Quantum approach for magnetic multilayers at finite temperatures

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The Green's function technique has been employed to examine the finite-temperature properties of a magnetic multilayer both in aligned and in canted spin configurations. A local coordinates system is introduced to describe complicated spin configurations which are determined by minimizing the free energy of the magnetic multilayer. Thermal averages of $S_m^{z_m}$ in each layer are selected to be independent variables, and the necessarily self-consistent equations are obtained successfully based on the random phase approximation. The temperature dependences of the coercivity, the hysteresis, and the magnetic multivalued recording have been discussed, and an application of the present approach to the canted spin state is given in a double-film system. [S0163-1829(98)02413-8]

I. INTRODUCTION

Magnetic multilayers received more and more attention in recent years because they showed high application potentials in magnetic devices and strong theoretical interests.[1] Many exciting phenomena, such as the giant magnetoresistance effect,[2] the giant magneto-impedance effect,[3] the magneto-optical recording,[4] the spin reorientation phase transition,[5] etc., were discovered based on the magnetic orders in quasi-two-dimensional systems at finite temperatures (even near the Curie temperature). Recently, many efforts were devoted to complicated coupled magnetic multilayers to improve the field sensitivity of the reading device,[6] to enhance the memory densities of the recording media,[7] and so on. Theoretically, a basic model which considers the exchange energy, the uniaxial anisotropy, and the Zeeman energy has been popularly adopted to discuss various properties of the magnetic multilayers.[8–16] Recently, a quantum theory based on this model was established to calculate the hysteresis loop and the coercivity of a magnetic multilayer,[14] and was applied to give a basic consideration of the magnetic multivalued (MMV) recording in double-film structures[15] and in magnetic granular film.[16] Some authors have tried to extend their methods to finite temperature, for example, using the molecular-field approximation which is good for the high-temperature case,[9] naively applying the Bose-Einstein statistics which is better for the very-low-temperature case[11] or some other assumptions.[12] On the other hand, some authors were devoted to making the model more realistic by including the long-ranged dipolar interactions in their Hamiltonian.[17]

The Green's function (GF) technique seems to be helpful to discuss a magnetic multilayer within the whole temperature region. The GF method was introduced into magnetic systems in 1959 by the pioneer work of Bogolyubov and Tyablibov who studied the thermodynamic properties of the spin-$\frac{1}{2}$ ferromagnetic systems.[18] In 1962, Tahir-Kheli and ter Haar extended that technique to arbitrary spin cases successfully.[19] Since then, many authors have used this approach to discuss various kinds of magnetic systems.[20,21] The most remarkable merit of the GF method is its approximate validity within the entire temperature region, which the other approaches such as spin-wave theory, molecular-field theory, and high-temperature expansion theory[22] did not possess.

In this paper, we would like to discuss the temperature dependences of several interesting properties in magnetic multilayers with the help of the GF technique based on the simple model.[8–14] Since the magnetic easy axes may be different from layer to layer in an arbitrarily layered system, a local coordinate system will be introduced into the system to optimize the spontaneous magnetized directions of each layer which are determined by minimizing the free energy. The temperature dependences of the coercivity and different thermal effects to the magnetic multivalued recording are discussed. Finally, an application to the canted spin configuration is presented.

II. FORMULAS OF THE GREEN'S FUNCTION APPROACH

For a general L-layer magnetic structure, a simple model Hamiltonian can be given as

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$$H=-\frac{1}{2}\sum_{m,m'}\sum_{\mathbf{r},\mathbf{r}'} I_{m,m'}(\mathbf{r},\mathbf{r}')\mathbf{S}_m(\mathbf{r})\cdot\mathbf{S}_{m'}(\mathbf{r}')-\mathbf{h}\cdot\sum_{m,\mathbf{r}}\mathbf{S}_m(\mathbf{r})$$
$$-\sum_m\sum_{\mathbf{r}} D_m[S_m^{z_m^0}(\mathbf{r})]^2, \tag{1}$$

