Magnetic properties of Heisenberg-type ferromagnetic films with a sandwich structure

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The Green's function method is developed for a sandwich structure in which a type-II ferromagnet is sandwiched between two ferromagnets of type I. The interface interactions are assumed to be ferromagnetic. Magnetization profiles and transition temperatures in such composite systems are derived for several sets of material parameters. It is shown that, if the surface anisotropy is weak, these thermodynamic quantities are affected definitely by the strong-exchange material, while if the surface anisotropy is strong, they are dependent on a competition between the effect of surfaces and the strong-exchange material. The relation to an infinite-multilayer system is also discussed.

I. INTRODUCTION

In recent years there has been considerable interest both experimentally and theoretically in the properties of magnetic superlattices formed from a superposition of layers of two different magnetic materials. Theoretically, within the Heisenberg model, most of the papers have been devoted to the study of low-temperature properties of such superlattices for a variety of artificial structures, e.g., a ferromagnet composed of two semi-infinite ferromagnets,[1,2] and a multilayer formed from alternating thin films of two ferromagnetic materials,[3,4] or of ferromagnetic and antiferromagnetic ones.[5] In ferromagnetic superlattices at low temperatures, one of the characteristic features is the existence of interface or surface magnon states appearing outside the magnon bands or in the gaps between many magnon bands of the bulk. In addition, a multilayer of alternating ferromagnetic and antiferromagnetic thin films exhibits more rich behaviors, reflecting several possible ground-state configurations under an externally applied field.[5]

Recently, Schwenk, Fishman, and Schwabl[6] have developed a phenomenological theory of ferromagnetic multilayers at finite temperatures based on a continuum model, and calculated the transition temperature and the magnetization profile of the composite systems. The more intriguing multilayer system where two different ferromagnets couple antiferromagnetically at the interfaces has been studied by Camley and Tilley[7,8] using both a mean-field approximation as well as a continuum model.

In this paper we consider an alternative artificial structure, namely, a symmetrical sandwich configuration of two different ferromagnetic thin films. The sandwich structure itself is useful for making various magnetic devices,[9] and also the double-layered structure serves experimentally as a model for investigating multilayer systems.[10] From a theoretical point of view, as will be shown in the present study, the simplicity of the structure enables us to calculate all the layer magnetizations at any temperature, and therefore, the transition temperature from a microscopic theory. These thermodynamic quantities are dependent on the strength of exchange couplings of each material, that of the interface interaction, and the thickness of each film, and are also dependent on the strength of the surface anisotropy at two surfaces. We develop a theory based on the Green's-function formalism, which is an extension of that having been successfully used to study thin films of one constituent.[11-13] The model Hamiltonian and Green's-function formalism are presented in Sec. II. The expressions for the layer magnetization and the transition temperature are derived in Sec. III. Their numerical results for some representative sets of the parameters described above are illustrated in Sec. IV. The conclusions and discussions, including a relation to a multilayer system, are also given in Sec. IV.

II. THE MODEL AND GREEN'S FUNCTION

We consider a simple cubic ferromagnet of spin-$\frac{1}{2}$ with a sandwich structure in which a type-II ferromagnet with an exchange constant J_2 and thickness N_2 is sandwiched between the same two ferromagnets of type I, each of which has an exchange constant J_1 and thickness N_1. These two constituents are coupled via a ferromagnetic exchange interaction of magnitude J' at the interfaces. A schematic view of this model is shown in Fig. 1. Different from an infinite stack of alternating thin films, a sandwich structure has two free surfaces, whose effect can be accounted for by introducing the Ising-like surface anisotropy.[11] We assume that the spins tend to be oriented parallel to the surface, say the y-z plane, and define the z and x axes as the easy and normal directions, respectively. We also assume N_2 to be even for the sake of convenience. Since all the interactions considered here are ferromagnetic, no qualitative change may arise by this assumption. This is not the case if antiferromagnetic interactions are involved.[5,7,8]

The Hamiltonian of our model may be written as

$$H=-\sum_{\langle nj,n'j'\rangle} J_{nj,n'j'}(S_{nj}^{x}S_{n'j'}^{x}+S_{nj}^{y}S_{n'j'}^{y}+\eta_{nn'}S_{nj}^{z}S_{n'j'}^{z}) , \tag{2.1}$$

where $J_{nj,n'j'}$ represents an exchange integral between nearest neighbors, taking either J_1, J_2, or J' depending

