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Quantum effects in Heisenberg antiferromagnetic thin films

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Quantum effects on surface magnetizations in antiferromagnetic thin films with Heisenberg spins ($S=\frac{1}{2}$) are investigated in detail theoretically. The system studied is a film with a body-centered-cubic lattice with (001) surfaces. Using a Green's-function technique, self-consistent layer magnetizations are obtained at an arbitrary temperature for a given set of surface parameters. At high temperatures the layer magnetization is smallest at the surface and increases gradually inward. However, at low temperatures, there is an unusual behavior of the layer magnetizations near the surface: the second-layer magnetization is smaller than the surface magnetization. This anomaly has no counterpart in ferromagnetic thin films and is interpreted as an effect of the quantum fluctuations.

I. INTRODUCTION

Surface effects in magnetic materials have been the subject of intensive theoretical[1–6] and experimental[7,8] studies during the last two decades. In particular, critical behaviors at surfaces have been investigated by different methods. For a recent review, the reader is referred to an article by Binder.[1] In general, below the magnetic transition temperature, the magnetization of the surface layer can be smaller, the same, or larger than the magnetization of a layer in the bulk. On the other hand, there has been a large number of studies dealing with localized surface spin waves at zero temperature.[2] In a previous work,[9] we have calculated the effects of these surface modes on the surface magnetization and critical temperature of cubic and body-centered-cubic (bcc) ferromagnetic thin films with quantum Heisenberg spins. A recent paper by Hong[5] treated the same question for the Ising case using a mean-field approximation. We have also considered antiferromagnetic thin films.[9] However, the profile of layer magnetization as a function of temperature has not been calculated.[9]

Rapid developments in the fields of magnetic multilayers and superlattices[10] make it necessary to have a microscopic understanding of single magnetic films. With this in mind, we investigate in this paper the effect of quantum fluctuations on the layer magnetizations in the vicinity of the surface at an arbitrary temperature in a bcc antiferromagnetic thin film with (001) surfaces. It has been shown that quantum fluctuations can cause unexpected effects at low temperatures in nonhomogeneous systems, for example, in frustrated Heisenberg spin lattices[11] or antiferromagnetic superlattices.[12] As it turns out, we found in the present work that quantum fluctuations cause an unusual behavior of the profile of magnetization in the direction normal to the film surface at zero and low temperatures: There is an oscillation of layer magnetizations restricted to the first four layers. At higher temperatures this behavior disappears and one recovers the common feature near the transition temperature observed in various systems including ferromagnetic thin films.[1,9] For our purpose we use the double-time Green's-function method developed by Zubarev[13] for *quantum* Heisenberg spins of amplitude one-half. We also perform Monte Carlo (MC) simulations of the same model, but with *classical* Heisenberg spins for comparison.

In Sec. II the model and the Green's-function formalism are presented. The results are shown in Sec. III where data from MC simulations of the corresponding system with classical spins are also displayed for comparison. Concluding remarks are given in Sec. IV.

II. MODEL AND FORMALISM

We consider a bcc antiferromagnetic (AF) thin film (TF) with (001) surfaces. The Hamiltonian is given by

$$H=2\sum_{i,j} J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j+2\sum_{i,j} D_{ij}S_i^zS_j^z \ , \tag{1}$$

where J_{ij} is the exchange interaction between the quantum Heisenberg spins $\mathbf{S}_i$ and $\mathbf{S}_j$ sitting at two nearest lattice sites i and j, respectively, and D_{ij} is an Ising-type anisotropic interaction between nearest neighbors (NN). Both J_{ij} and D_{ij} are supposed to be positive (AF interactions) and allowed to be different for surface and interior spins. Since D_{ij} is positive, the spins are aligned in the z direction perpendicular to the film surfaces. This model therefore may correspond to experimental systems for magneto-optical perpendicular recording. In the following let us consider a film with an even number of layers N and the same condition at the two surfaces, for simplicity. Let J_S and D_S be, respectively, the exchange and anisotopic interactions between a surface spin and a spin belonging to the second layer. The interactions between interior *nn* spins are J and D. Note that the spins within each layer are parallel and those between two adjacent layers are antiparallel.

