Critical properties of thin quantum and classical Heisenberg films

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The dependence of the critical temperature $T_c(l,\Delta)$ on film thickness l and ratio of surface (J_s) to bulk (J_b) exchange interaction strengths $\Delta \equiv J_s/J_b - 1$ in quantum and classical Heisenberg models are studied by using the effective-field theory within the framework of the differential operator technique. It is found that for $\Delta < \Delta_c$ (where Δ_c depends on the specific model), the critical temperature $T_c(l,\Delta)$ of the film is smaller than the corresponding bulk critical temperature T_c^b of the infinite system and as l is increased, T_c also increases approaching T_c^b for large values of l. However, in the case of $\Delta > \Delta_c$, T_c is larger than both the bulk T_c^b and the surface T_c^s critical temperatures of the corresponding semi-infinite systems and, as the film thickness l further increases, T_c decreases and approaches, for large value of l, the surface magnetic transition T_c^s.

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I. INTRODUCTION

Recently, the study of magnetic multilayer has attracted considerable attention and has been stimulated by recent technological progresses. In particular, modern high-vacuum techniques, such as the epitaxial growth ones, allow us to fabricate very thin magnetic films of controllable thickness.[1] The increasing technological importance of nanometer-scaled magnets results from the general trend of miniaturization in technical applications. These interesting magnetic properties of magnets on a nanometer scale provide both interesting opportunities for technology and questions for basic research.

On the other hand, from both the experimental and theoretical points of view, one particularly important phenomenon is the dependence of the transition temperature with respect to several parameters, such as the film thickness (l), the geometrical structure or the composition of the film and magnetic excitation. *A* technique or physical complication in some ultrathin films is the fact that experimentally it has been observed that T_c is finite (not null) only when $l > l_{\min}$, [an example is Ni films on Cu(100),[2] where a monolayer film $l=1$ is nonmagnetic]. Thus the search for monolayer film of elements which are nonmagnetic in bulk material remains a topic of considerable interest. Moreover, the dimensionality (two or three dimensions) in the thin films is not well established, therefore we can explore and test the universality hypothesis.

Theoretically, to fit the experimental results of the critical temperature in thin films, the following scaling relation has been used:[3–5]

$$T_c(l) = T_C(\infty)\left(1 - \frac{A}{l^{\lambda}}\right), \tag{1}$$

where $T_C(\infty)$ is the Curie temperature of the bulk and the parameters A and λ depend on the thickness of the films. The constant A may be interpreted as the number of monolayers under which the Curie temperature vanishes.

Some authors have employed an Ising model to study the dependence of T_c as a function of l.[6–9] The scaling theory predicts in Eq. (1) $\lambda = 1/\nu$, where ν is the correlation length exponent for the bulk system [three dimensional (3D)]. For example, the expected λ values for the 3D Ising and Heisenberg models are $\lambda = 1/0.6294 = 1.5884$,[10] and $\lambda = 1/0.7048 = 1.419$,[11] respectively. The Heisenberg model in thin films and superlattices has also been previously treated in the literature.[12–14] In this case, the Mermin-Wagner theorem[15] requires the transition temperature to vanish for the two-dimensional isotropic Heisenberg limit. However, it has been shown that even a very small amount of anisotropy may lead to magnetic order with substantial finite transition temperature[16,17] even for two dimensions.

Another interesting aspect in the study of magnetic films is concerning the dimensionality crossover region, when l changes from $l = l_{\min}$ (2D) to $l = l_1$ (nearly 3D). Usually, $l_1 = 6$ monolayers. One expects that near the surface of a film the couplings are somewhat weaker than in the bulk. Comparison of critical temperatures for films obtained by theoretical models[19] with experimental data measured on ultrathin films of Ni(001) grown on the Cu(001) substrate and Ni(111) on W(110) (Ref. 20) have shown that $T_c(l)$ is higher than the experimental results. More importantly, it rises too fast with increasing l in the region of dimensionality crossover. This discrepancy can be ascribed to the fact that the exchange coupling was assumed to be uniform throughout the film and also to the use of the Ising model to simulate the experimental data. In addition, in Ni(111) films the magnetization is in plane and therefore is more appropriately described by the *XY* model, while the ultrathin Ni(001) films, the magnetization is normal to the plane and the use of the Ising model should be justified (used as a first approximation) *a priori*, but the fact that $T_c[\mathrm{Ni(111)}] > T_c[\mathrm{Ni(001)}]$ physically implies that the use of this Ising model to simulate the Ni(001) films is not adequate. To theoretically describe the experimental data of the magnetic Ni(001) films the Heisenberg model should be more appropriate since $T_c(XY) > T_c$ (Heisenberg).

