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Temperature dependence of exchange bias and coercivity in ferromagnetic/antiferromagnetic bilayers

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Abstract. A model for the temperature dependence of exchange bias and coercivity in epitaxial ferromagnetic (FM)/ antiferromagnetic (AFM) bilayers is developed. In this model, the interface coupling includes two contributions, the direct coupling and the spin-flop coupling. The temperature dependence arises from the thermal disturbance to the system, involved in the thermal fluctuations of magnetization of AFM grains and the temperature modulation of the relevant magnetic parameters. In addition, the randomness of original orientations of easy axes of AFM grains after field cooling is taken into account. A self-consistent calculation scheme is proposed and numerical treatment is carried out. The results show that the temperature dependence of exchange bias and coercivity is closely related to the sizes of AFM grains and the interface exchange coupling constants. Especially, the exchange bias will have a peak and the blocking temperature will increase if the spin-flop coupling plays a role. On the other hand, the original orientation distribution of easy axes of AFM grains will affect exchange bias and coercivity prominently. The prediction has been well supported by experiments.

PACS. 75.30.Et Exchange and superexchange interactions – 75.50.Ee Antiferromagnetics – 75.30.Gw Magnetic anisotropy

1 Introduction

The existence of interfacial exchange coupling between a ferromagnetic (FM) layer and an antiferromagnetic (AFM) layer significantly modifies the magnetic properties of this kind of FM/AFM bilayers. The most well-known effect is the shift of hysteresis loop of the FM layer, called the exchange bias, which was firstly discovered in partially oxidized Co particles more than 40 years ago [1], then also verified in FM/AFM bilayers [2,3]. Other important effects have been observed, for example, almost all FM layers show an increase in the coercivity, and there exists a unusual shift in the ferromagnetic resonance [4–6]. These effects have attracted much attention due to their application to giant magnetoresistive spin-valve heads for high-density recording systems [7]. Many experimental results have been reported that the characteristics of exchange bias and coercivity in FM/AFM bilayers depend on the constituent materials, their thicknesses, the orientations of applied fields, and the temperature [2,3,8–10]. As usual, the exchange coupling can be modelled as an exchange anisotropy field which will add vectorially to an external field. However, magnetic devices, such as the read heads based on the above effects, are always operated in surroundings of variable temperatures, so the temperature dependence of exchange bias and coercivity needs to be illuminated.

Many experimental results [2,3,8–10] have shown that the temperature dependence of exchange bias and coercivity displays some common behavior while their relative values may be different. The principal characteristics is that both the exchange bias and coercivity decrease with increasing temperature at lower temperatures. However, with further increasing temperature the exchange bias generally gets smaller while the coercivity tends to increase and reach a peak. Finally the exchange bias approaches zero and the coercivity decreases again with increasing temperature up to a blocking temperature, at which the exchange bias disappears. It is interesting that, in some situations, one has also experimentally observed a peak or flat in curves of exchange bias versus temperature [9–12]. There are several models aiming at explaining qualitatively the temperature dependence of exchange bias and coercivity. For example, C. Hou et al. predicted that the peaks of exchange bias of FM/AFM bilayers related to temperature are more likely to observe if the Néel temperature is higher than the Curie temperature. However, in other situations with the Néel temperature lower than the Curie temperature, one also find a peak or flat

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region for exchange bias vs. temperature [12]. How to understand this phenomenon seems still difficult partly due to the details of the temperature dependence of magnetic parameters, such as the interface exchange coupling, the anisotropies of FM and AFM layers, and the size of AFM grains, remain unclear. Moreover, more recently, there has been much study of domain formation in the AFM from a microscopic point of view [13,14]. It could be expected that the domain formation in the AFM is crucial for the existence of exchange bias, especially for the compensated interfaces. In addition, it has been found that there may exist different behavior for ascending and descending branches in hysteresis loops corresponding to different reversal mechanisms [15–17].