We define the intrasublattice and intersublattice double-time Green's functions $G_{ij}(t,t')$ and $F_{ij}(t,t')$ by[13]

$$G_{ij}(t,t')=\langle\langle S_i^+(t);S_j^-(t')\rangle\rangle \ ,$$

i and j belonging to up sublattice, and (2)

$$F_{ij}(t,t')=\langle\langle S_i^+(t);S_j^-(t')\rangle\rangle \;,$$

i and j belonging to up and down sublattices, respectively, where $S_i^+(t)$ and $S_j^-(t)$ are the usual notations defined from the x and y spin components. We write the equations of motion for these functions, for example, for $G_{ij}(t,t')$; one has

$$\frac{i\,dG_{ij}(t,t')}{dt}=\langle[S_i^+(t),S_j^-(t')]\rangle\delta(t-t') -\langle\langle[H,S_i^+(t)];S_j^-(t')\rangle\rangle \;, \tag{3}$$

where $\delta(t-t')$ is the Dirac δ function, $\langle\cdots\rangle$ means thermal average, and $\hbar=1$ has been used. The higher-order Green's functions generated by the second term of (3) are treated by the Tyablikov decoupling scheme; for example,

$$\langle\langle S_i^z S_i^+(t);S_j^-(t')\rangle\rangle\approx\langle S_i^z\rangle\langle\langle S_i^+(t);S_j^-(t')\rangle\rangle \;. \tag{4}$$

This approximation neglects some fluctuations which may affect quantitatively the results shown below. It is, however, believed that the qualitative behavior found in this paper may not be altered by such a decoupling scheme since it is well known that in the limit of vanishing temperature, the Green's-function method reproduces to the three lowest-order terms in the temperature expansion of the magnetization obtained from the spin-wave theory.

We introduce now the following two-dimensional and time Fourier transforms:

$$\begin{aligned} G_{ij}(t,t')&=(1/\pi^2)\int\int dk_x dk_y(1/2\pi)\int d\omega\, e^{-i\omega(t-t')}G_{mm'}(\omega,k_x,k_y)e^{i\mathbf{k}\cdot(\mathbf{i}-\mathbf{j})} \;,\\ F_{ij}(t,t')&=(1/\pi^2)\int\int dk_x dk_y(1/2\pi)\int d\omega\, e^{-i\omega(t-t')}F_{mm'}(\omega,k_x,k_y)e^{i\mathbf{k}\cdot(\mathbf{i}-\mathbf{j})} \;, \end{aligned} \tag{5}$$

where $\mathbf{k}$ is a wave vector in the xy plane (with component k_x and k_y), $\mathbf{i}$ and $\mathbf{j}$ are the lattice-site positions, ω is a spin-wave frequency, and m (or m') $=1,2,\ldots,N$ denotes the z components of the position, i.e., the layer ordering number beginning with one surface ($n=1$) and terminating with the other surface ($n=N$). Since N is supposed to be even, one puts $N=2L$ and one can replace the indices m and m' of G by $2n$ and by $2n'$ respectively, with $n(n')=1,2,\ldots,L$. For the intersublattice function F, one replaces the indices m by $2n-1$ and m' by $2n'$, with $n,n'=1,2,\ldots,L$. The resulting equations of motion for G and F can be written in a matrix form:

$$\Delta(E)\mathbf{g}_{2n'}=\mathbf{u}_{2n'} \;, \tag{6}$$

where

$$\Delta(E)=\begin{bmatrix} E+A_1 & \alpha B_1 & 0 & & & 0\\ -\alpha B_2 & E-A_2 & -B_2 & & & 0\\ 0 & B_3 & E+A_3 & B_3 & & 0\\ & & \cdots & & & \\ 0 & & B_{N-1} & & E+A_{N-1} & \alpha B_{N-1}\\ 0 & & & & -\alpha B_N & E-A_N \end{bmatrix} \;, \tag{7}$$

$$\mathbf{g}_{2n'}=\begin{bmatrix} f_{1,2n'}\\ g_{2,2n'}\\ f_{3,2n'}\\ \cdots\\ f_{2L-1,2n'}\\ g_{2L,2n'} \end{bmatrix} \;, \tag{8}$$

