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PHASE TRANSITION IN MAGNETIC ULTRA-THIN FILMS

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Abstract. In this paper, we study the influence of surface anisotropy on the phase transition in antiferromagnetic and ferromagnetic ultra-thin films using the functional integral method. Besides, spin fluctuations are also given to illustrate these phase transitions. We find that the phase transition temperature of the ultra-thin films may be higher or lower than that of the corresponding bulk systems, which depends on the surface anisotropy. Moreover, we also determine crossover points at which the phase transition temperature is not influenced by the thickness of the thin film.

Keywords: thin film, surface anisotropy, spin fluctuation, phase transition, functional integral method

1 Introduction

Two-dimensional (2D) systems have been extensively studied during the past decades due to the rich physical properties that they exhibit, especially the variety of their interesting magnetic phase transitions. A large number of recent experimental and theoretical studies have shown that the order-disorder phase transition in magnetic ultra-thin films may differ significantly from that in the corresponding bulk systems [3, 7, 10]. In the general case, the phase transition temperature (Curie temperature for the ferromagnetic (FM) thin film and Néel temperature for the antiferromagnetic (AFM) thin film) of the ultra-thin films is lower than that in the bulk and decreases when the thickness of the film reduces. However, in some special cases, such as Gd [1], Tb [5], and $NbSe_2$ [2], the phase transition temperature of the ultra-thin films is higher than that of the bulk. In these works, the authors also suggested that the presence of very large surface anisotropies causes the magnetic order at the surfaces above the bulk Curie temperature. Hence, we can see that one of the most important contributions for the unusual properties in thin films is their anisotropy at the surface. In general, it can be said that atoms at the surface state create a new phase with special properties such as low symmetric order and a decrease of the number of nearest neighbors (NN), which may cause several interesting physical properties [8].

In this paper, we investigate the phase transition in the magnetic ultra-thin film on the basis of the Heisenberg model via spin fluctuations using the functional integral method [3]. However, according to the theorem of Mermin and Wagner [9], long-range order cannot exist in the 2D isotropic Heisenberg system at a finite temperature due to the presence of large thermal spin fluctuations. Therefore, we give a surface anisotropy in the isotropic Heisenberg model [6]. The spin fluctuations, the magnetization, and then the phase transition temperature in the film should be influenced strongly by the surface anisotropy. The paper is organized as follows: In the theory section, we briefly give the key results, where we calculated for the AFM and FM thin film using the functional integral method. Section 3 deals with numerical results and discussion. First, we investigate the effect of the anisotropy (parameters K_s and J_0) at the surface for different numbers of the thickness of the thin film. Next, we discuss the important role of the spin fluctuations in the phase transitions, which are mentioned in the above part.

2 Theory

Consider a 2D system having *m* monolayers on a simple square lattice in the *Oxy* plane. In the system, the monolayers of *A* spins and the monolayers of *B* spins are arranged alternately. Therefore, the Heisenberg Hamiltonian of the system has the following form [6]

$$\begin{aligned} H = &-\frac{1}{2}\sum_{n,n'\neq n,j,j'}\sum_{\alpha=x,y,z} J_o\left(\vec{r}_{nj}-\vec{r}_{n'j'}\right) S^{\alpha}_{Anj}S^{\alpha}_{Bn'j'} \\ &-\frac{1}{2}\sum_{n,j,j'}\sum_{\alpha=x,y,z} J_i\left(\vec{r}_{nj}-\vec{r}_{nj'}\right) S^{\alpha}_{Anj}S^{\alpha}_{Anj'} \\ &-\frac{1}{2}\sum_{n,j,j'}\sum_{\alpha=x,y,z} J_i\left(\vec{r}_{nj}-\vec{r}_{nj'}\right) S^{\alpha}_{Bnj}S^{\alpha}_{Bnj'} \\ &\frac{1}{2}\sum_{n=1,m}\sum_{j} K_s\left(S^{z}_{Anj}\right)^2-\frac{1}{2}\sum_{n=1,m}\sum_{j} K_s\left(S^{z}_{Bnj}\right)^2, \end{aligned} \tag{1}$$