From the theoretical point of view, one of the models more widely used to study the magnetic properties of surfaces is the Ising model. In this paper, the quantum and classical Heisenberg thin films are treated by the framework of

the effective-field theory (EFT). The EFT is based on the use of rigorous correlation identities as a starting point and utilizes a differential operator technique, introduced by Honmura and Kaneyoshi.[21] Although without introducing mathematical complexities, this approach, which is conceptually as simple as the standard mean-field approximation (MFA), shares with other methods a great versatility. These method may explicitly and systematically include correlation effects. Here, the formalism is illustrated by employing its simplest approximation version, in which spin-spin correlations are neglected.

Owing to its simplicity, the EFT has been exhaustively used to study a very large variety of problems such as percolation, ordered and disordered classical and quantum spin models, bulk and surface critical behavior of spin models, and, more recently, also the quantum Heisenberg model,[22–24] *n*-vector model,[25,26] Ising thin films,[8,9,13,14] and semi-infinite quantum spin-$\frac{1}{2}$ Heisenberg model.[27–29] In all the cases studied so far, it has turned out that this simple method leads to quite good results for the critical coupling even when the cluster sizes are taken by just using the simplest choice. On the other hand, the values of the critical exponents are all classical.

Our major concerns are the dependence of the transition temperature on the thickness of individual constituents in the cell and the influence of the value of the spin S ($\frac{1}{2}$ finite or ∞) on the phase-transition temperature. In Sec. II we outline the formalism and derive the equation that determines the critical temperatures of the film as function of interactions, spin, and film thickness. The phase diagrams of the film are discussed in Sec. III. Finally, Sec. V is devoted to a brief conclusion.

II. MODEL AND FORMALISM

The aim of this work is to investigate the phase diagram of quantum and classical Heisenberg thin films consisting of l two-dimensional layers on a simple cubic (001) lattice. The reduced Hamiltonian of the system is given by

$$\mathcal{H}=-\beta H=\sum_{\langle ij\rangle} K_{ij}\mathbf{S_i}.\mathbf{S_j}, \tag{2}$$

where $\beta=1/k_BT$, k_B the Boltzmann constant, $\mathbf{S_i}=(\mathbf{S_i^x},\mathbf{S_i^y},\mathbf{S_i^z})$ are spin-$\frac{1}{2}$ operators and spin vectors of $n^{1/2}$ ($n=3$ for the Heisenberg model) length at site i for the quantum and classical systems, respectively, and $K_{ij}=\beta J_{ij}$ represents the reduced exchange interaction between all pairs $\langle ij\rangle$ of nearest-neighboring sites. We choose the following values for the parameter K_{ij}: if the sites i and j are at the surface planes (labeled as $p=0$ and $p=l$, respectively), we take $K_{ij}=K_s$ (surface exchange interaction J_s); for all other neighboring sites, $K_{ij}=K_b$ (bulk exchange interaction J_b).

The thermal expectation value of a general function of spin components Ω_N can be obtained by the exact generalized Callen-Suzuki identity,[30–32]

$$\langle\Omega_N\rangle=\left\langle\frac{\mathrm{Tr}_N\Omega_N\exp(\mathcal{H}_\mathcal{N})}{\mathrm{Tr}_N\exp(\mathcal{H}_\mathcal{N})}\right\rangle, \tag{3}$$

where the partial trace Tr_N is taken over the set of N spins variables specified by a finite system Hamiltonian $\mathcal{H}_\mathcal{N}$, Ω_N is a thermodynamic quantity which is a function of all the N spins of the cluster, and $\langle\cdots\rangle$ indicates the canonical thermal average taken over the ensemble defined by the complete Hamiltonian $\mathcal{H}$.