In the present work, we will investigate the temperature dependence of exchange bias and coercivity associated with magnetic parameters of materials in order to understand the above phenomena and to find an optimal composition of FM/AFM bilayers to improve their performance. The Fulcomer and Charap model [18] is generalized to include both the bilinear (direct exchange) and biquadratic (spin-flop) exchange couplings. While the FM layer is assumed to be perfect with uniaxial anisotropy, the AFM layer is composed of many AFM grains, and each grain has uniaxial anisotropy. Furthermore, the temperature dependence results from the thermal disturbance including the thermal fluctuations of magnetization of antiferromagnetic grains and the temperature modulation of magnetic parameters. Especially, the orientation distribution of easy axes of antiferromagnetic grains resulted from temperature driving during the sample preparation under cooling field is considered. Based on the above model, it is more reasonable to illustrate the occurrence of a peak in the temperature dependence of exchange bias of FM/AFM systems when the Néel temperature is lower than the Curie temperature if the interface coupling includes the spin-flop coupling.

The paper is organized as follows: Section 2 is devoted to the model and method for investigating the temperature dependence of FM/AFM bilayers. In Section 3 the numerical results for the dependence of exchange bias and coercivity on the temperature and on other parameters are obtained and discussed. Finally, a main conclusion is given in Section 4.

2 Model and method

We first introduce the model to derive the dependence of exchange bias and coercivity on the temperature and other parameters for exchange coupled FM and AFM bilayers. Our model is based on the Meiklejohn-Bean model and its extension. As a phenomenological model, the domain wall pinning may be combined into the anisotropy energy of AFM grains and the domain wall motion approximately through the effect of energy barriers for AFM grains with different sizes. Then our model is to consider that the FM magnetization and AFM net sublattice magnetization to be uniform due to the fact that almost all experiments have been done in magnetic fields high enough to saturate the FM magnetization, while AFM grains have superparamagnetism. An FM/AFM system is now composed of one perfect FM layer coupled to N AFM grains which are arranged in a layer. The thickness $t_{\rm FM}$ of the FM layer is thin enough that there is no any domain walls inside the ferromagnet. All AFM grains have the same height d but different interface areas S_i. All spins in each grain are assumed to behave coherently, because all the AFM grains are assumed so small that they can be taken as single domains. On the other hand, individual grains are considered with cylindrical symmetry, so the contact surface between each pair of grains appears small, and the contribution from the interaction of intergrains is neglectable compared with that of innergrains. Additionally, the intergrain coupling will hinder the reversal of a grain under an applied field, it is possible to combine this effect into the anisotropic parameters. Therefore, for simplicity, it is also assumed here that there is no any interaction between the AFM grains, but exchange couplings exist between the FM layer and individual AFM grains. The behavior of AFM grains is closely related to their superparamagnetism. Furthermore, both the FM layer and AFM grains have only uniaxial magnetic anisotropies with the FM easy axes parallel to the same direction of the external field. The interface is considered to lie in the x-y plane with the z axis normal to the interface. All magnetization and applied field are assumed within the x-y plane.

Many experiments have confirmed that there is no helical structure along the z axis in a thin FM layer. For example, Parkin et al. [19] have observed that a uniform magnetization distributed throughout the thickness of a 400 Å $Ni_{80}Fe_{20}$ layer coupled with a $Fe_{50}Mn_{50}$ layer. In fact, an early theoretical study also suggested it [20]. Therefore, we can believe that the FM moments rotate uniformly in the presence of an applied field. In general, the bilinear coupling between the ferromagnet and antiferromagnet play a dominating role for exchange bias [2,3]. However, for a large amount of FM/AFM systems, it is attested theoretically and experimentally that the existence of a 90° angle coupling between magnetization directions at FM/AFM interface is normal and is shown to be related to the well-known "spin-flop" state [21–26], which is due to the frustration of spins at the interface, i.e., the interfacial AFM moments will tend to align themselves perpendicular to the FM easy axis. This spin-flop state contributes a biquadratic coupling to FM/AFM bilayers. Theoretical works have shown that the biquadratic coupling cannot be responsible for exchange bias but contribute to coercivity. However, when both bilinear and biquadratic coupling terms exist in FM/AFM bilayers at the same time, the exchange bias will be perturbed by the biquadratic term while it is mainly determined by the bilinear coupling. Even for a partially compensated interface, the exchange bias can be affected by the biquadratic coupling [27–30]. We therefore consider that such a spin-flop configuration could occur in our FM/AFM bilayers, and investigate its implications. So, in the present model, the bilinear as well as the biquadratic interface coupling terms are taken into account.