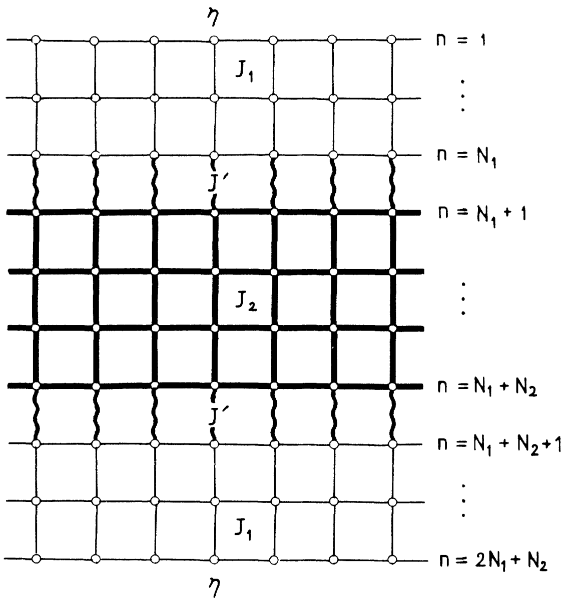

FIG. 1. Schematic view of the sandwich structure studied in this paper.

on the location of the nearest-neighbor pairs, the index n labels a layer as denoted in Fig. 1, and j the lattice point in the y-z plane. The Ising-like anisotropy $\eta_{nn'}$ in Eq. (2.1) describes the surface anisotropy such that $\eta_{nn'}=\eta\ (>1)$ only if n and n' are on the same surface layer, and $\eta_{nn'}=1$ otherwise.

We now employ the double-time temperature-dependent Green's function[14]

$$G_{n,j,n'j'}(t-t')=\langle\langle S^+_{nj}(t);S^-_{n'j'}(t')\rangle\rangle , \tag{2.2}$$

then write the equations of motion within the random-phase approximation,[11,15]

$$\langle\langle S^z_{nj}S^+_{n''j''};S^-_{n'j'}\rangle\rangle \simeq \mu_n\langle\langle S^+_{n''j''};S^-_{n'j'}\rangle\rangle , \tag{2.3}$$

where μ_n is the layer magnetization of the nth layer defined by

$$\mu_n=\langle S^z_{nj}\rangle . \tag{2.4}$$

By taking the Fourier transform with respect to time, then utilizing the translational invariance in the y-z plane, we get the equations of motion for the Green's functions $G_{nn'}(\omega,\mathbf{k})$, $\mathbf{k}=(k_y,k_z)$, in matrix form:

$$\begin{pmatrix} \omega-W_1 & m'_1 & & & \\ m_2 & \omega-W_2 & m'_2 & & \\ & \ddots & \ddots & \ddots & \\ & m_{N-1} & \omega-W_{N-1} & m'_{N-1} \\ & & m_N & \omega-W_N \end{pmatrix}\begin{pmatrix} G_{1n} \\ G_{2n} \\ \vdots \\ G_{N-1,n} \\ G_{Nn}\end{pmatrix}=\frac{1}{\pi}\begin{pmatrix}\mu_1\delta_{1n} \\ \mu_2\delta_{2n} \\ \vdots \\ \mu_{N-1}\delta_{N-1,n} \\ \mu_N\delta_{Nn}\end{pmatrix} , \tag{2.5}$$

where

$$W_n=\begin{cases} zJ_1\mu_1(\eta-\gamma_k)+J_1\mu_2, & n=1 \\ zJ_1\mu_n(1-\gamma_k)+J_1(\mu_{n-1}+\mu_{n+1}), & 2\le n\le N_1-1 \\ zJ_1\mu_{N_1}(1-\gamma_k)+J_1\mu_{N_1-1}+J'\mu_{N_1+1}, & n=N_1 \\ zJ_2\mu_{N_1+1}(1-\gamma_k)+J'\mu_{N_1}+J_2\mu_{N_1+2}, & n=N_1+1 \\ zJ_2\mu_n(1-\gamma_k)+J_2(\mu_{n-1}+\mu_{n+1}), & N_1+2\le n\le N/2 \end{cases} \tag{2.6}$$

with $z=4$ and

$$\gamma_k=\tfrac{1}{2}(\cos k_y+\cos_z) . \tag{2.7}$$

Due to the symmetrical structure of the sandwich configuration, the other half of the elements of W_n are given by