where *n* and *n′* are layer indices; $\vec{r}_{nj}$ is the position vector of the j^{th} spin in the n^{th} monolayer; the 1st term in (1) is the exchange interaction between spin S^{α}_{Anj} and spin $S^{\alpha}_{Bn'j'}$ in the NN monolayers. In this paper, we only consider the case of $S_A = S_B$ with two alignments of spins *A* and spins *B*, which are FM ($J_0 > 0$) or AFM ($J_0 < 0$); the second and third terms are the FM exchange interactions between the NN spins in the same monolayer ($J_i > 0$); the last is the uniaxial anisotropic term of the spins in the *Oz* direction (the *Oz* axis is perpendicular to the plane of the thin film), which is called *out-plane anisotropy*, we only consider the anisotropy at the surface and ignore that in the inner layers of the thin film. All the energies and temperatures are measured in the unit of the exchange constant *J* throughout the paper.

We choose the *Oz* direction to be the average alignment of the spins, so the spin fluctuations are defined as follows:

$$\begin{aligned} \delta S^{z}_{A(B)nj} &= S^{z}_{A(B)nj} - \left\langle S^{z}_{A(B)nj}\right\rangle,\ \delta S^{x}_{A(B)nj} \\ &= S^{x}_{A(B)nj},\ \delta S^{y}_{A(B)nj} = S^{y}_{A(B)nj} \end{aligned} \tag{2}$$

where $\beta^{-1} = k_B T$ and $\left\langle ...\right\rangle = \mathrm{Tr}\ \mathrm{e}^{-\beta H}...\Big/\mathrm{Tr}\ \mathrm{e}^{-\beta H}$. With the Fourier transformation of the spin operators

$$\begin{aligned} \delta S^{\alpha}_{A(B)nj}\left(\vec{k}\right) &= \frac{1}{\sqrt{N}}\sum_{j}\delta S^{\alpha}_{A(B)nj}\exp\left[-\mathrm{i}\vec{k}\vec{r}_{nj}\right], \\ &\alpha = x,y,z. \end{aligned} \tag{3}$$

where *N* is the number of the spins in every monolayer, Hamiltonian (1) of the system is rewritten as

$$\begin{aligned} H &= H_0 + H_{\text{int}}, \\ H_0 &= -\sum_{n,j}\frac{y_{nA}}{\beta}S^{z}_{Anj}-\sum_{n,j}\frac{y_{nB}}{\beta}S^{z}_{Bnj} \\ &\quad -\frac{N}{2}\sum_{n,n'\neq n,j'} J_o\left(\vec{k}=0\right)\left\langle S^{z}_{Anj}\right\rangle\left\langle S^{z}_{Bn'j'}\right\rangle \\ &\quad -\frac{N}{2}\sum_{n,j'} J_i\left(\vec{k}=0\right)\left\langle S^{z}_{Anj}\right\rangle\left\langle S^{z}_{Anj'}\right\rangle, \\ H_{\text{int}} &= -\frac{1}{2}\sum_{n,n',\vec{k}}\sum_{\alpha=x,y,z} J_{nn'}\left(\vec{k}\right)\delta S^{\alpha}_{n\vec{k}}\delta S^{\alpha}_{n',-\vec{k}}. \end{aligned} \tag{4}$$

where

$$\begin{aligned} y_{nA} &= \beta\left(\frac{1}{2}\sum_{n'\neq n,j'} J_o\left(\vec{k}=0\right)\left\langle S^{z}_{Bn'j'}\right\rangle+\sum_{j'} J_i\left(\vec{k}=0\right)\left\langle S^{z}_{Anj'}\right\rangle+K_s\delta_{n,1}\left\langle S^{z}_{Anj}\right\rangle+K_s\delta_{n,m}\left\langle S^{z}_{Anj}\right\rangle\right), \\ y_{nB} &= \beta\left(\frac{1}{2}\sum_{n'\neq n,j'} J_o\left(\vec{k}=0\right)\left\langle S^{z}_{An'j'}\right\rangle+\sum_{j'} J_i\left(\vec{k}=0\right)\left\langle S^{z}_{Bnj'}\right\rangle+K_s\delta_{n,1}\left\langle S^{z}_{Bnj}\right\rangle+K_s\delta_{n,m}\left\langle S^{z}_{Bnj}\right\rangle\right); \end{aligned} \tag{5}$$