where $I_{m,m'}(\mathbf{r},\mathbf{r}')$ is the exchange interaction between the spins. z_m^0 are the magnetic easy axes for each layer and they need not be the same in general. For simplicity, let us assume that z_m^0 axes in each layer are in the x-z plane, and use $\{\phi_m\}$ to denote the angle between the z_m^0 and z axes: $\hat{z}_m^0\cdot\hat{z}=\cos\phi_m$. $\mathbf{h}$ is the applied field. In this paper, we only study the case that the external field is applied perpendicularly. It is helpful to introduce the local coordinate (LC) transformation[13] $S_m^z(\mathbf{r})=\cos\theta_m S_m^{z_m}(\mathbf{r})+\sin\theta_m S_m^{x_m}(\mathbf{r})$, $S_m^x(\mathbf{r})=\cos\theta_m S_m^{x_m}(\mathbf{r})-\sin\theta_m S_m^{z_m}(\mathbf{r})$, where $\{\theta_m\}$ denote the spontaneous magnetized direction which are different from layer to layer and should be determined later by minimizing the free energy. After the LC transformation, the Hamiltonian can be divided into the following three parts:

$$H=H_1+H_2+H_3, \tag{2}$$

where

$$H_1=-\frac{1}{2}\sum_m\sum_{\mathbf{r},\mathbf{r}'} I_{m,m}(\mathbf{r},\mathbf{r}')\mathbf{S}_m(\mathbf{r})\cdot\mathbf{S}_m(\mathbf{r}'), \tag{3}$$

$$H_2=-\frac{1}{2}\sum_{m,m'}\sum_{\mathbf{r},\mathbf{r}} I_{m,m'}(\mathbf{r},\mathbf{r})\{\cos(\theta_m-\theta_{m'})[S_m^{z_m}(\mathbf{r})S_{m'}^{z_{m'}}(\mathbf{r})$$
$$+S_m^{x_m}(\mathbf{r})S_{m'}^{x_{m'}}(\mathbf{r})]+S_m^{y_m}(\mathbf{r})S_{m'}^{y_{m'}}(\mathbf{r})\}-h\sum_{m,\mathbf{r}}\cos\theta_m S_m^{z_m}(\mathbf{r})$$
$$-\sum_{m,\mathbf{r}} D_m\{\cos^2(\theta_m-\phi_m)[S_m^{z_m}(\mathbf{r})]^2$$
$$+\sin^2(\theta_m-\phi_m)[S_m^{x_m}(\mathbf{r})]^2\}, \tag{4}$$

$$H_3=-\frac{1}{2}\sum_{m,m'}\sum_{\mathbf{r},\mathbf{r}'} I_{m,m'}(\mathbf{r},\mathbf{r})\sin(\theta_m-\theta_{m'})[S_m^{x_m}(\mathbf{r})S_{m'}^{z_{m'}}(\mathbf{r})$$
$$-S_m^{z_m}(\mathbf{r})S_{m'}^{x_{m'}}(\mathbf{r})]-\frac{1}{2}\sum_{m,\mathbf{r}} D_m\sin[2(\theta_m-\phi_m)]$$
$$\times[S_m^{x_m}(\mathbf{r})S_m^{z_m}(\mathbf{r})+S_m^{z_m}(\mathbf{r})S_m^{x_m}(\mathbf{r})]-h\sum_{m,\mathbf{r}}\sin\theta_m S_m^{x_m}(\mathbf{r}). \tag{5}$$

The equations of motion can be obtained with the help of the spin operator commutations. After that, the following random phase approximation[18,19] (RPA) can be applied:

$$S_m^{\pm}(\mathbf{r},t)S_{m'}^{z_{m'}}(\mathbf{r}',t)\simeq S_m^{\pm}(\mathbf{r},t)\langle S_{m'}^{z_{m'}}\rangle,\quad m\neq m'\quad\text{or}\quad \mathbf{r}\neq\mathbf{r}', \tag{6}$$

where the notation $\langle\cdots\rangle$ means thermal averages and is defined by $(1/Z_p)\Sigma_n\langle n|e^{-\beta H}\cdots|n\rangle$ in which $Z_p=\Sigma_n\langle n|e^{-\beta H}|n\rangle$ is the partition function.