In order to apply the EFT to the present model, we consider a simple cluster with two spins located at each pth layer ($p=0, 1, 2\cdot\cdot, l$). The Hamiltonian for this cluster then reads

$$\mathcal{H}_2^{(p)}=K_p\mathbf{S_{1p}}\cdot\mathbf{S_{2p}}+\mathbf{a_{1p}S_{1p}^z}+\mathbf{a_{2p}S_{2p}^z}, \tag{4}$$

where $a_{jp}=K_p\Sigma_\delta S_\delta^z+K_{p+1}S_{j(p+1)}^z+K_{p-1}S_{j(p-1)}^z$, δ labels the nearest neighbors of the spin S_{jp}^z in the pth layer, $S_{j(p\pm1)}^z$ is the corresponding nearest neighbor of the spin S_{jp}^z in the $(p\pm1)$th layer, respectively, and $K_0=K_l=K_s$, $K_p=K_b$ for $p\neq0$, l and $K_{-1}=0$.

Using Eq. (4) in Eq. (3) in the axial approximation,[22,23] and also the differential operator technique (see Ref. 22), the average magnetization at the pth layer $m_p=\langle(S_{1p}^z+S_{2p}^z)/2\rangle$ is expressed by

$$m_p=\langle\exp(a_{1p}D_x+a_{2p}D_y)\rangle g_p(x,y)|_{x,y=0}, \tag{5}$$

with

$$g_p(x,y)=\frac{\sinh(x+y)}{\cosh(x+y)+\exp(-2K_p)\cosh\sqrt{(x-y)^2+4K_p^2}} \tag{6}$$

for the quantum case and

$$g_p(x,y)=\frac{\sinh(x+y)}{\cosh(x+y)+\phi(K_p)\cosh(x-y)} \tag{7}$$

for the classical Heisenberg model, where $\phi(x)=[1-L(3x)]/[1+L(3x)]$ and $L(x)=\coth(x)-1/x$ is the Langevin function.

To evaluate the exponential factors appearing in Eq. (5) we use the van der Waerden identity for the two-state spin system [i.e., $\exp(\lambda S_i^z)=\cosh(\lambda)+S_i^z\sinh(\lambda)$] to express the layer magnetization m_p ($p=0-l$) in terms of multisite correlation functions. However, it is clear that if we try to treat exactly all the boundary spin-spin correlations present in Eq. (5), the problem becomes unmanageable. Here, in the effective-field theory,[23] we use a decoupling procedure that ignores all high-order spin correlations on both right-hand sides of Eq. (5) (implicit way), namely

$$\langle S_{ip}^z\cdot S_{jp}^z\cdots S_{kp}^z\rangle\simeq m_p\cdot m_p\cdots m_p, \tag{8}$$

where $i\neq j\neq\cdots\neq k$ and $m_p\equiv\langle S_{ip}^z\rangle$. The approximation (8) neglects correlations between different spins but takes exactly into account self-correlations such as $\langle(S_{ip}^z)^2\rangle=1$. In the usual mean-field approximation (MFA) all the self- and multispin correlations are neglected. From this approach we get the following set of equations for the layer magnetizations in a simple cubic lattice,

$$m_0=(\alpha_{sx}+m_0\beta_{sx})^3(\alpha_x+m_1\beta_x)\times(\alpha_{sy}+m_0\beta_{sy})^3(\alpha_y+m_1\beta_y)g_0(x,y)|_{x,y=0}, \quad (9)$$

for the zeroth surface magnetization ($p=0$),

$$m_1=(\alpha_x+m_1\beta_x)^3(\alpha_x+m_0\beta_x)(\alpha_y+m_1\beta_y)^3(\alpha_y+m_0\beta_y)\times(\alpha_x+m_2\beta_x)(\alpha_y+m_2\beta_y)g_1(x,y)|_{x,y=0}, \quad (10)$$