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and $J_{nn'}(\vec{k})$ is the elements of a square matrix of *m* order $J(\vec{k})$, an example for matrix $J(\vec{k})$ with *m* = 4:

$$J(\vec{k}) = \begin{pmatrix} K_s + J_i(\vec{k}) & J_o(\vec{k}) & 0 & 0 \\ J_o(\vec{k}) & J_i(\vec{k}) & J_o(\vec{k}) & 0 \\ 0 & J_o(\vec{k}) & J_i(\vec{k}) & J_o(\vec{k}) \\ 0 & 0 & J_o(\vec{k}) & K_s + J_i(\vec{k}) \end{pmatrix} \tag{6}$$

and

$$\delta S^{\alpha}_{\vec{k}} \delta S^{\alpha}_{-\vec{k}} = \begin{pmatrix} \delta S^{\alpha}_{1A\vec{k}}\delta S^{\alpha}_{1A,-\vec{k}} & \delta S^{\alpha}_{1A\vec{k}}\delta S^{\alpha}_{2B,-\vec{k}} & \delta S^{\alpha}_{1A\vec{k}}\delta S^{\alpha}_{3A,-\vec{k}} & \delta S^{\alpha}_{1A\vec{k}}\delta S^{\alpha}_{4B,-\vec{k}} \\ \delta S^{\alpha}_{2B,\vec{k}}\delta S^{\alpha}_{1A,-\vec{k}} & \delta S^{\alpha}_{2B\vec{k}}\delta S^{\alpha}_{2B,-\vec{k}} & \delta S^{\alpha}_{2B\vec{k}}\delta S^{\alpha}_{3A,-\vec{k}} & \delta S^{\alpha}_{2B\vec{k}}\delta S^{\alpha}_{4B,-\vec{k}} \\ \delta S^{\alpha}_{3A\vec{k}}\delta S^{\alpha}_{1A,-\vec{k}} & \delta S^{\alpha}_{3A\vec{k}}\delta S^{\alpha}_{2B,-\vec{k}} & \delta S^{\alpha}_{3A\vec{k}}\delta S^{\alpha}_{3A,-\vec{k}} & \delta S^{\alpha}_{3A\vec{k}}\delta S^{\alpha}_{4B,-\vec{k}} \\ \delta S^{\alpha}_{4B\vec{k}}\delta S^{\alpha}_{1A,-\vec{k}} & \delta S^{\alpha}_{4B\vec{k}}\delta S^{\alpha}_{2B,-\vec{k}} & \delta S^{\alpha}_{4B\vec{k}}\delta S^{\alpha}_{3A,-\vec{k}} & \delta S^{\alpha}_{4B\vec{k}}\delta S^{\alpha}_{4B,-\vec{k}} \end{pmatrix}.$$

In (6),

$$\begin{aligned} J_i(\vec{k}) &= \sum_{\vec{h}} J_i(\vec{h}) \exp(-i\vec{k}\vec{h}) = 2J_i \cos(k_x.\mathrm{a}) + 2J_i \cos(k_y.\mathrm{a}), \\ J_o(\vec{k}) &= \sum_{\vec{h}} J_o(\vec{h}) \exp(-i\vec{k}\vec{h}) = J_o, \end{aligned} \tag{7}$$

where *a* is the distance between the two NN spins in a monolayer of the thin film and *b*(*y*) is the Brillouin function

$$b(y_{A,B}) = (S_{A,B} + \frac{1}{2})\mathrm{cth}(S_{A,B} + \frac{1}{2})y_{A,B} - \frac{1}{2}\mathrm{cth}\frac{y_{A,B}}{2}. \tag{8}$$