For the on-site interactions to which the above ordinary RPA cannot be applied, we adopt the decoupling scheme of Lines[20] as follows

$$S_m^{\pm}(\mathbf{r},t)S_m^{z_m}(\mathbf{r},t)+S_m^{z_m}(\mathbf{r},t)S_m^{\pm}(\mathbf{r},t)\simeq\Psi_m\langle S_m^{z_m}\rangle S_m^{\pm}(\mathbf{r},t), \tag{7}$$

where

$$\Psi_m=\frac{3\langle(S_m^{z_m})^2\rangle-S_m(S_m+1)}{(\langle S_m^{z_m}\rangle)^2}. \tag{8}$$

Abbreviations are used as follows:

$$\overline{S_m}=\langle S_m^{z_m}(\mathbf{r})\rangle,\quad \overline{S_m^2}=\langle(S_m^{z_m}(\mathbf{r}))^2\rangle. \tag{9}$$

Variational parameters $\{\theta_m\}$ should be determined by minimizing the free energy,

$$\frac{\partial}{\partial\theta_m}f=\frac{\partial}{\partial\theta_m}[-KT\cdot\ln(Z_p)]$$
$$\simeq\frac{1}{Z_p}\sum_n\left\langle n\left|e^{-\beta H}\frac{\partial H}{\partial\theta_m}\right|n\right\rangle$$
$$=\left\langle\frac{\partial}{\partial\theta_m}H\right\rangle=0, \tag{10}$$

which, based on the RPA, takes the following form:

$$-\sum_{m'} I_{m,m'}\sin(\theta_m-\theta_{m'})\overline{S_m}\,\overline{S_{m'}}-h\sin\theta_m\overline{S_m}$$
$$-\frac{1}{2}D_m\sin[2(\theta_m-\phi_m)][\overline{S_m^2}-\langle(S_m^x)^2\rangle]=0. \tag{11}$$

It is interesting to note that the above condition is the same as $[S_m^+,H_3]=0$ on the basis of the RPA, which implies that the excitations are stable. It should be noted that Eq. (10) is correct only if $\partial H/\partial\theta_m$ commute with H. We show in the Appendix that this is true based on the RPA.

With the help of the commutators and a usual Fourier transformation, we arrive at

$$[\tilde{S}_n^+(\mathbf{k}),H]\simeq\sum_m F_{n,m}(\theta_m,\mathbf{k})\tilde{S}_m^+(\mathbf{k})+\sum_m G_{n,m}(\theta_m,\mathbf{k})$$
$$\times\tilde{S}_m^-(-\mathbf{k}), \tag{12}$$

where $F_{n,m}$ and $G_{n,m}$ are some functions of θ_m, $\mathbf{k}$ and can be calculated straightforwardly, and $\tilde{S}_m^{\pm}$ is defined by $\tilde{S}_m^{\pm}=S_m^{\pm}/\sqrt{2\overline{S_m}}$.

Introducing a Bogolyubov type $(\hat{U},\hat{V})$ transformation to Eq. (12), it can be proved that if $\{\hat{U},\pm i\hat{V}\}^T$ are selected as the eigenvectors of the matrix

$$\mathcal{H}(\mathbf{k})=\begin{pmatrix}\mathcal{F}(\mathbf{k}) & i\mathcal{G}(\mathbf{k})\\ i\mathcal{G}(\mathbf{k}) & -\mathcal{F}(\mathbf{k})\end{pmatrix}, \tag{13}$$

where $\mathcal{F}(\mathbf{k})$ and $\mathcal{G}(\mathbf{k})$ in the matrix $\mathcal{H}(\mathbf{k})$ are two $L\times L$ matrices whose elements are defined by $F_{m,m'}(\mathbf{k},\theta)$ and $G_{m,m'}(\mathbf{k},\theta)$, we have

$$[A_m^+(\mathbf{k}),H]=E_m(\mathbf{k})A_m^+(\mathbf{k}),$$

$$[A_m^-(-\mathbf{k}),H]=-E_m(\mathbf{k})A_m^-(-\mathbf{k}), \tag{14}$$

where $\{E_m(\mathbf{k}),-E_m(\mathbf{k})\}$ are corresponding eigenvalues of the matrix $\mathcal{H}(\mathbf{k})$ (see Ref. 14 for a comparison).