Based on the discussion above, the total energy of an exchange coupled FM/AFM bilayer in the presence of an applied field H is given by

$$E = K_{\rm FM} S_{\rm FM} t_{\rm FM} \sin^2\theta - H M_{\rm FM} S_{\rm FM} t_{\rm FM} \cos\theta + \sum_{i=1}^{N} \big[K_{\rm AFM} S_i d \sin^2\phi_i - f(\alpha_i) J_{\rm E1} S_i \cos(\theta - \alpha_i - \phi_i) + f(\alpha_i) J_{\rm E2} S_i \cos^2(\theta - \alpha_i - \phi_i) \big], \quad (1)$$

where $K_{\rm FM}$ and $K_{\rm AFM}$ are the uniaxial anisotropy constants of the FM layer and AFM grains, respectively; θ, ϕ_i and α_i are the angles between the FM magnetization and the FM anisotropy axis, the AFM sublattice magnetization and the AFM anisotropy axis of the ith grain, and its AFM anisotropy axis and the applied field which is collinear with the cooling field, the subscript i is summed over all AFM grains; $f(\alpha_i)$ is the distribution function for the occupied probability of the ith AFM grain with its easy axis deviating from the cooling field denoted by α_i, which depends on the sample's preparation, and the nucleation process of the AFM layer under the cooling field; $M_{\rm FM}$ is the saturated magnetization of the FM layer with its interface area $S_{\rm FM}$ and thickness $t_{\rm FM}$. The first term in equation (1) is the uniaxial anisotropy energy of the FM layer, while the second term is the Zeeman energy. In the square bracket in equation (1), the first term is the uniaxial anisotropy energy of an AFM grain; the second term is the direct unidirectional coupling energy with the bilinear coupling constant $J_{\rm E1}$; the third term is the spin-flop coupling energy with the biquadratic coupling constant $J_{\rm E2}$, and in general, $J_{\rm E1} \gg J_{\rm E2}$.

At equilibrium, the first derivative of the total energy E with respect to the angle ϕ_i must be equal to zero. From equation (1), we can obtain a set of equations satisfying energy minimum, which determine the angle ϕ_i under fixed values of θ and α_i. Actually there are two values of ϕ_i for any given values of θ and α_i to satisfy the energy minimum equations. This means that when the magnetization of the FM layer rotates around, each AFM grain has two stable states in all possible directions of its magnetization. Hereafter, we call these stable states correspondingly with "state 1" and "state 2", respectively. In order to obtain the equilibrium energy of the system for fixed values of θ and α_i, we should know the occurrence probability of both state 1 and state 2 of each AFM grain magnetization. Using $E_{i1(2)}$ to denote the energy of the ith AFM grain in state 1 (2), we can get the probability of this AFM grain magnetization in state 1 as

$$p_{i1} = 1/\left\{1 + \exp[(E_{i2} - E_{i1})/k_{\rm B}T]\right\}. \quad (2)$$

The probability of its AFM magnetization in state 2 denoted by p_{i2} is obtained by the condition $p_{i1} + p_{i2} = 1$. Then statistically the equilibrium energy of the ith AFM grain can be written as

$$E_i(\theta, \alpha_i) = p_{i1} E_{i1} + p_{i2} E_{i2}, \quad (3)$$

where α_i could be distributed from $-\pi/2$ to $\pi/2$. However, for a certain deposition temperature under which the sample is prepared after field cooling, a set of α_i, i.e., $f(\alpha_i)$, is determined. The total equilibrium energy of the system is obtained by summing over individual energies of all grains with the probability distribution of their anisotropy axes $f(\alpha_i)$, so

$$E(\theta) = \sum_{i=1}^{N} \int_{-\pi/2}^{\pi/2} (p_{i1} E_{i1} + p_{i2} E_{i2}) f(\alpha_i) d\alpha_i. \quad (4)$$