The free energy of the thin film is calculated as follows:

$$\begin{aligned} F = -\frac{1}{\beta}\ln\mathrm{Tr}\left(e^{-\beta H}\right) &= -\frac{1}{\beta}\ln\mathrm{Tr}\left(e^{-\beta H_0}\right) - \frac{1}{\beta}\ln\int d\varphi \exp\left\{-\frac{1}{2}\sum_{n,\vec{k},\alpha}\varphi^{\alpha}_{n}(\vec{q})\varphi^{\alpha}_{n}(-\vec{q})\right\} \\ &\times\left\langle \mathrm{T}\exp\left\{\sum_{n,n',\vec{k},\alpha}\left(\beta J_{nn'}(\vec{k})\right)^{1/2}\varphi^{\alpha}_{n}(\vec{q})\,\delta S^{\alpha}_{n'}(\vec{q})\right\}\right\rangle_0, \end{aligned} \tag{9}$$

where

$$d\varphi = \prod_{\alpha,n}\int_{-\infty}^{+\infty}\frac{d\varphi^{\alpha}_{n}(0)}{\sqrt{2\pi}}\prod_{\vec{q}\neq 0}\int_{-\infty}^{+\infty}\frac{d\varphi^{\alpha,c}_{n}(\vec{q})}{\sqrt{\pi}}\int_{-\infty}^{+\infty}\frac{d\varphi^{\alpha,s}_{n}(\vec{q})}{\sqrt{\pi}}. \tag{10}$$

Using the functional integral method given in details in [3], we achieve the last expression of the free energy for the thin film

$$\begin{aligned} F = \frac{N}{2}\sum_{n}J_1(0)\left\langle S^z_{nA}\right\rangle\left\langle S^z_{nB}\right\rangle + \frac{N}{2}\sum_{n\neq n'}J_2(0)\left\langle S^z_{nA}\right\rangle\left\langle S^z_{n'A}\right\rangle + \frac{N}{2}\sum_{n\neq n'}J_2(0)\left\langle S^z_{nB}\right\rangle\left\langle S^z_{n'\mathrm{B}}\right\rangle \\ -\frac{N}{\beta}\sum_{n}\ln\frac{\mathrm{sh}(S_A + 1/2)y_{nA}}{\mathrm{sh}(y_{nA}/2)} - \frac{N}{\beta}\sum_{n}\ln\frac{\mathrm{sh}(S_B + 1/2)y_{nB}}{\mathrm{sh}(y_{nB}/2)} \\ +\frac{1}{2\beta}\sum_{\vec{k}}\ln\left(\det\left[I - C(\vec{k})\right]\right) + \frac{1}{2\beta}\sum_{\vec{k},\omega}\ln\left(\det\left[I - E(\vec{k},\omega)\right]\right) \end{aligned} \tag{11}$$

where

$m = 4:$

$$C(\vec{k}) = \begin{pmatrix} \beta(K_s + J_i(\vec{k}))b' y_{1A} & \beta J_o(\vec{k})b' y_{2B} & 0 & 0 \\ \beta J_o(\vec{k})b' y_{1A} & \beta J_i(\vec{k})b' y_{2B} & \beta J_o(\vec{k})b' y_{3A} & 0 \\ 0 & \beta J_o(\vec{k})b' y_{2B} & \beta J_i(\vec{k})b' y_{3A} & \beta J_o(\vec{k})b' y_{4B} \\ 0 & 0 & \beta J_o(\vec{k})b' y_{3A} & \beta(K_s + J_i(\vec{k}))b' y_{4B} \end{pmatrix}$$

and

$$E(\vec{k}) = \begin{pmatrix} \frac{\beta(K_s + J_i(\vec{k}))b y_{1A}}{y_{1A} - i\beta\omega} & \frac{\beta J_o(\vec{k})b y_{2B}}{y_{2B} - i\beta\omega} & 0 & 0 \\ \frac{\beta J_o(\vec{k})b y_{1A}}{y_{1A} - i\beta\omega} & \frac{\beta J_i(\vec{k})b y_{2B}}{y_{2B} - i\beta\omega} & \frac{\beta J_o(\vec{k})b y_{3A}}{y_{3A} - i\beta\omega} & 0 \\ 0 & \frac{\beta J_o(\vec{k})b y_{2B}}{y_{2B} - i\beta\omega} & \frac{\beta J_i(\vec{k})b y_{3A}}{y_{3A} - i\beta\omega} & \frac{\beta J_o(\vec{k})b y_{4B}}{y_{4B} - i\beta\omega} \\ 0 & 0 & \frac{\beta J_o(\vec{k})b y_{3A}}{y_{3A} - i\beta\omega} & \frac{\beta(K_s + J_i(\vec{k}))b y_{4B}}{y_{4B} - i\beta\omega} \end{pmatrix}.$$

The dependence of the phase transition temperature on the thickness of the thin film can be derived from the logarithmic singularity of the free energy in the zero field, $y = 0$, and in the long wavelength limit $\vec{k} \to 0$:

$$\det\left[I - C(\vec{k})\right] = 0, \tag{12}$$

in (12) $\beta = \beta_C = \frac{1}{k_B T_C}$.