$$W_{N+1-n}=W_n,\quad n=1,2,\ldots,N/2 , \tag{2.8}$$

where $N(=2N_1+N_2)$ is the total number of layers. The same symmetrical relation as Eq. (2.8) holds for other quantities, and hereafter, unless otherwise noted, we shall not write such relations. The off-diagonal elements m_n and m'_n in Eq. (2.5) are given by

$$m_n=\begin{cases} J_1\mu_n, & n=2,\ldots,N_1 \\ J'\mu_{N_1+1}, & n=N_1+1 \\ J_2\mu_n, & n=N_1+2,\ldots,N_1+N_2 \\ J'\mu_{N_1+N_2+1}, & n=N_1+N_2+1 \\ J_1\mu_n, & n=N_1+N_2+2,\ldots,N \end{cases} \tag{2.9}$$

and

$$m'_n=m_{N+1-n},\quad n=1,\ldots,N-1 . \tag{2.10}$$

In solving Eq. (2.5) to obtain the Green's functions, it is advantageous to use the tridiagonal form of the coefficient matrix in Eq. (2.5). Following Selzer and Majlis,[16] we define a subdeterminant such that $D_{N-p}(\omega)$

means the $N-p$-th order determinant, obtained by deleting the first p rows and columns in the coefficient matrix in Eq. (2.5). We then have the recurrence relation

$$D_{N-p}(\omega)=(\omega-W_{p+1})D_{N-1-p}(\omega)-m_{p+2}m'_{p+1}D_{N-2-p}(\omega)\ ,\quad p=0,1,\ldots,N-2 \tag{2.11}$$

with

$$D_1(\omega)=\omega-W_N,\quad D_0(\omega)=1\ . \tag{2.12}$$

Due to the symmetry, $D_{N-p}(\omega)$ is equivalent to the subdeterminant which is obtained by deleting the last p rows and columns in the coefficient matrix in Eq. (2.5). Using th expression $D_{N-p}(\omega)$ thus defined, we get

$$G_{nn}(\omega,\mathbf{k})=\frac{\mu_n D_{N-n}(\omega)D_{n-1}(\omega)}{\pi D_N(\omega)},\quad n=1,\ldots,N/2\ . \tag{2.13}$$

This expression for the Green's functions, combined with the recurrence relations (2.11) and (2.12), will be found to be useful for calculating layer magnetizations in the following sections.

III. LAYER MAGNETIZATION AND TRANSITION TEMPERATURE

In this section, we derive the expressions for the layer magnetization and the transition temperature. With the aid of the spectral theorem for the Green's functions

$$\langle S_{nj}^- S_{nj}^+\rangle=\lim_{\epsilon\to 0}\frac{1}{\pi^2}\int_0^\pi\int d\mathbf{k}\, i\int_{-\infty}^{\infty}d\omega[\exp(\beta\omega)-1]^{-1}[G_{nn}(\omega+i\epsilon,\mathbf{k})-G_{nn}(\omega-i\epsilon,\mathbf{k})] \tag{3.1}$$

and the relation

$$\frac{1}{\omega-i\epsilon}-\frac{1}{\omega+i\epsilon}=2\pi i\delta(\omega)\ , \tag{3.2}$$

we have

$$\langle S_{nj}^- S_{nj}^+\rangle=2\mu_n\Phi_n\ , \tag{3.3}$$

where

$$\Phi_n=\frac{1}{\pi^2}\int_0^\pi\int d\mathbf{k}\sum_{i=1}^{N}\frac{D_{N-n}(\omega_i)D_{n-1}(\omega_i)}{[\exp(\beta\omega_i)-1]D'_N(\omega_i)} \tag{3.4}$$

with $\beta=1/k_BT$, k_B the Boltzmann constant, T the absolute temperature, and

$$D'_N(\omega)=\frac{d}{d\omega}D_N(\omega)=2\sum_{n=1}^{N/2}D_{N-n}(\omega)D_{n-1}(\omega)\ . \tag{3.5}$$

In Eq. (3.4) ω_i means the N solutions of $D_N(\omega)=0$, which can be factorized into two polynomials of $N/2$ degree,

$$D_{N/2}(\omega)\pm m_{(N/2)+1}D_{(N/2)-1}(\omega)=0\ . \tag{3.6}$$

These equations can be solved by using the recurrence relations (2.11) and (2.12) to calculate the coefficients of the polynomials.