According to Refs. 18 and 19, after a careful calculation, one can get similar self-consistent equations as the bulk spin system,

$$\overline{S_m}=\frac{[S_m-\Phi_m][1+\Phi_m]^{2S_m+1}+[S_m+1+\Phi_m][\Phi_m]^{2S_m+1}}{[1+\Phi_m]^{2S_m+1}-[\Phi_m]^{2S_m+1}}, \tag{15}$$

$$\overline{S_m^2}=S_m(S_m+1)-[1+2\Phi_m]\overline{S_m}, \tag{16}$$

except for different forms of Φ_m:

$$\Phi_m=\frac{1}{N}\sum_{l,\mathbf{k}}\frac{|U_{m,l}|^2+|V_{m,l}|^2e^{\beta E_l(\mathbf{k})}}{e^{\beta E_l(\mathbf{k})}-1}. \tag{17}$$

Therefore, we have $3L$ parameters $\{\theta_m,\overline{S_m},\overline{S_m^2};\ m=1,2,\ldots,L\}$, and $3L$ self-consistent equations, Eqs. (11), (15) and (16), so that we can uniquely determine a state by solving these equations. When the temperature approaches zero, one may find that the present approach will automaticlly recover spin wave theory which is believed to be the best for the zero-temperature case.[8,14]

The poles of the Green's function are the elementary excitations $E_m(\mathbf{k})$, while the gap is defined as the minimum value of the excitations: $\Delta(h)=\min[E_m(\mathbf{k})]$. According to Ref. 14, a spin configuration determined by Eq. (11) may be a stable or metastable state if there is a positive gap $(\Delta(h)>0)$ in elementary excitations to suppress the strong thermal fluctuations. The negative field h^{coer} at which the gap comes to zero is understood as the coercive force, $\Delta(h^{\mathrm{coer}})=0$, since the state cannot be metastable any longer at this field due to the strong fluctuations. Thus, all the physically interesting properties, such as the spin configurations, the surface magnon modes, the temperature dependence of the coercivity, the Curie temperature of the layered structures, etc., can be obtained by some simple numerical calculations.

III. TEMPERATURE DEPENDENCES OF THE COERCIVITY AND THE HYSTERESIS

First, we consider the coercivity and the hysteresis loop, which can be obtained analogous to the zero-temperature case.[14] When an external magnetic field is decreasing from a positive saturation value, the spin configuration must be $\{\theta_m=0\}$, and the induced magnetization of the system can be calculated by $M(h)=(1/L)\Sigma_m\overline{S_m}$ with the help of Eq. (15). As a applied field decreases across zero, the spins do not turn over at once although the configuration $\{\theta_m=\pi\}$ should actually be the ground state with a lower energy. In fact, in the state $\{\theta_m=0\}$, the magnon excitation still possesses a positive gap so that it is a metastable state. As a result, one can calculate the magnetic properties tracing the present spin configuration $\{\theta_m=0\}$ until the external field comes to a field when the excitation gap approaches zero. This field is just the coercive field. Thus, the total hysteresis loops at finite temperatures can be determined.

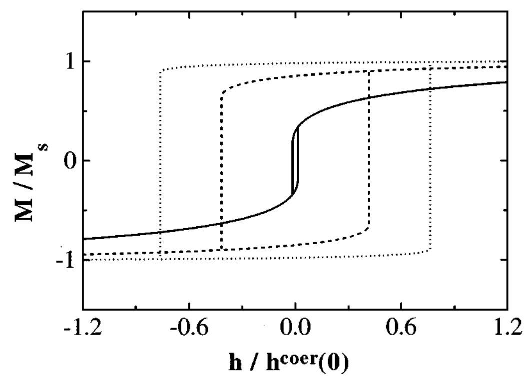

FIG. 1. Hysteresis loops for model 1 at low temperature (dotted line), at moderate temperature (dashed line), and near the Curie temperature (solid line).

