Mechanisms for the multistep spin reorientation of ultrathin Fe films on Gd

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The multistep spin reorientation transition of ultrathin iron films on bulk gadolinium is described theoretically. We find that it is necessary to include cubic terms in the magnetic anisotropy energy expansion in order to explain this phenomenon. Furthermore, the signs of the anisotropy coefficients required to explain the coexistence of both first- and second-order phase transitions in this spin reorientation transition are obtained. We can then model this system using either a surface or a bulk driven model, or a combination of both.

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I. INTRODUCTION

Phase transitions are a common phenomenon encountered in nearly every branch of physics.[1] Modeling the driving forces and the order of the transition often leads to a deeper understanding of the underlying physical processes involved. Magnetism, in particular, is a rich field in this regard due to the vector nature of the order parameter. Integral to this is the concept of magnetic anisotropy, i.e., the difference in energy for various orientations of the magnetization with respect to a sample. The anisotropy plays a role in every magnetic device,[2] and applications have allowed, for example, magneto-optical devices to surpass the diffraction limit of magneto-optical storage devices. These ''super-resolution'' devices rely on a temperature-driven spin-reorientation transition, where the magnetization of a thin film rotates from in-plane to perpendicular with increasing temperature.[3] An understanding of these spin reorientation transitions is an important source of knowledge regarding magnetic anisotropy. This information is invaluable because ab initio calculations in even the simplest systems are difficult,[4] and it is currently not feasible to predict, from first principles, the behavior of complicated alloys and multilayered systems.

For the most part, single magnetic films supported on nonmagnetic substrates have been investigated.[5,6] In these systems, the magnetization vector reorients from in the plane of the film to perpendicular as the temperature changes. This phenomenon can occur either as the temperature increases or decreases. For example, in Fe and Co films the magnetization rotates into the plane as the temperature ramps up,[7–9] while in films of Ni grown on Cu it rotates out of the plane.[10,11] All of these spin reorientation transitions are single step, continuous transitions, and can be understood using only the linear (K_1) and quadratic (K_2) terms of the anisotropy energy expansion for a single magnetic film. In addition, these films are thin enough that the entire film magnetization rotates uniformly across the thickness of the film.

However, devices are generally composed of many different magnetic layers that can have significantly different magnetic orientations and properties. These properties include the Curie temperature, higher order anisotropies, interlayer and intralayer exchange, and crystal structure. In order to begin understanding these systems, it is necessary to investigate the properties of more complicated, magnetic multilayer structures.

Recently, we reported experimental evidence of a multistep spin reorientation transition in a magnetic bilayer system consisting of an ultrathin (1.5 atomic layers) amorphous film of Fe on a thick, bulklike Gd(0001) film.[12] The first step in this unique spin reorientation transition is a continuous, reversible transition as the temperature increases from an in-plane orientation of Fe-rich surface magnetization into a slightly canted, out-of-plane state. The next step occurs at still higher temperature, and is a discontinuous, hysteretic transition of the surface magnetization to a nearly perpendicular, out-of-plane state. Both steps take place in the temperature interval from 260 to 280 K, i.e., below the Gd Curie point, $T_{C,\mathrm{Gd}}=292.5$ K. Subsequent magnetic studies using the polar magneto-optic Kerr effect,[13] have shown that the Gd layers near the interface participate in the spin reorientation transition. However, it was not possible to determine to what extent these interfacial layers are perturbed from the data at hand.

The goals of the present work are (1) to reveal the physical mechanisms underlying these two phase transitions, (2) to find the area in parameter space (in terms of the magnetic properties of both films) required for both a first order and second order phase transition to occur, and (3) to understand and model the extent to which the Gd interfacial layers participate in the spin reorientation transition. The approach used is based on the accepted viewpoint that magnetic spin reorientations are physical realizations of phase transitions as described by Landau theory.[14–16] We take the perpendicular component of the surface layer vector magnetization as an order parameter and model the behavior of the entire system as the temperature changes, taking into account the layer dependent magnetization properties.

II. FIRST STEP—CONTINUOUS PHASE TRANSITION FROM IN-PLANE TO CANTED

Here we discuss the physical mechanism that gives rise to the continuous, low temperature transition from the in-plane to canted magnetization state of the Fe-rich surface. This

transition is characterized by the appearance of long-range order perpendicular to the plane of the film, and is accompanied by a reversible peak in the susceptibility.[12,13] It can therefore be considered a second-order phase transition into a state with a perpendicular magnetization component. In order to describe this behavior it is necessary to use a multilayer approach. This is due to the fact that there must be a non-uniform magnetization profile established (i.e., a domain wall) in the interfacial region between the canted surface magnetization and the in-plane magnetized bulk Gd.