3 Numerical results and discussion

The numerical results of the dependence of the reduced phase transition temperature $\tau_C = k_B T_C / J$ on the thickness of the thin film are shown in Fig. 1 with the various values of the uniaxial anisotropy parameter K_s at the surface of the thin film. From Fig. 1, we can see two obvious cases for the phase transition temperature of the thin film according to the values of K_s/J:

Case 1: $K_s / J < 1$; this case is called the weak surface anisotropy. The phase transition temperature rather quickly increases with the increasing monolayer number and reaches that of the bulk with an identical value of K_s, which agrees with the experimental results given in [7] and [10]. In this case, the exchange interaction between spins in the bulk systems is more than that in the thin film due to a decrease in the number of NN spins, which results in a reduction in the magnetic order, and thus a decreased τ_C.

Case 2: $K_s / J > 1$ is the strong surface anisotropy. Contrary to Case 1, the phase transition temperature decreases when the thickness increases, and the phase transition temperature of the ultra-thin films is higher than that of the bulk, which may be used to illuminate the experimental results reported in [5], in which the authors proposed that very high anisotropy strongly affects the magnetic ordering at the surface layer of the Tb samples. Physically, we can understand that

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the magnetic order is firmer in the thin film than in the corresponding bulk system because the thin film possesses a strong surface anisotropy that favors the FM/AFM order, while no such anisotropy occurs in the bulk system.

The boundary value between the two cases above depends on the values of the parameters J_i and J_0. For example, when J_i/J = J_0/J = 1, the boundary value K_s/J = 1 (Fig. 1). Besides, from Fig. 1, one can see that the phase transition temperature increases with K_s/J when we fix the monolayer number *m* and quickly increase/decrease with the increasing of *m* in the ultra-thin region when K_s/J is fixed. It is obvious that τ_C is affected strongly by the surface anisotropy K_s/J when the thickness *m* is small, which results from an appearance of the surface atoms with low symmetric order and a significant reduction of nearest neighbors in the ultra-thin film. In both cases of the strong and weak anisotropies, the phase transition temperature tends to that of the bulk when *m* increases.

Fig. 2 shows the dependence of the reduced phase transition temperature on the uniaxial anisotropic parameters for different numbers of monolayers. We call this a $K_s/J, \tau_C$ phase diagram, and here we choose $J_0/J = J_i/J = 1$. From this figure., we can define a crossover point with the critical parameter $K_{sC}/J = 1$, at which the phase transition temperature of the thin film does not depend on the thickness of it. Therefore, from this crossover point, we can determine the critical temperature of the corresponding bulk system. This special point corresponds to the green dot line (m, τ_C, K_s/J = 1) in Fig. 1. We think the existence of this point is due to the geometry of the thin film and the influence of the surface anisotropy. The spin on the surface interacts with the four NN spins in the same monolayer and one spin in the NN monolayer. Whereas, the one in the bulk system interacts with the four NN spins in the same monolayer and two spins in the NN monolayers. Therefore, the crossover point corresponds to the case when the surface anisotropy parameter K_s/J offsets the inadequacy of an NN spin. We give examples for this crossover point when changing the exchange parameters. When J_0/J = 1 and J_i/J =1, we have K_{sC}/J = 1; when J_0/J = 0.5 and J_i/J =1, we have K_{sC}/J = 0.5.