for the first layer magnetization, and

$$m_l=(\alpha_{sx}+m_l\beta_{sx})^3(\alpha_x+m_{l-1}\beta_x)\times(\alpha_{sy}+m_l\beta_{sy})^3(\alpha_y+m_{l-1}\beta_y)g_l(x,y)|_{x,y=0}, \quad (11)$$

for the lth surface magnetization ($p=l$), where $\alpha_\nu=\cosh(K_bD_\nu)$, $\beta_\nu=\sinh(K_bD_\nu)$, $\alpha_{s\nu}=\cosh(K_sD_\nu)$, $\beta_{s\nu}=\sinh(K_sD_\nu)$, and $g_l(x,y)$ corresponds to Eqs. (6) and (7) for the quantum and classical Heisenberg models, respectively.

Equations (9)–(11) give, self-consistently, the layer magnetizations for any temperature. However, as we are interested in the calculation of the ordering near the transition, the usual argument that the layer magnetization m_p tends to zero as the temperature approaches its critical value allows us to consider only linear terms in m_p. In this case, we obtain the following set of simultaneous equations:

$$m_o=3B_sm_0+B_1m_1, \quad (12)$$

$$m_1=3B_2m_1+B_2m_0+B_2m_2, \quad (13)$$

$$\vdots$$

$$m_l=3B_sm_l+B_sm_{l-1}, \quad (14)$$

with

$$B_s=2\alpha_{sx}^3\alpha_{sy}^2\beta_{sy}\alpha_x\alpha_yg_o(x,y)|_{x,y=0}, \quad (15)$$

$$B_1=2\alpha_{sx}^3\alpha_{sy}^3\beta_y\alpha_xg(x,y)|_{x,y=0}, \quad (16)$$

and

$$B_2=2\alpha_x^4\alpha_y^4\beta_xg(x,y)|_{x,y=0}. \quad (17)$$

By performing a tedious but straightforward calculation, the coefficients B_s, B_1, and B_2 can be determined by the use of the relation $\exp(aD_x+bD_y)f(x,y)=f(x+a,y+b)$.

In a matricial notation Eqs. (12)–(14) can be rewritten in the form

$$\mathbf{A}\begin{pmatrix} m_0 \\ m_1 \\ \cdot \\ \cdot \\ m_{l-1} \\ m_l \end{pmatrix}=\mathbf{0}, \quad (18)$$

with

$$\mathbf{A}=\begin{pmatrix} a & 1 & 0 & 0 & \cdot & \cdot & \cdot \\ 1 & b & 1 & 0 & \cdot & \cdot & \cdot \\ 0 & 1 & b & 1 & 0 & \cdot & \cdot \\ \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot & \cdot & 1 & b & 1 \\ \cdot & \cdot & \cdot & \cdot & \cdot & 1 & a \end{pmatrix}_{l\times l}, \quad (19)$$

where $a=(3B_s-1)/B_1$ and $b=(3B_2-1)/B_2$. This set of equations can be satisfied by nonzero magnetizations m_p only if the determinant of the matrix $\mathbf{A}$ is zero, namely $\det\mathbf{A}=\mathbf{0}$. This condition yields l different solutions for the critical temperature. The largest solution is interpreted as the Curie temperature of the thin film.[33]

On the other hand, to obtain the bulk and surface critical temperatures of the semi-infinite ($l\to\infty$) classical and quantum Heisenberg models, we follow the same procedure of Refs. 26 and 27 to get a new set of equations given by

$$m_o=3B_sm_0+B_1m_1, \quad (20)$$

$$m_1=3B_2m_1+B_2m_0+B_2m_2, \quad (21)$$

and

$$m_p=3B_2m_p+B_2m_{p-1}+B_2m_{p+1}, \quad p\geqslant 2. \quad (22)$$

Assuming now that $m_{p+1}=\lambda m_p$ for $p\geqslant 2$,[34] we have the following secular equation:

$$\mathbf{A_s}\begin{pmatrix} m_0 \\ m_1 \end{pmatrix}=\begin{pmatrix} W_1 & 1 \\ 1 & W_2+\lambda \end{pmatrix}\begin{pmatrix} m_0 \\ m_1 \end{pmatrix}=\mathbf{0}, \quad (23)$$

where $W_1=(3B_s-1)/B_1$, $W_2=(3B_2-1)/B_2$, and $\lambda=(-W_2-\sqrt{W_2^2-4})/2$. Note that $W_2<0$, therefore $\lambda<1$, so we have an exponential decrease for the magnetization $m_{p>1}$ in the ordered phase.

III. PHASE DIAGRAMS

Let us first consider the limit $l\to\infty$. In this case the semi-infinite system exhibits three different types of phases, namely: the bulk phase (**B**) where both bulk and free surface are magnetized; the surface phase (**S**) where only the free surface is magnetized; and the paramagnetic phase (**P**) where both bulk and surface are disordered. For a sufficiently high surface coupling enhancement $\Delta>\Delta_c$ ($\Delta=J_s/J_b-1$) there exists a **S**-**P** phase transition in addition to the usual **B**-**P** phase transition, and $\Delta=\Delta_c$ corresponds to the special phase transition where **S** and **B** order simultaneously occur. The bulk transition temperature is obtained by setting $m_p=m_{p+1}=m_b$ ($l\to\infty$) in Eq. (22). In this way we have the same results previously obtained in Ref. 22. The reduced bulk transition temperature $T_c^b\equiv K_c^{-1}=5.031$ and 4.891 for the classical and quantum Heisenberg models, respectively, are comparable to $T_c^b=4.329$ (Ref. 11) and 3.35 (Ref. 35) obtained by Monte Carlo simulation (here, and in what follows, we are measuring the temperature in units of J_b/k_B,

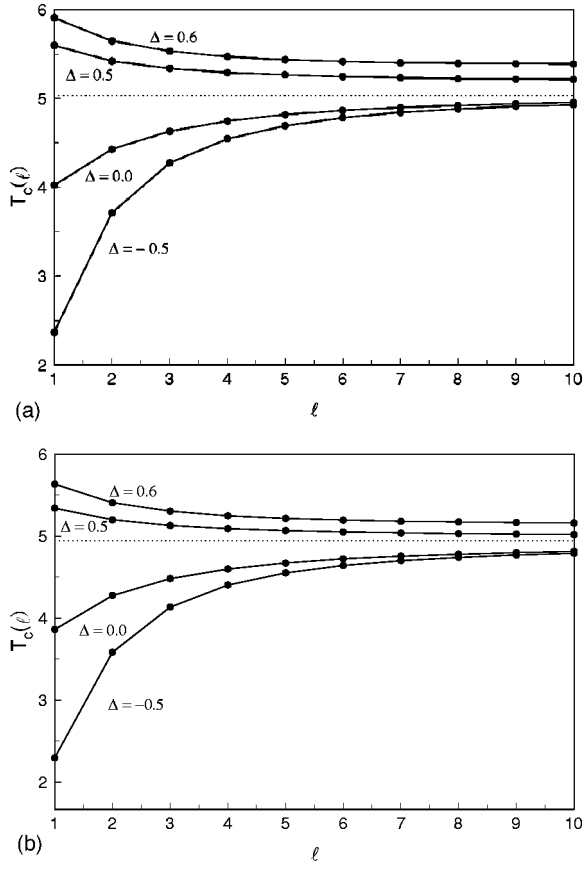

FIG. 1. Reduced critical temperature $T_c(l)$ as a function of the film thickness l for several values of the surface to bulk exchange ratio Δ. The dotted line corresponds to bulk critical temperature. (a) Classical Heisenberg, (b) quantum Heisenberg.