$$\mathbf{u}_{2n'}=\begin{bmatrix} 0\\ 2M_2\delta_{2,2n'}\\ 0\\ \cdots\\ 0\\ 2M_{2L}\delta_{2L,2n'} \end{bmatrix} \;, \tag{9}$$

where $\delta_{2n,2n'}$ is the Kronecker symbol, $E=\omega/J$, $g_{2n,2n'}=JG_{2n,2n'}$, $f_{2n+1,2n'}=JF_{2n+1,2n'}$, $M_{2n}=\langle S_{2n}^z\rangle$ (magnetization per spin of the $2n$th layer), $M_{2n+1}=-\langle S_{2n+1}{}^z\rangle$, and

$$\begin{aligned} A_1&=8M_2(\alpha+d_s) \;,\\ A_2&=8M_1(\alpha+d_s)+8M_3(1+d) \;,\\ A_{2n-1}&=8(M_{2n-2}+M_{2n})(1+d),\quad n=2,\ldots,L-1 \;,\\ A_{2n}&=8(M_{2n-1}+M_{2n+1})(1+d),\quad n=2,\ldots,L-1 \;,\\ A_{2L-1}&=8M_{2L}(\alpha+d_s)+8M_{2L-2}(1+d) \;,\\ A_{2L}&=8M_{2L-1}(\alpha+d_s) \;,\\ B_m&=8\gamma_{\mathbf{k}}M_m,\quad m=1,2,\ldots,2L \;,\\ \alpha&=J_s/J\\ d_s&=D_s/J \;,\\ d&=D/J \;,\\ \gamma_{\mathbf{k}}&=\cos(k_x a)\cos(k_y a) \;, \end{aligned}$$

with a being half of the lattice constant.

The spin-wave spectrum is obtained by solving the secular equation $\det\Delta(E)=0$, for a given set of α, d, and d_s, and for a given $\mathbf{k}$. Now let us denote these N solutions for a wave vector $\mathbf{k}$ by $E_{k,i}$ with $i=1,\ldots,N$. Using the spectral theorem[13] which relates the correlation function $\langle S_i^- S_j^+ \rangle$ to the Green's functions and after some algebra,[9,12] the magnetization of the even layer $2n$ at the temperature T for spin $\frac{1}{2}$ is given by

$$M_{2n}=\tfrac{1}{2}-(1/\pi^2)\times\int\int d\mathbf{k}\sum_i a_{2n}(E_{k,i})\times[\exp(E_{k,i}/T)-1]^{-1} \quad (i=1,\ldots,N) \tag{10}$$

for $n=1,\ldots,L$, where $a_{2n}(E_{k,i})=|\Delta_{2n,2n}(E_{k,i})|/\Pi_j(E_{k,i}-E_{k,j})$, with $j\neq i$, and $|\Delta_{2n,2n}(E_k)|$ is the determinant obtained by replacing the column $2n$ of $\Delta(E_k)$ by Eq. (9), with $n'=n$. One has to solve self-consistently the L coupled equations (10) to obtain the L layer magnetizations from the surface to the half of the film (the other half is symmetric due to the assumption of identical surfaces).

The layer magnetizations at $T=0$ are obtained by noticing that in the sum of (10) the contributions from positive $E_{k,i}$ are zero and the exponential terms are zero for negative $E_{k,i}$.

III. RESULTS

Before showing our results, let us mention that the calculation of the critical temperature T_c as a function of film thickness and surface anisotropy has been done in our previous work[9] using the approximation which assumes a uniform magnetization for all layers. In principle, we can use the formalism presented in Sec. II to calculate T_c taking into account the correct profile of layer magnetizations as the temperature T tends to T_c. This is done in a few examples shown below.

For numerical integrations, 20^2 points have been taken in the Brillouin zone and a precision of 0.1% is required for self-consistency at low temperatures. At higher temperatures (typically $T>1.2$), a precision of 1% is required in order to keep the number of iterations reasonable (below 10 times).