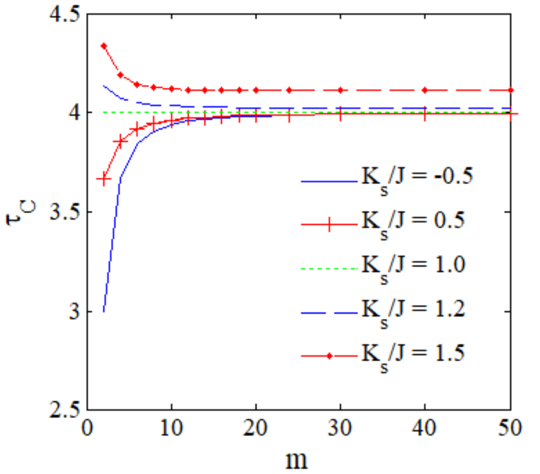

Fig. 1. Dependence of reduced phase transition temperature on thickness of thin film with various values of surface anisotropic parameter K_s/J (J_0/J = 1, J_i/J = 1)

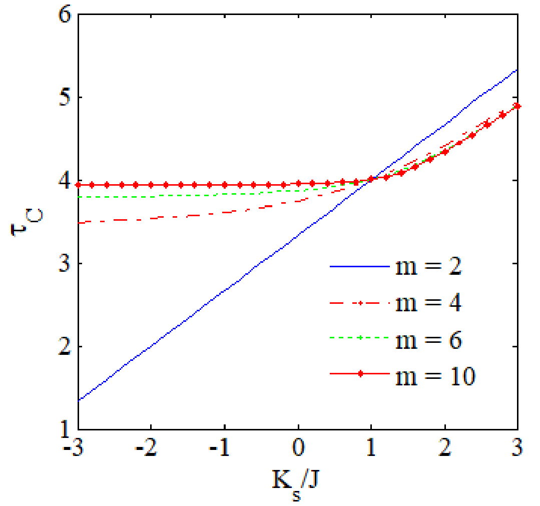

Fig. 2. Dependence of reduced phase transition temperature on surface anisotropic parameter K_s/J when increasing thickness of thin film (J_0/J = 1 and J_i/J = 1)

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Moreover, from Fig. 2, we also find that the phase transition temperature increases with an increas of K_s/J. That is because, in this paper, we choose the *Oz* direction for both the average alignment and the direction of the uniaxial anisotropy of the spins, so the parameter K_s/J will support the magnetic order in the *Oz* direction and then the phase transition temperature in the thin film. In [4], the authors also showed that a positive uniaxial anisotropic parameter ($K_s/J > 0$) favors large values of the spin's *z*-projection, and the thin film has an easy-axis in the *Oz* direction, which is an energetically favorable direction of spontaneous magnetization; with a negative uniaxial anisotropic parameter ($K_s/J <$ 0), the spin tends to minimize the *z*-component of its magnetic moment so that the system has an easy-plane orthogonal to the *Oz* axis. These theoretical points can be explained from the spin fluctuations given in Fig. 3 and Fig. 4. From Fig. 3, we can see that the *x*/*y*-components $\delta S^{x,y}$ of the spin fluctuation are large for $K_s/J < 0$ and decrease significantly when $K_s/J > 0$ and vice versa for the *z*-component δS^z of the spin fluctuation, which leads to a reduction of the total spin fluctuation given in Fig. 4. Hence, the magnetization m_z and then the phase transition temperature (defined at $m_z \ \tau_C \ = 0$ in Fig. 5) of the thin film also increase correspondingly. Thus, we find out that the influence of the spin fluctuations on the magnetic order in the ultra-thin film is significant and can be managed by the surface anisotropy.

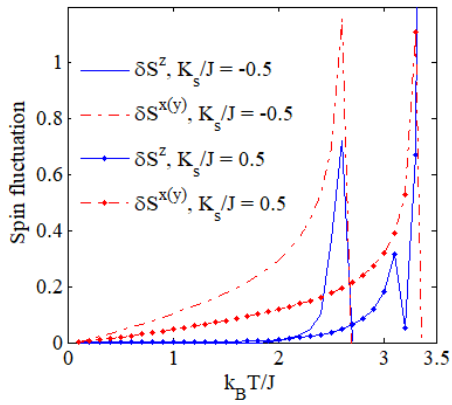

Fig. 3. x, *y* and *z*-components of spin fluctuation as function of reduced temperature *T*/*J* with different values of surface anisotropic parameter K_s/J (*m* = 2)