It is important to note that $E(\theta)$ should be obtained by $E_i(\theta, \alpha_i)$ self-consistently. This means that the total energy $E(\theta)$ must reach the minimum value for different distribution α_i. This is fulfilled by numerical calculations.

Now the total energy $E(\theta)$ is determined by the magnitude and the orientation of an applied field. By minimizing the total energy $E(\theta)$ in any fixed value of the applied field, we can make clear the magnetization process of the FM/AFM bilayer. Then according to reference [31], we can calculate coercivity and exchange bias. In the process of our calculations, the energy is scaled by $K_{\rm AFM} d S_{\rm FM}$, i.e.,

$$\epsilon(\theta) \equiv E(\theta)/K_{\rm AFM} d S_{\rm FM} = k_{\rm FM} \sin^2\theta - h\cos\theta + \sum_{i=1}^{N} \{ s_i \sin^2\phi_i - f(\alpha_i) j_1 s_i \cos[\theta - (\alpha_i + \phi_i)] + f(\alpha_i) j_2 s_i \cos^2[\theta - (\alpha_i + \phi_i)] \} \quad (5)$$

where $k_{\rm FM} = K_{\rm FM} t_{\rm FM}/K_{\rm AFM} d$, $h = H M_{\rm FM} t_{\rm FM}/K_{\rm AFM} d$, $s_i = S_i/S_{\rm FM}$, $j_1 = J_{\rm E1}/K_{\rm AFM} d$, $j_2 = J_{\rm E2}/K_{\rm AFM} d$. The coercivity and the exchange bias are scaled by $M_{\rm FM} t_{\rm FM}/K_{\rm AFM} d$.

For a real sample, AFM grains have a rather continuous distribution in their sizes over a certain range. Typically, we assume that the sizes of AFM grains exhibit log-normal distribution [32]

$$P = C \exp\left\{ -[\ln(s_i/s_0)]^2/8\sigma^2 \right\}, \quad (6)$$

where $s_0 = S_0/S_{\rm FM}$ is the reduced average area in which S_0 is defined as the average area for all AFM grains; C and σ are two constants. It is reasonable to take $C = 1$, $\sigma = 0.27$. In fact, the number of AFM grains N is variable, but is satisfied by $N s_0 \equiv 1$.

There are two advantages of our model compared to previous work [11,18,32–34]. Firstly, there is a competition between the bilinear and biquadratic terms, which may provide versatile choices for the exchange bias effect. It means that the interface is treated as partial compensation by the biquadratic coupling. Secondly, the thermal fluctuation of magnetization of AFM grains as well as the original orientation distribution of easy axes of AFM grains after field cooling are considered and determined self-consistently. Especially, the latter is exactly new and more practical compared to the previous work [34].

3 Numerical results and discussion

In the numerical calculations, we take the reduced values of the magnetic parameters (such as $k_{\rm FM}$, s_i and $j_{1(2)}$ etc.)

just for convenience to describe both FM and AFM layers. Obviously, the values of these related magnetic parameters used in the numerical calculations are not arbitrary, but instead determined by exchange and anisotropy energies associated with the antiferromagnet and interface in real materials. So to assure the reasonableness of our theoretical approach, we would adopt the related values of magnetic parameters based on the $Ni_{80}Fe_{20}/Fe_{50}Mn_{50}$ bilayer. In the present FM/AFM system, it is assumed that the Curie temperature of the ferromagnet is much greater than the Néel temperature of the antiferromagnet as usual. Thus, the temperature relation of AFM magnetization and the magnetocrystalline anisotropy will dominate the temperature dependence of exchange bias and coercivity. In contrast, it has no problem to assume that the FM magnetization and its magnetocrystalline anisotropy are independent of temperature.

