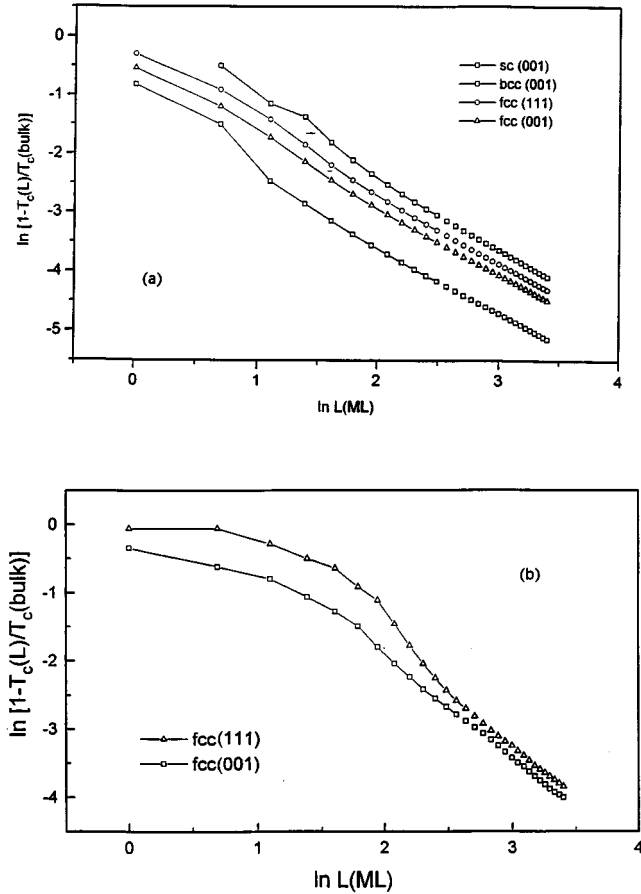


FIG. 4. Illustration of the finite-size scaling for films with (a) uniform coupling and (b) variable coupling.

the basis of the fact that Ising model and isotropic Heisenberg model yield almost the same $T_C(l)$ for $l \geq 7$ [38], it is plausible to assume so because as far as T_C is concerned the thicker film behaves like a 3D bulk.

The transition temperature shift exponent λ , which is the inverse of the 3D correlation length exponent ν [39], can be found from our theoretical results. The shift exponent is defined by

$$\frac{T_C(\text{bulk}) - T_C(l)}{T_C(\text{bulk})} = C_0 l^{-\lambda}, \quad (15)$$

where C_0 is a constant and may be interpreted as the number of monolayers under which the Curie temperature vanishes. In Fig. 4(a), we plot $\ln[1 - T_C(l)/T_C(\text{bulk})]$ versus $\ln l$ for uniform films of all four cases. It is interesting to note that as l decreases, the four straight lines are basically parallel with

a common slope which yields $\lambda = 1.280$. This plot also shows that all four curves start to deviate from the straight line at about $L \sim 10$. On the other hand, Eq. (15) fails completely for $L \leq 5$ if it may be regarded as approximately acceptable for $5 < L \leq 10$. Thus the shift exponent loses its meaning in the dimensionality crossover region in which the finite-size scaling law is supposed to be invalid. This also indicates, to some extent, the depth that surface effects can penetrate into the bulk, suggesting the number of monolayers on which the coupling strength is modified by the presence of surfaces.

For magnetic films with variable coupling, $T_C(l)$ presented in Fig. 3 are inserted into Eq. (15). The resulting log-log plot for Ni(001) is shown in Fig. 4(b) along with that for Ni(111) obtained just by fitting the data. The situation is similar to the case of uniform films. However, the two lines are straight and parallel for $L \geq 15$, indicating the wider region of dimensionality crossover observed in experiments. The shift exponent in this case is $\lambda = 1.439$, which implies a 3D correlation length component $\nu = 0.695$. Therefore it seems that the concept of finite-size scaling is verified by experiments down to a film thickness of ~ 15 ML. Although the present calculation cannot claim the universality of the finite-size scaling, there is no indication of any violation outside the region of dimensionality crossover for all cases considered.

In conclusion, we have found that the lattice dependence of the absolute Curie temperature is mainly a reflection of the coordination number. The uniaxial nature of Ising model has its limitation in applications to realistic samples. It should be of interest to mention that the present theory can be applied to quantum spin films on the isotropic Heisenberg model with no difficulty. A uniform spin film on the isotropic Heisenberg model, however, predicts no phase transition when $l = 1$, and hence cannot account for the data. It is necessary to include anisotropic interactions arising from low-dimensional spin fluctuations [40], which is considered to be responsible for the ordering [41]. Further research along this line is being carried out, and results will be published elsewhere in the future.

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