Figure 1 shows the spin-wave spectrum in the case of free surfaces; i.e., the surface parameters are the same as the bulk ones ($\alpha=1$ and $d_s=d=0.01$), for $N=4$ and 8, using the *self-consistent values* of the layer magnetizations at $T=0$. The surface spin-wave branches are denoted by S. These surface spin waves are damped from the sur-

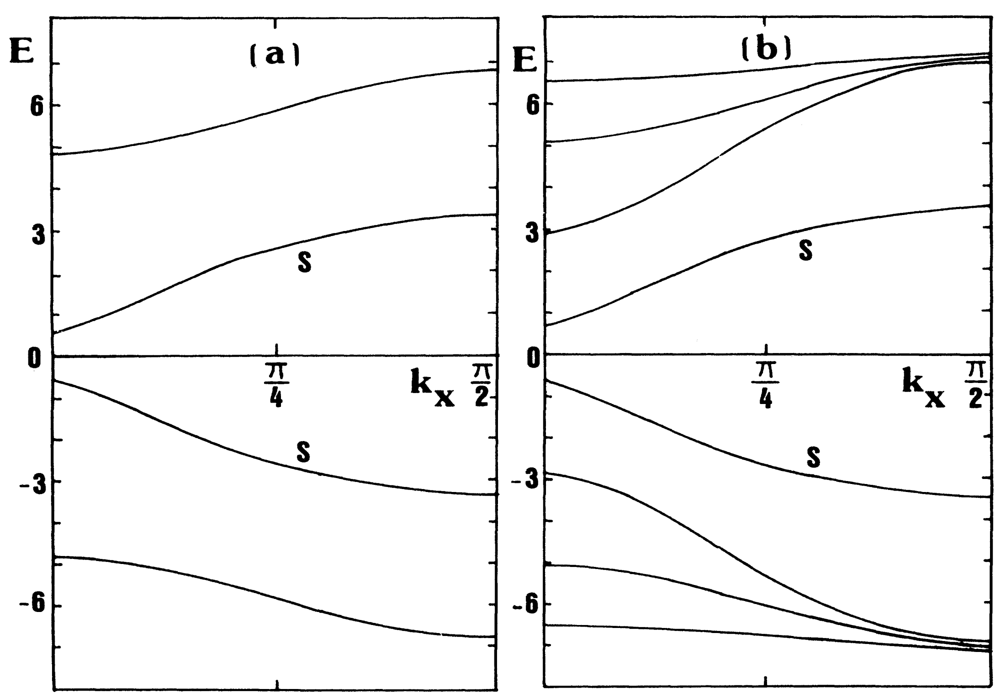

FIG. 1. Spin-wave energy E vs k_x with $k_y=0$, at $T=0$, using the self-consistent values of layer magnetizations for $\alpha=1$, $d=d_s=0.01$, and (a) $N=4$, (b) $N=8$. The surface spin-wave branches are indicated by S. E and T are in units of J.

faces inward, as can be seen from the calculation of their amplitudes, which are not shown here (see Ref. 9). It is noted that for $N>4$ there is a multidegeneracy of the bulk branches at particular points of the Brillouin zone if layer magnetizations are supposed to be uniform.[9] When self-consistent values are used, this degeneracy is removed [see Fig. 1(b) at $k_x=\pi/2$]. The removal of degeneracy yields simple poles for the Green's functions, making the calculations easier. Otherwise, one has to make calculations with higher-order poles of the Green's functions (see the discussion of this point in Ref. 9).

We show in Fig. 2 the profile of magnetization in the direction perpendicular to the film surfaces at several temperatures, for $N=4$ and 8, with free surfaces ($\alpha=1$ and $d_s=d=0.01$). As seen, at low temperatures, *the magnetization of the second layer is smaller than that of the surface.* The values of M_1^0 and M_2^0 for $N=4$ are, respectively, 0.432 and 0.410. For $N=8$, the layer magnetizations are 0.447, 0.424, 0.438, and 0.437 starting from one surface. These values do not, however, vary significantly for larger N, at least up to $N=20$. Note that the third layer has a very slightly larger magnetization than the fourth layer, which cannot be distinguished on the scale of Fig. 2. This oscillatory behavior of layer magnetizations near the surface is also found for $N>8$