In our model, two contributions are included for the temperature dependence. The first is associated with magnetic parameters, such as the exchange couplings at the interface and the uniaxial anisotropy of AFM grains. The second is involved in the superparamagnetism of AFM grains. For the former, it may be reasonable to consider that the temperature dependence of magnetic parameters for AFM bulks is still available to grains when their sizes are larger than several nanometers. It is found in the Stoner's theory that the AFM magnetization $M_{\mathrm{AFM}} \propto \left[1-(T/T_{\mathrm{N}})^2\right]^{1/2}$ [35], which can be used to specify the AFM anisotropy $K_{\mathrm{AFM}} \propto M_{\mathrm{AFM}}^3$, the interface exchange couplings $J_{\mathrm{E1}} \propto M_{\mathrm{AFM}}$, and $J_{\mathrm{E2}} \propto M_{\mathrm{AFM}}^2$ [11], where T is the temperature of the system, and T_{N} is the Néel temperature. For the latter, the original distribution of easy axes of AFM grains for samples prepared after field cooling is considered and determined self-consistently, and thermal disturbance of magnetic states of AFM grains is described by equations (2) and (3).

The numerical result of the distribution $f(\alpha_i)$ is shown in Figure 1. It is clear that the distribution $f(\alpha_i)$ is different for different depositing temperature T_{D} under which samples are prepared. As shown in Figure 1, we can find that at higher deposition temperature the easy axes of grains can be taken as randomly distribution in x-y plane, but at lower deposition temperature the easy axes of grains are mainly laid on the cooling field direction. We find that $f(\alpha_i)$ is generally a little similar to the canonical distribution. This is reasonable. For a given sample, when the measured temperature increases the FM magnetization will be reduce, but the distribution $f(\alpha_i)$ is almost fixed. In the following calculations to investigate the temperature dependence of exchange bias and coercivity, we take $k_{\mathrm{B}}T_{\mathrm{D}}/K_{\mathrm{AFM}}S_0 d = 1.0$.

For various average sizes of AFM grains, the temperature relations of exchange bias and coercivity are shown in Figure 2. With increasing the sizes of AFM grains, both the exchange bias and coercivity increases at any temperature. In addition, the blocking temperature at which the exchange bias disappears also increases. Namely, there is size dependence for the blocking temperature. These results are in agreement with experiments

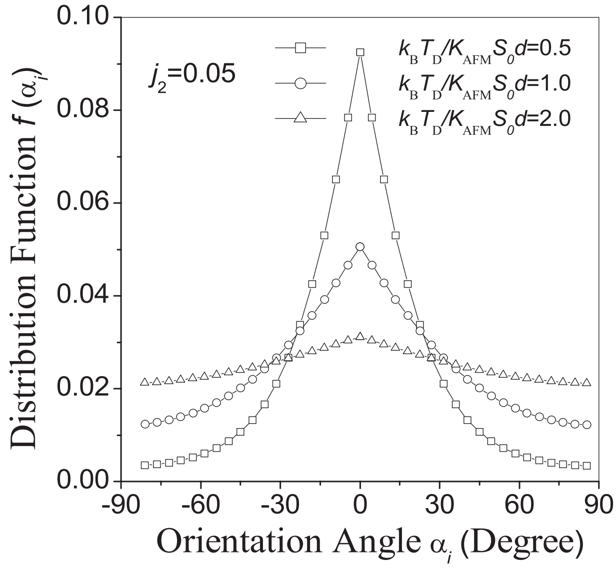

Fig. 1. Orientation distribution $f(\alpha_i)$ of AFM easy axes for different depositing temperatures.