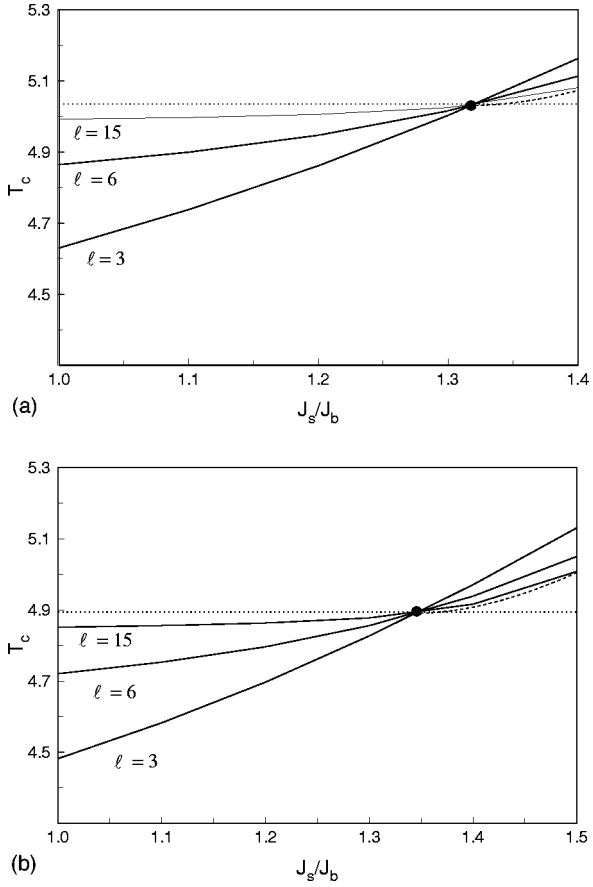

FIG. 2. Reduced critical temperature $T_c(l)$ as a function of the surface to bulk exchange ratio J_s/J_b for several values of the thickness l. The dashed and dotted lines corresponds to results in the limit $l\to\infty$ (semi-infinite system) and bulk temperature, respectively. (a) Classical Heisenberg, (b) quantum Heisenberg.

i.e., $T\equiv K^{-1}$). The present formalism also shows the correct qualitatively result $T_c^b(\text{classical})>T_c^b(\text{quantum})$. The critical frontier separating the paramagnetic-ordered surface phases is determined from the condition $\det \mathbf{A_s}=\mathbf{0}$, namely

$$W_1(W_2+\lambda)=1. \tag{24}$$

The multicritical point at Δ_c is obtained from the above equation with $T=T_c^b$ which gives $\Delta_c=0.332$ and 0.349 for the classical and quantum Heisenberg models, respectively. These values of Δ_c have not been only calculated in the literature for the case of the semi-infinite classical Heisenberg model, but also the semi-infinite quantum Heisenberg model has been recently analyzed by the present EFT method in Refs. 27 and 28.

For thin films Eq. (19) can be solved numerically in order to get the critical temperature $T_c(l)$ as a function of the thickness l of the film. Taking the limit $l\to\infty$ we find: (i) the same values of the bulk critical temperature T_c^b above reported for the classical and quantum Heisenberg models on a simple cubic lattice ($z=6$) for $\Delta<\Delta_c$ and (ii) the same surface critical temperature $T_c^s(\Delta)$ as given by Eq. (24) for $\Delta>\Delta_c$. Figure 1 shows the reduced critical temperature T_c as a function of l for different values of the surface interaction Δ in thin films with *infinite* [classical Heisenberg, Fig. 1(a)] and *finite* [quantum Heisenberg, Fig. 1(b)] spins. We can see that now the phase diagrams are quite different from the corresponding ones for the semi-infinite systems (see Refs. 26 and 27). The main difference is that in the film instead of the possibility of the existence of the critical temperatures T_c^b and T_c^s (surface critical temperature) we get only a well defined film critical temperature T_c which depends on the film thickness. The new multicritical point (Δ_c) can be defined as that particular Δ value at which the film critical temperature $T_c(l)$ does not depend on film thickness. The numerical value of Δ_c is the same as those found for the semi-infinite system. Furthermore, according to the definition of Δ_c, it can be expected that the crossover point in Fig. 1 should define also the critical temperature of the three-dimensional infinite bulk system, where the surface and the Δ parameter are of no importance. The bulk T_c^b critical temperature is represented by the dotted line. Figure 1 shows also that for $\Delta<\Delta_c$ the critical temperature $T_c(l)$ of the film is smaller than the bulk critical temperature T_c^b. It increases with the film thickness l and approaches T_c^b asymptotically as the number of layers becomes large. For $\Delta=\Delta_c$ the critical temperature $T_c(l)$ is independent of l, and equal to T_c^b. When $\Delta>\Delta_c$, the critical temperature of the film is greater than