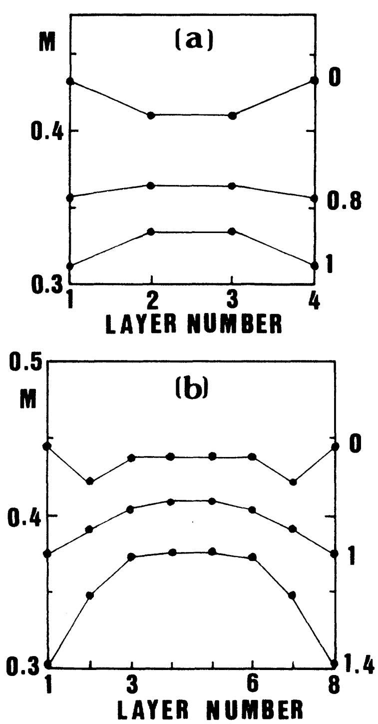

FIG. 2. Layer magnetizations across the film thickness (solid circles) at several temperatures (values indicated on the right-hand side in unit of J) in the case $\alpha=1$ and $d=d_s=0.01$, for (a) $N=4$, (b) $N=8$. Lines are guides to the eye. See text for comments.

and is always restricted to the first four layers. As T is increased, the surface magnetization becomes smaller, and the layer magnetization is increased gradually inward. The behavior at high T has been commonly found experimentally and theoretically in various systems, not restricted to antiferromagnetic thin films.[1] However, the *crossover* to the behavior at low T has never been noticed before, although unusual zero-point spin contractions for $N=4$ have been briefly reported.[9]

The layer magnetizations versus temperature T are shown in Fig. 3 in the case of free surfaces for $N=8$. The solid circles including the Néel temperature T_N are calculated (for T_N, only a precision of 5% is required). The broken lines are arbitrary interpolations.

Let us discuss the physical meaning of the unusual behavior of magnetization profile near the surfaces at low temperatures shown in Figs. 2 and 3. It is evident that this behavior comes from the quantum fluctuations resulting from *antiferromagnetic interactions near a surface.* Without surfaces, the quantum fluctuations due to antiferromagnetic interactions give a uniform zero-point spin contraction. So the presence of a surface gives rise to the spatial dependence of quantum fluctuations. At higher temperatures, the effect of quantum fluctuations is not relevant compared to that of thermal excitations. So the magnetization becomes smallest at the surface due to the lack of neighbors and is increased gradually inward, as also found commonly in various systems including ferromagnetic surfaces.[1] For the discussion below, let us call *crossover temperature* the temperature at which the surface magnetization becomes smaller than the second-layer magnetization.

We consider now the effects of surface exchange interaction α and surface anisotropy d_s on the magnetization near the surfaces. The layer magnetizations for

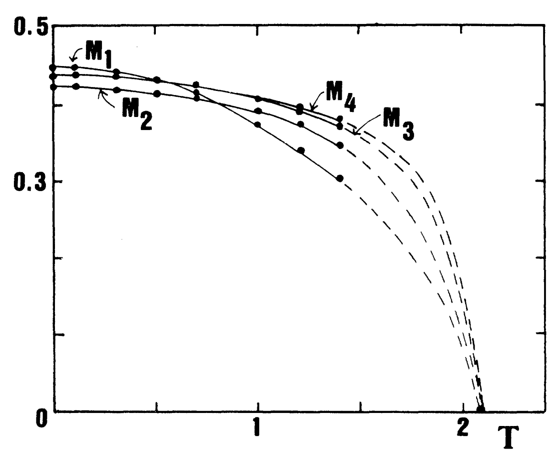

FIG. 3. Self-consistent layer magnetizations of the first four layers vs temperature T (in units of J) for $N=8$ with $\alpha=1$ and $d=d_s=0.01$. Calculated points are shown by solid circles, solid lines are guides to the eye, and dashed lines are arbitrary intrapolations. See text for comments.

TABLE I. Self-consistent layer magnetizations at $T=0$, for several couples of (α,d_s) in the case of $N=8$ and $d=0.01$. In each cell the first line shows the magnetizations of the surface and the second layer, while the second line displays the magnetizations of the third and fourth layer. See text for comments.

d_s / α	0.01		0.1		0.2	
0.5	0.466	0.434	0.476	0.446	0.482	0.454
	0.433	0.437	0.440	0.442	0.443	0.444
0.75	0.455	0.427	0.466	0.440	0.473	0.450
	0.435	0.437	0.443	0.443	0.447	0.445
1	0.447	0.424	0.458	0.435	0.465	0.444
	0.438	0.437	0.444	0.442	0.448	0.445
1.5	0.435	0.415	0.447	0.428	0.453	0.435
	0.440	0.437	0.448	0.442	0.451	0.445
2	0.425	0.409	0.437	0.421	0.445	0.430
	0.444	0.436	0.449	0.443	0.453	0.445

$\alpha=0.5$ and 1.5 are shown in Figs. 4 and 5, respectively. Above the crossover temperature, the surface magnetization is smallest even for α larger than 1, and its curvature depends strongly on α. At zero temperature, the surface magnetization, though always larger than the second-layer magnetization, is smaller than the magnetizations of the third and fourth layers for α larger than 1.