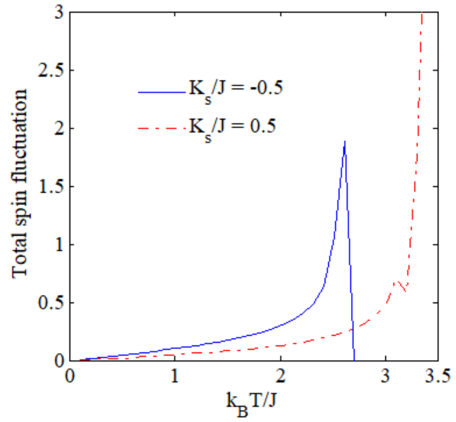

Fig. 5. Total spin fluctuation as a function of reduced temperature *T*/*J* with different values of surface anisotropic parameter K_s/J (*m* = 2)

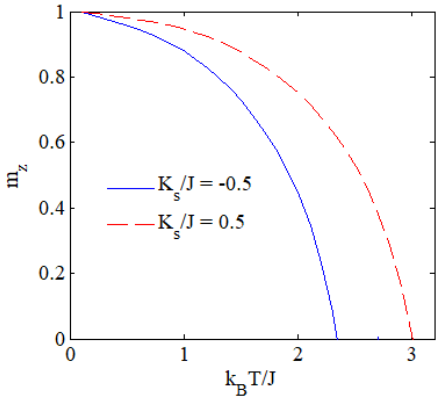

Fig. 4. *z*-component of spin magnetic moment as function of reduced temperature *T*/*J* with different values of surface anisotropic parameter K_s/J (*m* = 2)

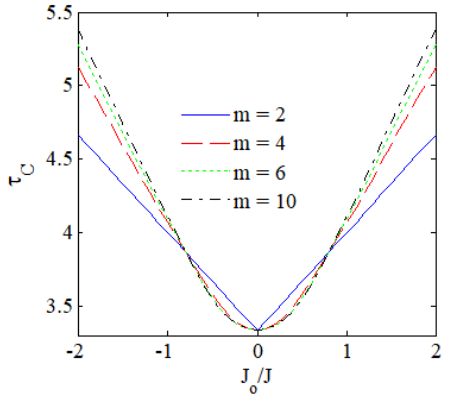

Fig. 6. Dependence of reduced phase transition temperature on exchange parameter J_0/J when increasing thickness of thin film ($K_s/J = 0.5$ and $J_i/J = 1$)

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Besides, in this paper, we also consider the dependence of the reduced phase transition temperature on the exchange parameter J_0/J between spins in two NN monolayers with two cases: FM ($J_0 < 0$) and AFM ($J_0 > 0$). We find that the FM or AFM exchange interaction (i.e., sign of J_0) does not affect the phase transition in the magnetic film. Only the value of J_0 takes an important role (Fig. 6) because the exchange parameter J_0 causes an alignment of the spins in the NN monolayers in the FM order $\begin{matrix} \cdots\uparrow\uparrow\uparrow\cdots \\ \cdots\uparrow\uparrow\uparrow\cdots \end{matrix}$ with $J_0 > 0$ or the AFM order $\begin{matrix} \cdots\uparrow\uparrow\uparrow\cdots \\ \cdots\downarrow\downarrow\downarrow\cdots \end{matrix}$ with $J_0 < 0$. The larger the value of J_0 is, the more stable is the FM or AFM order, so the phase transition temperature τ_C increases with the increase in the value of J_0/J. In this figure., we also find a crossover point due to the presence of the uniaxial anisotropy at the surface of the thin film $K_s/J = 0.5$.

4 Conclusions

In this paper, using the functional integral method, we investigate the $K_s/J, \tau_C$ and $J_o/J, \tau_C$ phase diagrams of the magnetic thin films via the thermal spin fluctuations. From these diagrams, we determine the crossover points at which the thickness of the thin film does completely not affect the phase transition temperature of the system. The exchange interaction and the uniaxial anisotropy at the surface make a change in the thermal spin fluctuations, which strongly affects the phase transition of the thin film.

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