both the bulk T_c^b and the surface T_c^s critical temperatures of the corresponding semi-infinite systems and decreases as l gets larger.

In Fig. 2 we present the dependence of the reduced critical temperature on the surface-to-bulk ratio J_s/J_b for several film thicknesses l for the classical [Fig. 2(a)] and quantum [Fig. 2(b)] Heisenberg thin films. Up to the critical value of surface exchange enhancement (black point in Fig. 2), $\Delta_c = 0.332$ and 0.349 for the semi-infinite classical and quantum Heisenberg models, respectively, the transition temperature increases with increasing the film thickness while above it, it decreases. This is ascribed to the fact that the ratio of surface spins to bulk spins decreases with increasing the film thickness and the importance of the difference in exchange interactions within the surface J_s and bulk J_b in determining the overall ordering temperature is progressively reduced as l increases. At the multicritical point $\Delta=\Delta_c$, $T_c(l)$ is independent of l and equal to T_c^b. This is really the case, which can be seen also in Fig. 2, where the bulk T_c^b and the surface T_c^s critical temperatures of the corresponding semi-infinite systems are represented by the dotted and dashed lines, respectively. In particular, the critical curve for T_c^s presented in Fig. 2 for the semi-infinite quantum Heisenberg model is the same result as that previously obtained in Refs. 26 and 27.

IV. CONCLUDING REMARKS

In this paper we have extended to the classical and quantum spin-$\frac{1}{2}$ Heisenberg thin films a type of effective-field theory previously developed to the usual Ising thin films.[7–9] We have generated a set of formal relations which are suitable to explicitly incorporate correlations through some sort of successive approximation scheme. The method is here illustrated in its simplest approximation version, in which correlations are neglected. Within this framework we discuss the temperature dependence as function of the number l of spin layers in the film. The formalism of transition temperature derivation obtained above is general and can be used in the study of thin film of various thicknesses and lattice structures. We treat in the present study the simple cubic lattice, and a critical value $\Delta_c = 1-(J_s/J_b)_c$ has been found in the phase diagram in the $(T, J_s/J_b)$ plane for various thickness, where Δ_c corresponds the common point of all the critical curves. The value of Δ_c obtained is the same as that of the corresponding semi-infinite system. The film has one critical temperature which is lower than the bulk critical temperature for $\Delta<\Delta_c$ and larger than both the T_c^b and the surface T_c^s critical temperatures for $\Delta>\Delta_c$. We observe that for $\Delta<\Delta_c$, $T_c(l)$ increases with the increasing of the film thickness l to approach T_c^b, and for $\Delta>\Delta_c$, $T_c(l)$ decreases to approach asymptotically the surface critical temperature T_c^s.

Finally, owing to its simplicity, the EFT method developed here can be used to study the thin film with other coordination number, interlayer exchange coupling dependence with respect to pairs of layers, effect of dilution, etc. Research in this direction is now in progress.

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$$H=K\sum_{\langle i,j\rangle}[(1-\Delta)(\sigma_i^x\sigma_j^x+\sigma_i^y\sigma_j^y)+\sigma_i^z\sigma_j^z],$$

where $\Delta=0$ corresponds to the isotropic Heisenberg model ($T_c=0$). In Ref. 16(a) [also in Ref. 16(c)] it has been shown that $T_c\simeq[\ln(1/\Delta)]^{-1}$ in the limit $\Delta\to 0^+$.

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