qualitatively [36,37], and illustrate that the pinning effect of the AFM layer for the occurrence of exchange bias is of key importance. Without the biquadratic coupling the exchange bias always decreases monotonously with increasing temperature, which is in good agreement with experimental measurements. Also with increasing temperature, the coercivity firstly decreases distinctly, then increases and approaches a peak at around the blocking or Néel temperature, finally decreases again. However, when there is a biquadratic coupling at the interface between the FM and AFM layers, the curve shape of the temperature dependence of exchange bias becomes convex as shown in Figure 2b, while the coercivity increases at any temperature as shown in Figure 2d, and the blocking temperature increases as well. A possible explanation would be that the biquadratic coupling leads to increase the FM magnetization inhomogeneity, which is responsible for more effective pining in higher temperature. FM/AFM bilayers with large exchange bias and high blocking temperature are very applicable to design magnetic devices.

The calculated results of the temperature dependence of exchange bias and coercivity for various values of the bilinear coupling are shown in Figure 3. The coercivity gradually increases with increasing the bilinear coupling (see Fig. 3b), but the exchange bias increases more distinctly (see Fig. 3a). However, it is found that when $j_1 \geq 2.0$, the AFM layer will follow the reversal of the FM layer, and results in a nonbiased hysteresis loop and a large measurable coercivity, as pointed out by our previous result [30]. For small bilinear coupling, there are obvious peaks for exchange bias and coercivity if the biquadratic coupling plays a role. For the larger bilinear and special biquadratic couplings we can observe a large temperature region in which both the exchange bias and coercivity are almost unchangeable, which is also useful for the performance of magnetic devices. Moreover, as shown in the insets, we

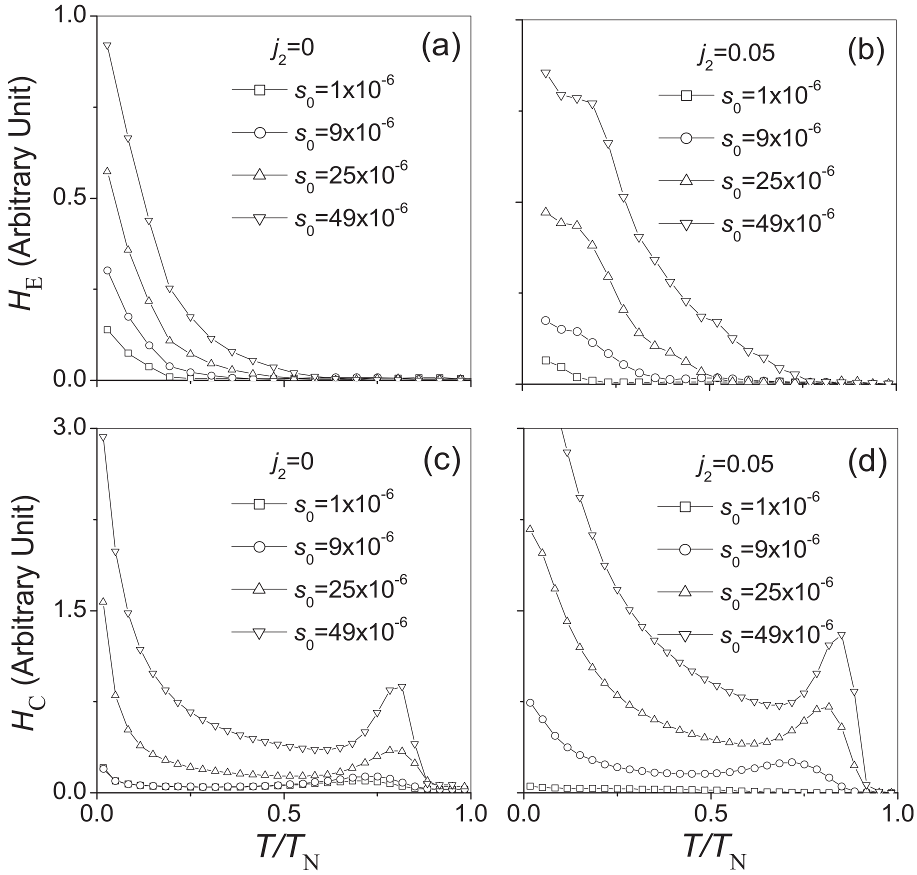

Fig. 2. Exchange bias (a), (b) and coercivity (c), (d) vs. temperature for the parameters $k_{FM} = 0.5$, $j_1 = 0.5$.