For the model

$$\text{model 1:}\quad L=4,\quad I_{m,m'}(\mathbf{r},\mathbf{r}')=J,\quad D/J=1.0,$$

the hysteresis loops of a magnetic thin film at different temperatures are shown in Fig. 1 for comparison. When the temperature increases, the coercivity of the system is weakened, the hysteresis loops are smoothened, and the induced magnetization is more sensitive to the temperature. Especially in the case that the temperature approaches the Curie temperature, the present ferromagnetic system is very similar to a paramagnetic system—the coercivity is nearly zero and the magnetization is very sensitive to the external field. A very small negative field can turn the spins over (Fig. 1). The temperature dependence of the coercivity is shown in Fig. 2. This phenomenon has been well known and been applied in technological magnetism successfully. It usually costs a very strong magnetic force to write a message onto a domain which already has one at room temperature. To facilitate the process, the engineer heats the magnetic material so as to lower the coercivity, and then a small negative magnetism is

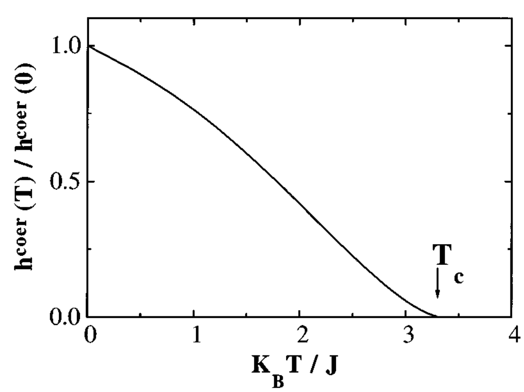

FIG. 2. The temperature dependences of the coercivity for model 1.

enough to turn the spins over. After that, the engineer cools the system to room temperature and such a message can be fixed successfully.

By the way, in this paper, we have chosen the uniaxil anisotropy to be much larger than those of the real material since we only want to show the qualitative picture of the results. Actually, we have made the calculations for the smaller anistropy case and find that the physical pictures are completely the same.

IV. MAGNETIC MULTIVALUED RECORDING

In this section, we will discuss how the thermal fluctuations influence the magnetic multivalued recording.[7,15] The basic idea of the MMV recording is to find such materials whose hysteresis loops are multistep shaped. A quantum model for the MMV recording has been proposed in Ref. 15 where the zero-temperature case is studied. It is a double-film structure where the main parameters are

$$I_{m,m}(\mathbf{r},\mathbf{r}')=J_m,\ \ I_{m,m'}(\mathbf{r},\mathbf{r})=I,\ \ D_m(2S_m-1)=\widetilde{D}_m,$$

$$\phi_m=0.$$

It is argued in Ref. 15 that the MMV recording can be achieved when the interlayer interaction I is not very strong compared to the anisotropy D_m and the coercivities of the two magnetic thin films must not be very close to each other. Without losing generality, it is assumed that $\widetilde{D}_1<\widetilde{D}_2$.

According to Ref. 15, the possible metastable states are the following four ones: $A(\theta_1=\theta_2=0)$, $B(\theta_1=\pi,\ \theta_2=0)$, $C(\theta_1=0,\ \theta_2=\pi)$, and $D(\theta_1=\theta_2=\pi)$. The metastable regions of the four states can be found following the steps described in the last section. One may find that the metastable region of spin state A is $h>h_A$, the metastable region for spin state B is $[-h_B^1,h_B^2]$, and straightforwardly, the metastable regions for spin states C and D are $[-h_B^2,h_B^1]$ and $(-\infty,-h_A]$, respectively.[15] According to Ref. 15, the condition for realizing the MMV recording is that the existing regions of the metastable states should overlap with each other.

In the zero-temperature case, it is found that the intralayer interaction J_m has nothing to do with these critical fields, and the condition for realizing the MMV recording is fully determined by the interlayer interaction I as well as the anisotropy $\widetilde{D}_m$.[15] However, in the finite-temperature case, the thermally excited magnons depend strongly on the intralayer interaction. As a result, if the Curie temperatures T_C of the two materials (depending on J_mS_m) have a large difference, the thermal fluctuations must affect the two films in distinct ways, and there may be some new and interesting results.

Subsequently, we show two different cases.