For the case $S=\frac{1}{2}$, assumed in the present study, it follows that

$$\langle S_{nj}^- S_{nj}^+\rangle=\tfrac{1}{2}-\mu_n\ , \tag{3.7}$$

so that equating this with Eq. (3.3) we get

$$\mu_n=\frac{1}{2(1+2\Phi_n)},\quad n=1,2,\ldots,N/2\ , \tag{3.8}$$

which forms a set of self-consistent equations for the layer magnetization μ_n.

The transition temperature T_c of the whole system is determined from requiring that when approaching T_c from below all the μ_n's vanish. In a composite system, however, it occurs that some μ_n's become significantly small before reaching T_c, whereas others are still finite. Similar situations may arise even in a single film of one constituent, as was pointed out by Diep *et al.*[12] Therefore, it is convenient to scale all the quantities in terms of μ_L, which does not vanish even in the immediate vicinity of T_c. In the present system it is appropriate to choose $L=1$ when $J_1>J_2$ and $L=N/2$ when $J_1<J_2$. By using

$$\exp(\beta\omega_i)-1\simeq\mu_L\beta_c\tilde{\omega}_i \tag{3.9}$$

as $T\to T_c$ ($\beta\to\beta_c$), Φ_n defined in Eq. (3.4) can be approximated as

$$\Phi_n\simeq\frac{1}{\mu_L\beta_c}\tilde{\Phi}_n\ , \tag{3.10}$$

where the overtilde designates a scaled quantity in terms of μ_L and

$$\tilde{\Phi}_n=\frac{1}{\pi^2}\int_0^\pi\int d\mathbf{k}\sum_{i=1}^{N}\frac{\tilde{D}_{N-n}(\tilde{\omega}_i)\tilde{D}_{n-1}(\tilde{\omega}_i)}{\tilde{\omega}_i\tilde{D}'_N(\tilde{\omega}_i)}\ . \tag{3.11}$$

Then, from Eq. (3.8) we get the expression to determine T_c:

$$\beta_c=4\tilde{\Phi}_L\ ,\qquad \tilde{\mu}_n=\frac{\tilde{\Phi}_L}{\tilde{\Phi}_n},\quad n=1,\ldots,L-1,L+1,\ldots,N/2\ . \tag{3.12}$$

IV. NUMERICAL RESULTS AND DISCUSSION

Solving the self-consistent equations (3.8) and (3.12) by performing numerical integrations, we can estimate the magnetization μ_n of each layer and the transition temperature T_c of the whole system. Of special interest is how film 1 with exchange constant J_1 and film 2 with exchange constant J_2 interact through the interface interaction of magnitude J'. In addition, two free surfaces, whose effect is taken into account by the Ising-like anisotropy η, would significantly influence the values of μ_n and T_c. A variety of combinations of these parameters, together with the thicknesses N_1 and N_2, are possible. In order to understand the basic features of the magnetization profiles in sandwich structures, we select several representative sets. In the following the number of layers in each film is fixed such that $N_1=9$ and $N_2=6$, for a total of $N=24$ layers. The layer indices from 1 to 9 denote the layers in film 1, those from 10 to 12 denote the layers in film 2, and those of more than 12 are symmetric about the middle.

We first consider the case with $J_1>J_2$. The system may exhibit the properties of magnetic double layers of film 1 separated by a film 2 of weak exchange couplings. Since film 2 contributes less, the properties of film 1 will be reflected on the whole system. In Fig. 2 magnetization profiles at various temperatures are shown for the case with (a) strong surface anisotropy, $\eta=1.5$, and (b) weak surface anisotropy, $\eta=1.02$. The parameters have the values $J_1=2$, $J_2=1$, and $J'=1.5=(J_1+J_2)/2$. We see in Fig. 2(a) that larger magnetization appears on the surface layers and a few inner layers subsequent to each surface layer, which we call surface regions in the following. As the temperature increases near T_c only the surface regions remain as having finite magnetization, whereas other inner layers become almost paramagnetic. Thus, T_c in the present case corresponds to the temperature at which the magnetization of the surface regions vanish, and is estimated to be 2.723. This T_c is much higher than the bulk transition temperature $T_{c1}(\infty)=1.978$ of film 1 due to strong surface anisotropy,[12,13] and remains constant with a change in N_1 because the surface region is localized only less than a few layers from the surface.