Table I shows the layer magnetizations at zero temperature for several values of α and d_s. Several remarks are in order.

(i) For a given d_s, the magnetizations of the first two layers are decreased with increasing α. This is interpreted as effect of quantum fluctuations of the Néel state: The stronger antiferromagnetic interaction yields the larger quantum fluctuations. Note the opposite tendency of the magnetizations of the third and fourth layers.

(ii) For any value of α, the surface magnetization is always larger than the second-layer magnetization.

(iii) For a given value of α, the layer magnetizations are increased with increasing d_s, contrary to the effect of α described above; thus the Ising-like surface anisotropy reduces the quantum fluctuations as it is expected.

Finally, we note that even for large surface anisotropies ($d_s=0.2$, for example), the magnetization of the surface above the crossover temperature is still smaller than that of an interior layer (not shown here), unlike the case of a simple cubic thin film where the reverse is true for large surface anisotropy (the so-called "hard surface").[9] This may be due to the fact that there is no in-plane interaction for bcc films with (001) surfaces.

In order to compare to the quantum case, we have performed MC simulations of the same system with *classical* Heisenberg spins of unit length. The method has been described in detail elsewhere.[14] We used the sample sizes of $L\times L\times N$ spins, where N is the film thickness and L the other dimensions. Periodic boundary conditions were applied in the L directions. In our simulations we used $L=20$ and $N=4$, 6, 8, 10, and 12, with 5000 MC steps

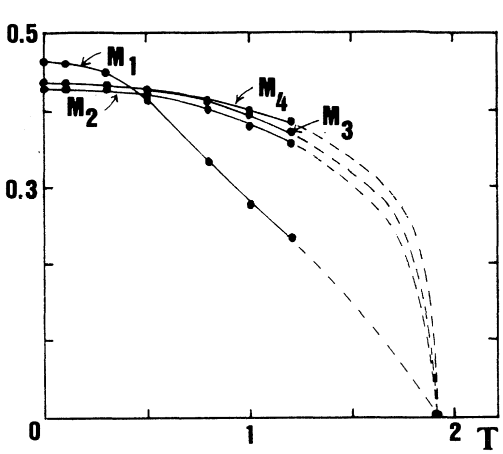

FIG. 4. Same caption as that of Fig. 3 with $\alpha=0.5$.

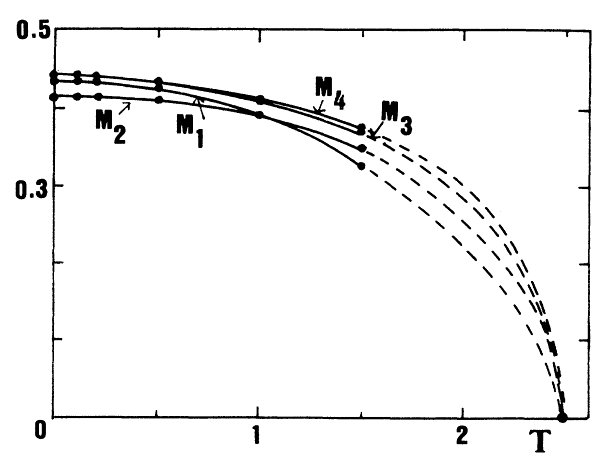

FIG. 5. Same caption as that of Fig. 3 with $\alpha=1.5$.