A. Normal case

If the magnetic material which has a small coercivity [determined by $\widetilde{D}_m=D_m(2S_m-1)$] also possesses a low Curie temperature, that is, $J_1S_1<J_2S_2$, the thermal fluctuations do not change the basic picture of the MMV recording. In this case, the coercive force of the first film decreases more quickly than the second one when temperature increases, so that the difference of the two coercivities are enlarged. To be specific, the following model parameters are chosen:

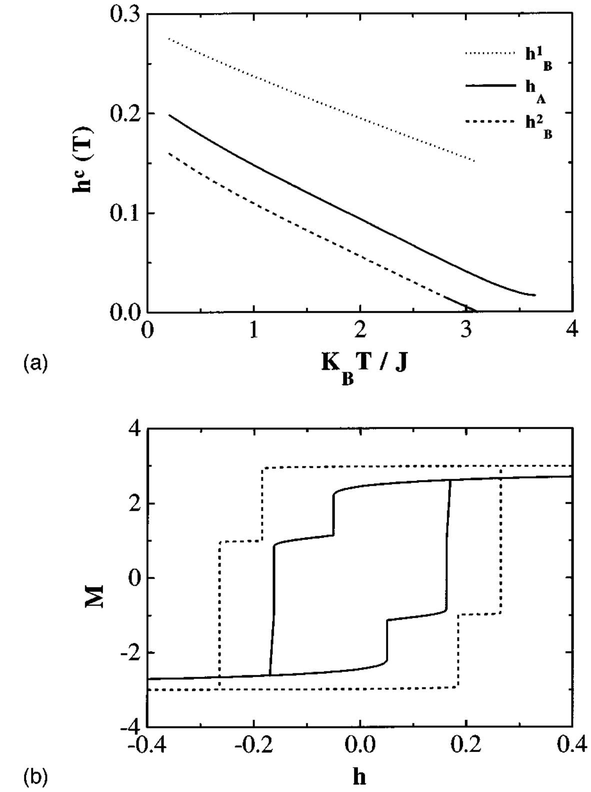

FIG. 3. (a) The critical values h_A, h_B^1, and h_B^2 as a function of the temperature for model 2. (b) Comparison of the magnetic multivalued recording for model 2 at low (dashed line) and high (solid line) temperatures.

$$\text{Model 2:}\quad S_1=1,\ S_2=2,\ \widetilde{D}_1=0.2,\ \widetilde{D}_2=0.3,$$

$$J_1S_1=3,\ J_2S_2=4,\ I=0.01.$$

The critical values h_A, h_B^1, and h_B^2 are shown in Fig. 3(a) as a function of the temperature, and the hysteresis loops of the systems at low and high temperatures are compared in Fig. 3(b). It can be clearly seen that the thermal fluctuations cannot change the main feature of the MMV recording so that such materials are good candidates for MMV recording.

B. Abnormal case

On the other hand, if $J_1S_1>J_2S_2$, thermal fluctuations are nontrivial. Although the coercivities of the two films will both decrease as the temperature increases, the velocities of the decrement are, however, different (see Fig. 2 for reference). Sooner or later, the coercivity of the first layer will become larger than the second one. At that time, different interesting phases are possible to appear. The following model is studied as an illustration:

$$\text{Model 3:}\quad S_1=1,\ S_2=2,\ \widetilde{D}_1=0.2,\ \widetilde{D}_2=0.3,$$

$$J_1S_1=5,\ J_2S_2=2,\ I=0.01.$$

h_A, h_B^1, and h_B^2 are shown together in Fig. 4(a). Three temperature regions are found for the MMV recording, and the