When η is weak, as seen in Fig. 2(b), inner layers of film 1 have larger magnetization than the surfaces,[12,13] so that surface effect is less important. T_c is then determined mainly by the inner layers, and therefore, in contrast to the case with strong surface anisotropy, it is sensitive to the number of layers. We have $T_c=1.808$ slightly lower than $T_{c1}(\infty)$. As will be shown in Fig. 4, T_c in the present case increases (decreases) as N_1 increases (decreases), approaching the bulk transition temperature $T_{c1}(\infty)$.[12]

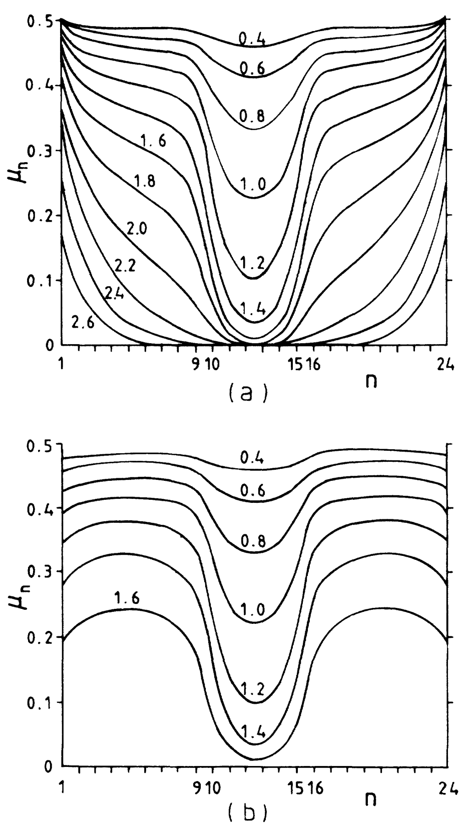

FIG. 2. Magnetization profiles for various temperatures attached to each curve. (a) The case with strong surface anisotropy $\eta=1.5$, and (b) the case with weak surface anisotropy $\eta=1.02$. The exchange constants have the values $J_1=2$, $J_2=1$, and $J'=1.5$.

Next, we consider the opposite case with $J_1<J_2$. When η is weak, film 2 of strong exchange couplings would largely contribute to the system. However, if η is strong, the transition temperature and magnetization profiles would depend on a competition between the surface regions and film 2. In Fig. 3 an example is shown for (a) the temperature dependence of the layer magnetization and the average magnetization of all layers (dotted line), and (b) the magnetization profiles at various temperatures. The parameters have the values $J_1=1$, $J_2=3$, $J'=2=(J_1+J_2)/2$, and $\eta=1.5$. To avoid errors that will appear for this set of parameters due to the numerical integrations in Eqs. (3.4) and (3.11), we have assumed a weak in-plane Ising-like anisotropy $\eta_b=1.005$ in the inner layers. Then, $1-\gamma_k$ in Eq. (2.6) is replaced by $\eta_b-\gamma_k$. We observe in Fig. 3(b) that larger magnetization appears in the surface regions of film 1 as well as the layers in film 2. As seen from Fig. 3(a), curve 1 representing the surface layer drops rapidly toward $T_{c1}=1.363$, which is the transition temperature of film 1 obtained by putting $J'=0$. At higher temperatures curves 10, 11, and 12, representing the layers in film 2, dominate and they drop rapidly to $T_c=2.731$. Therefore, in the present case there are two types of regions which can undergo phase transitions, i.e., the surface regions and film 2. The transition temperature of the whole system is determined by which region has a higher transition temperature. As can be seen from Fig. 3(a), the layer magnetizations μ_9, μ_8, and μ_7 of film 1 remain as having finite values as $T\to T_c$, indicating the strong correlation between film 2 and the successive layers 9, 8, and 7 in film 1 via the interface interaction. Since a surface region is also restricted to within about three layers, an interference between the surface regions and film 2 would occur only if film 1 is sufficiently thin.

Finally we want to relate our study to an infinite ferromagnetic multilayer system, which has been analyzed by Schwenk *et al.*[6] using a macroscopic Ginzburg-