find that the consideration of the original orientation distribution of easy axes of AFM grains is useful to the occurrence of a plateau or broader peak in curves of coercivity or exchange bias vs. temperature, especially for larger bilinear coupling. This is a particularly interesting result compared with the previous work [34]. The results that there is a peak for coercivity near the Néel temperature and a narrow plateau for exchange bias in certain temperature range can be used to illustrate R. Jungblut's experiment qualitatively [12]. In fact, in R. Jungblut's case, the [111]-oriented samples display a biquadratic interlayer exchange coupling due to monoatomic steps at the interfaces [23,38].

In order to investigate the effect of the biquadratic coupling, we have also calculated the temperature relations of exchange bias and coercivity for different biquadratic coupling as shown in Figure 4. With increase of the biquadratic coupling, we find that the coercivity increases near zero temperatures but decreases at higher temperatures, while the exchange bias always decreases at any temperature. Interestingly, for certain temperature region as shown in Figure 4b, the coercivity almost unchanges while the exchange bias decreases with increasing biquadratic coupling, but there is a narrow plateau (see Fig. 4a). It suggests that the biquadratic coupling should be limited in order to obtain practicable exchange bias and lower coercivity.

Finally, it is interesting to note from our calculations that when the Curie temperature is not much greater than the Néel temperature, the peak of the coercivity always appear, no matter whether the biquadratic coupling is there or not. In this case, the FM magnetization will also depend on the temperature as the AFM magnetization.

4 Conclusion

We have presented a model and calculations to investigate the temperature dependence of exchange bias and coercivity that come from the exchange coupling at the interface between an FM layer and an AFM grain layer. In the present model, the FM layer is considered perfect with uniaxial anisotropy while the AFM layer consists of many independent AFM grains which are single-domain like. The interface coupling includes two contributions, the direct coupling and spin-flop coupling. The temperature dependence results from the thermal activated transition between the equilibrium states of AFM grains, and the temperature modulated relevant magnetic parameters. It is interesting and more practical that the original distribution $f(\alpha_i)$ of easy axes of AFM grains from sample preparation after field cooling has been considered. Meanwhile, a self-consistent calculation scheme is established, and the self-consistent numerical results show that $f(\alpha_i)$ is very