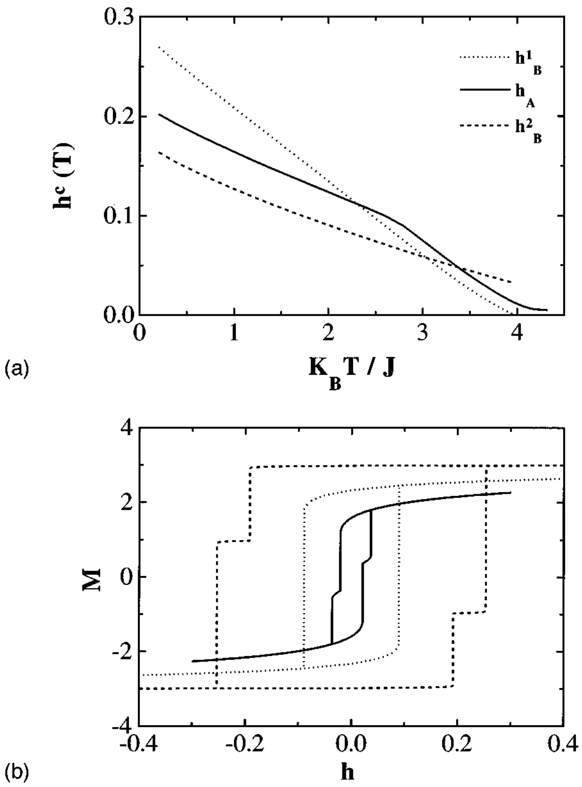

FIG. 4. (a) The critical values h_A, h_B^1, and h_B^2 as a function of the temperature for model 3. (b) Comparison of the magnetic multivalued recording for model 3 at low (dashed line), moderate (dotted), and high (solid line) temperatures.

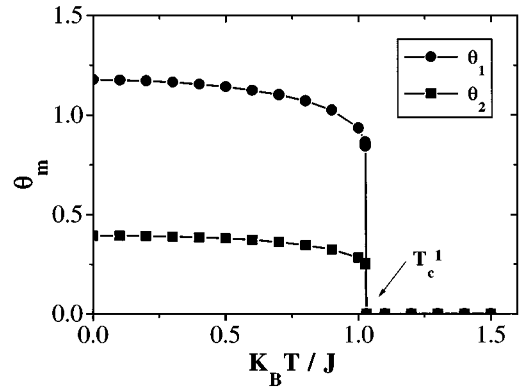

FIG. 5. The spin configuration θ_1 (up triangles) and θ_2 (circles) as a function of the temperature for model 4.

hysteresis loops in different cases are shown in Fig. 4(b) for comparison. When the temperature increases from zero, the multistep shape is obscured. At some temperatures, the two coercivities are so close to each other that the multistep shape disappears in the hysteresis loop. As the temperature is high enough, caused by different sensitivities to thermal fluctuations, it is possible to show another kind of multistep-shaped hysteresis loop in which the second film turns over earlier than the first one. In a technological process, these kinds of materials must be avoid being used as the recording media for MMV recording.

V. APPLICATION TO THE CANTED SPIN CONFIGURATION

In this section, we would like to give an application of the present method to the canted spin configuration case by discussing the temperature sensitivities of the spin configuration of a double-film structure.

Experimentally, a spin reorientation transition in ultrathin film was observed as the temperasture increases,[5] which is believed to be the result of a competition between the perpendicular anisotropy in the surface and the dipolar interactions which favor the spins to lie in the plane. This phenomenon has been appropriately explained by Ref. 17 through introducing the dipolar interaction term in their Hamiltonian and using a molecular-field approximation. We will not discuss this effect in this paper. On the other hand, the capping technique has been popularly used in experimental magnetism. By capping a magnetic thin film with different easy axes to another one, the total double-film structure may display many new and interesting effects.[4,6] We would like to discuss the following double-layer system as an illustration. Suppose the first film has an in-plane magnetic easy axis $\phi_1 = \pi$ and the second film a perpendicular magnetic easy axis $\phi_2 = 0$. In this case, unlike the last two sections, the nonlinear equations (11) are never trivial.

The following model is studied:

$$\text{model 4:} \quad D_1 = 0.2, \; D_2 = 0.2, \; J_1 = 1, \; J_2 = 2, \; I = 0.1,$$

$$S_1 = S_2 = 1.$$

The spin configuration $\{\theta_m\}$ is shown in Fig. 5 as a function of temperature. It can be clearly seen that there is a spin reorientation phase transition in a critical temperature T_c^1. The physics can be understood as follows: The spin configurations $\{\theta_m\}$ are determined by the competition of the effective anisotropy and the effective interlayer exchange interactions [Eq. (11)]. However, the temperature dependences of those terms are determined by the Curie temperature T_C, and different materials will have distinct temperature dependences (see Fig. 2). For the present system, the capping film has a low T_C and the recording one has a high T_C. When the temperature increases, the capping film becomes softer and softer (the coercivity and effective anisotropy decreases and decreases). As a result, at a critical temperature when the effectively in-plane anisotropy in capping film cannot compete with the effectively perpendicular anisotropy in recording film as it does in the zero-temperature case, the spins are all aligned in the z direction so that the spin reorientation transition occurs. One can easily understand another type of spin reorientation transition: If the capping film has a higher Curie temperature, the spins in the other film will be eventually aligned along the magnetic easy axis of the capping film.