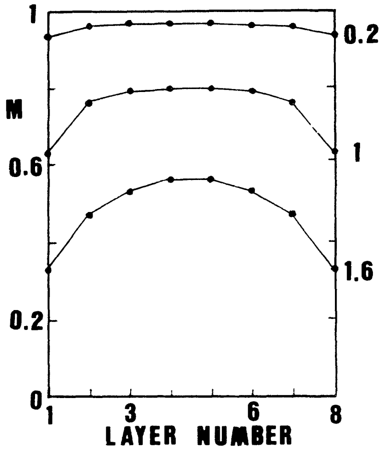

FIG. 6. Layer magnetizations across the film thickness (solid circles) at several temperatures T (values indicated on the right-hand side in units of J) obtained by Monte Carlo simulations in the case of classical Heisenberg spins of unit length, $N=8$, $d=d_s=0.01$, and $\alpha=1$. Lines are guides to the eye. See text for comments.

per spin discarded for equilibrating and 10 000 steps per spin for averaging. Figure 6 shows the profile of magnetization in the direction normal to the film surfaces at several temperatures for $N=8$, $\alpha=1$, and $d_s=d=0.01$, while Fig. 7 displays the layer magnetizations versus T for $N=8$, $d_s=d=0.01$, and $\alpha=1$ and 0.5. As seen, there is no crossover of layer magnetizations at low temperatures unlike the quantum case shown earlier. It is noted that the surface magnetization varies almost linearly with T even at very low temperatures, in agreement with the result for semi-infinite sample of simple cubic lattice with classical Heisenberg spins.[1]

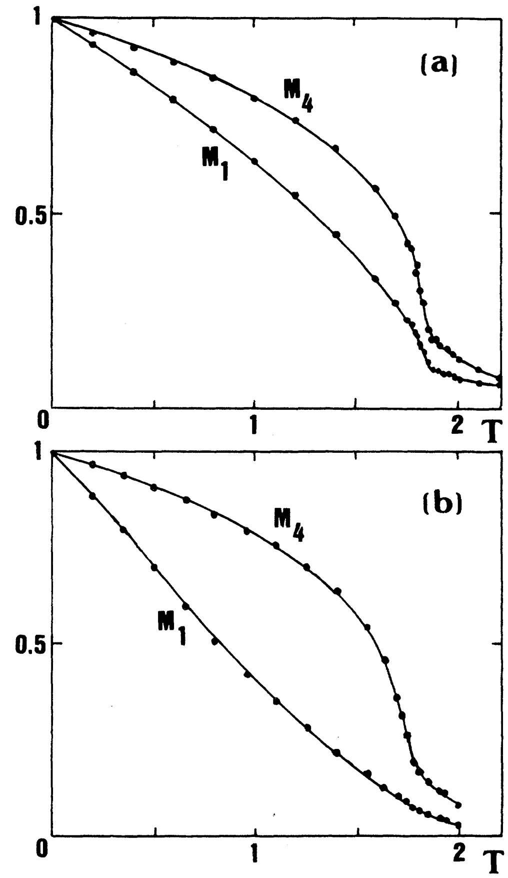

FIG. 7. Magnetizations of the first (M_1) and fourth (M_4) layers vs T (in units of J) obtained by Monte Carlo simulations in the case of classical Heisenberg spins of unit length, $N=8$, $d=d_s=0.01$, and (a) $\alpha=1$, (b) $\alpha=0.5$. For the clarity of the figure, M_2 and M_3, being between M_1 and M_4, are not shown.

IV. CONCLUDING REMARKS

In conclusion, let us emphasize that by a Green's-function method with the Tyablikov decoupling scheme we have found an unusual behavior of layer magnetizations at low temperatures in bcc antiferromagnetic thin films with NN interactions between *quantum* Heisenberg spins and (001) surfaces: There is an oscillation of the layer magnetizations in the vicinity of the surface (restricted to the first four layers) with the surface magnetization larger than the second layer one, contrary to the high-temperature behavior. This crossover between the magnetizations of the surface and the second layer has never been found before in antiferromagnetic thin films and has no counterpart in ferromagnetic thin films. Monte Carlo simulations with *classical* spins do not show this behavior at low temperatures. Therefore, we conclude that the anomaly results from the quantum fluctuations. However, the question why these fluctuations are strongest on the second layer, not on the surface, remains to be interpreted. We believe that this effect is also present in structures other than the bcc film studied here. We hope that this work will stimulate further experiments at very low temperatures on antiferromagnetic thin films.

ACKNOWLEDGMENTS

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