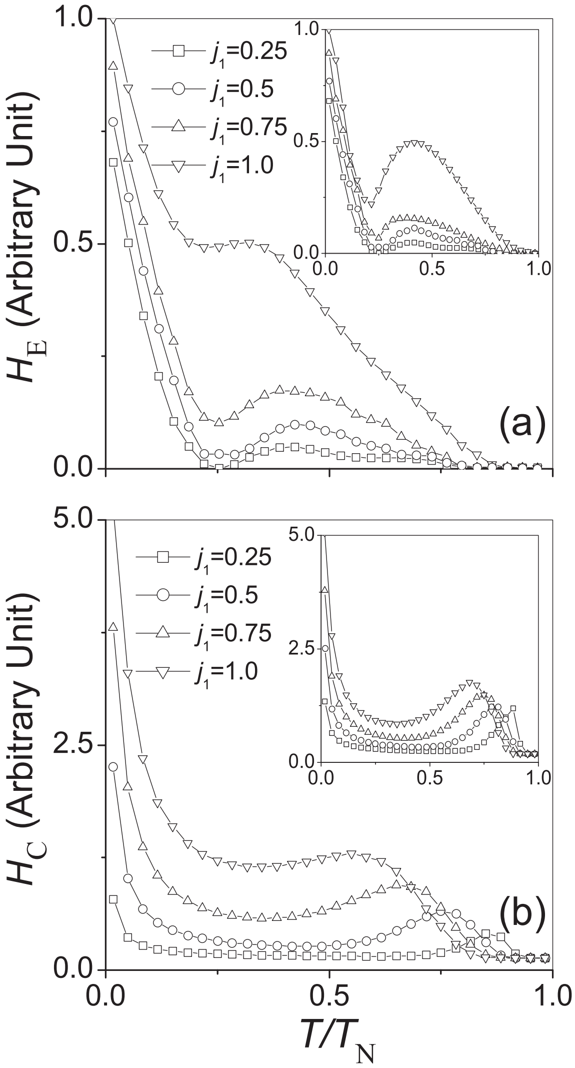

Fig. 3. Exchange bias (a) and coercivity (b) vs. temperature for the parameters $k_{\mathrm{FM}} = 0.5$, $s_0 = 16 \times 10^{-6}$, and $j_2 = 0.05$.

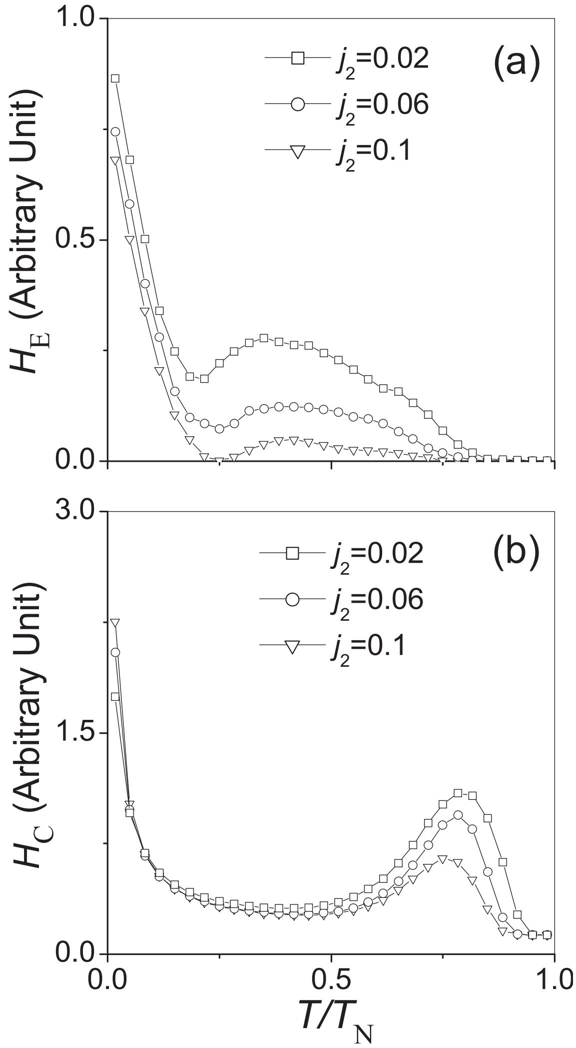

Fig. 4. Exchange bias (a) and coercivity (b) vs. temperature for the parameters $k_{\mathrm{FM}} = 0.5$, $s_0 = 16 \times 10^{-6}$, and $j_1 = 0.5$.

similar to the canonical distribution. In general, at low temperatures, the AFM state in each grain is stable as the FM magnetization rotates and results in higher exchange bias and coercivity. But with increasing temperature they decrease due to the fact that the AFM state becomes unstable. Moreover, in the case that the Curie temperature is much greater than the Néel temperature and there is a biquadratic coupling, the peak of the exchange bias and coercivity can appear simultaneously. Otherwise, the exchange bias always decreases with increasing temperature while the peak of coercivity still exists. These results are well confirmed by the experimentally observed peaks or upward inflections in the curves of the temperature dependence of both exchange bias and coercivity. For certain values of bilinear and biquadratic couplings, there are wide plateaus in which the exchange bias and coercivity are almost independent of temperature, and the orientations distribution of easy axes of AFM grains seems to be useful to induce the plateaus of exchange bias and coercivity to occur. This fact is useful for designing magnetic devices.

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