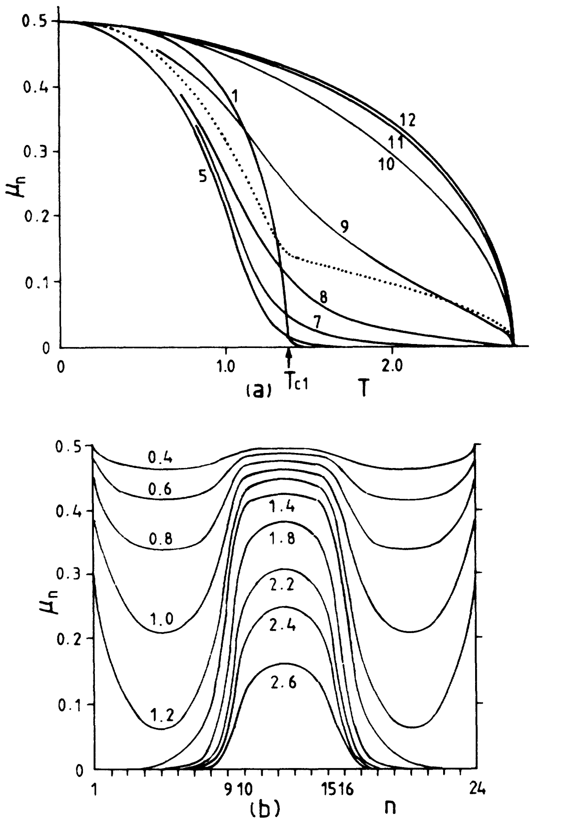

FIG. 3. (a) Temperature dependence of the layer magnetization. The number attached to each curve means the layer index as defined in Fig. 1. The dotted line represents the average magnetization. (b) Magnetization profiles for various temperatures attached to each curve. Parameters have the values $J_1=1$, $J_2=3$, $J'=2$, and $\eta=1.5$ (strong surface anisotropy).

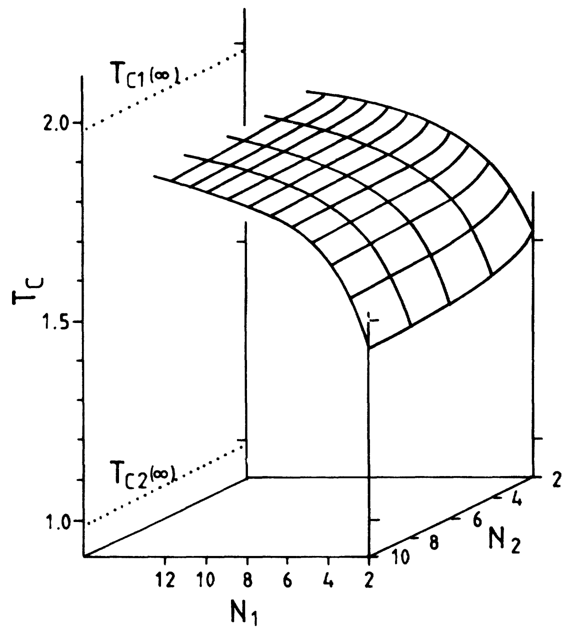

FIG. 4. Variation of the transition temperature T_c with respect to N_1 and N_2 for the same case as in Fig. 2(b). $T_{c1}(\infty)$ and $T_{c2}(\infty)$ mean the bulk transition temperatures of material 1 and material 2, respectively.

Landau theory. To our knowledge at present a corresponding study using a Green's-function theory has not been developed.[17] A similar approach to ours would bring the coefficient matrix in Eq. (2.5) to nontridiagonal form due to an infinite repetition of two kinds of thin films. We can, however, simulate the infinite multilayer system by the double-layered structure illustrated in Fig. 2(b). The variation of the transition temperature T_c with respect to N_1 and N_2 is plotted in Fig. 4. We see that T_c increases as N_1 increases, approaching the bulk transition temperature $T_{c1}(\infty)=1.978$ of material 1. As for the variation with respect to N_2, we find that only the T_c curve corresponding to $N_2=2$ deviates appreciably from the others. This means that the interaction between strong-exchange materials via a weak-exchange one has an effect on T_c only when the weak-exchange material is sufficiently thin. These results are qualitatively similar to the ones obtained by Schwenk *et al.*[6] using a continuum model.

In this paper we have developed a microscopic theory of ferromagnetic films with a sandwich structure based on the Green's-function formalism, and examined magnetization profiles and transition temperatures for several sets of material parameters. We have seen that if the surface anisotropy is weak, the properties of the system are determined mainly by the strong-exchange material, while if the surface anisotropy is strong, they are dependent on a competition between the effect of surfaces and the strong-exchange material. Therefore the magnitude of surface anisotropy is important in determining the magnetic properties of sandwiches. Hence, if the interface introduces an anisotropy effect like the surfaces, the transition temperature and the magnetization profile would change significantly, but the feature may be analogized from the present study.

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