VI. CONCLUSION

To summarize, in this paper, we have combined the Green's function technique with the previous local coordinates transformation to discuss some finite-temperature properties of magnetic multilyer systems. The nonlinear equations for determining the spontaneously magnetized directions in each layer have been derived by the minimization of the free energy, and the necessarily self-consistent equations have been obtained successfully following the standard Green's function technique. Some applications of the method have been presented. The temperature dependences of the coercivity and the hysteresis loop for layered structures have been shown, and the thermal fluctuations to the magnetic multivalued recording have been discussed, in which two cases are found to display different sensitivities to the thermal fluctuations. Finally, we study a more complicated model: the double-film structure composed of two magnetic films with different magnetic easy axes. The spin configurations are shown to have interesting sensitivities to the temperature.

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APPENDIX

In this appendix, we will show that Eq. (10) is correct under the random phase approximation, Eqs. (6)–(9).

Equation (10) should be correct if $\partial H/\partial\theta_m$ commute with H. Since the other terms have no contributions to Eq. (10), we need only consider $\partial H_2/\partial\theta_m$ which is

$$\frac{\partial H_2}{\partial\theta_m}=-\sum_{m'} I_{m,m'}\sin(\theta_m-\theta_{m'})S_m^{z_m}S_{m'}^{z_m}-h\sin\theta_m S_m^{z_m} -\frac{1}{2}D_m\sin[2(\theta_m-\phi_m)][(S_m^{z_m})^2-(S_m^x)^2]. \quad \text{(A1)}$$

Noting that under the same RPA as Eqs. (6)–(9), the commutations of $S_m^{z_m}$ with Hamiltonian are

$$[S_m^{z_m},H_1]=0, \quad \text{(A2)}$$

$$[S_m^{z_m},H_2]\overset{\text{RPA}}{\simeq}0, \quad \text{(A3)}$$

$$\begin{aligned}[S_m^{z_m},H_3]&=-\sum_{m'} I_{m,m'}\sin(\theta_m-\theta_{m'})S_m^{z_{m'}}S_m^{y_m}-h\sin\theta_m S_m^{y_m}\\&\quad-\frac{1}{2}D_m\sin[2(\theta_m-\phi_m)][S_m^{y_m}S_m^{z_m}+S_m^{z_m}S_m^{y_m}]\\&\overset{\text{RPA}}{\simeq}\Bigg\{-\sum_{m'} I_{m,m'}\sin(\theta_m-\theta_{m'})\overline{S}_{m'}\\&\quad-h\sin\theta_m-\frac{1}{2}D_m\sin[2(\theta_m-\phi_m)]\overline{S}_m\Psi_m\Bigg\}S_m^{y_m}.\end{aligned} \quad \text{(A4)}$$

According to Eq. (8), we find that the condition $[S_m^{z_m},H]=0$ coincides with Eq. (11). In other words, only when one has chosen a spin configuration which satisfies Eq. (11) can the condition $[S_m^{z_m},H]=0$ be automatically ensured under the RPA. If that is the case, it is not difficult to show that

$$[\partial H_2/\partial\theta_m,H]\simeq 0 \quad \text{(A5)}$$

under the RPA.

By the way, we would like to point out that the contributions from the terms $(S_m^+)^2+(S_m^-)^2$ have been neglected as an approximation which has also been adopted by other authors.[8–15] Actually, those terms have a spin-state mixing effect which is very important for ''easy-plane'' single-ion anistropy case.[23] For the present easy-axis case, the effect is significant only in very few cases.[23] To incorporate this effect, a characterestic angle (CA) method has been established for spin-1 and spin-$\frac{3}{2}$ cases.[23] Since the method has the limit of no university, we did not apply the CA method in the present paper.

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