The criterion for the onset of the continuous spin reorientation phase transition in this type of system was derived in our recent work (Ref. 17), in terms of the reduced surface and subsurface anisotropy constants and the exchange interaction in the surface layer. These are defined as k_S, k_B, and γ, respectively [see Ref. 17, Sec. VI, Eqs. (42) and (43)], and the general solution to this problem is given in closed form for an arbitrarily large number of magnetic layers in the surface region.

This evolution of the surface magnetization into a canted state can be understood intuitively by considering that the orientation of the surface moment is affected by two factors: the anisotropy energy of the Fe-rich surface (that favors the perpendicular orientation) and the exchange energy between the Fe-rich surface and the in-plane magnetized bulk Gd. The experimental data imply that at low temperature, the exchange interaction between the bulk and the surface is stronger than the surface anisotropy energy. To understand the continuous reorientation of the surface magnetization from in-plane to canted as the temperature increases, one must realize that the bulk magnetization falls off faster than the surface because Gd has a lower Curie temperature than the Fe-rich surface. Therefore, at some point the exchange interaction between surface and bulk becomes weaker than the surface anisotropy, and the surface will begin to cant out of plane.

In the absence of the second discontinuous step observed in Ref. 12, this analysis would be sufficient to describe the Fe spin reorientation transition, and would allow us to determine the reduced magnetic constants, as discussed in Ref. 17. This would result in a simple model that includes only the linear term in the perpendicular anisotropy and the exchange energy between surface and bulk. However, the appearance of the second step at higher temperature means that, while the criterion for the onset of this transition still holds, a more complicated model that includes higher order terms in the anisotropy energy expansion is necessary.

III. SECOND STEP—DISCONTINUOUS PHASE TRANSITION OF THE SURFACE FROM CANTED TO PERPENDICULAR

The second step of the reorientation transition is characterized by a discontinuous jump of the Fe-rich surface magnetization from canted to perpendicular, and can therefore be classified as a first-order phase transition. This spin reorientation transition is hysteretic with temperature cycling, and occurs slightly below the bulk Gd Curie temperature. Another important aspect of this transition is that as the temperature is ramped up through the transition, there is a pronounced maximum in both the perpendicular component of the electron spin polarization and the polar magneto-optic Kerr effect (MOKE).[12,13] The observation that this effect is stronger in the MOKE data shows that it originates in the interfacial Gd layers, and indicates that they are participating in spin canting-along with the Fe-rich surface layer.

The fact that there is a discontinuous, hysteretic transition of the magnetization direction means that the energy of the system must have two minima as a function of the angle of the film magnetization. This requirement can only be met by including up to at least cubic terms in the anisotropy energy expansion. Just as important, however, is the fact that the values of these anisotropy coefficients and the exchange terms must meet specific requirements in order to obtain a coexistence of both first- and second-order phase transitions over the course of the spin reorientation transition. These higher-order terms may originate either at the surface or in the bulk. Consideration of these possibilities results in the two models discussed below: (1) a surface driven model, with the higher-order terms originating solely in the Fe over layer; and (2) a bulk driven model with the higher order terms originating solely in the Gd. We explore both of these possibilities in Secs. III A and III B. In both models we develop a full multilayer approach, where the Fe-rich surface is the first layer and the underlying Gd film is divided into many different layers. The response of the system in these two models is calculated for both electron spin polarization and MOKE measurements.

A. Surface driven model of the discontinuous transition

This model assumes that the discontinuity of the magnetization vs. temperature originates from the intrinsic properties of the Fe-rich surface layer. In Sec. III A 1 we illustrate the nature of the discontinuous step within a magnetic bilayer approach (i.e., surface and bulk), and demonstrate that it is not possible to describe the multistep spin reorientation transition without accounting for at least cubic terms in the anisotropy energy expansion.

A multi-layer approach is then applied (Sec. III A 2), and solved numerically to find the computer simulated magnetization profile in the Gd film. We find that the simulated MOKE and surface magnetization data match well with the experimental data.

1. Surface driven model—bilayer approach using surface anisotropy vs bulk exchange

In the surface driven model, the thermodynamic potential of the Fe/Gd system may be written as a series expansion in the magnetization angle, $\cos^2\theta$, ($\theta=\theta_1-\pi$):

$$\Phi_S = K_{1,S}, M_S^2\cos^2\theta + K_{2,S}M_S^4\cos^4\theta + K_{3,S}M_S^6\cos^6\theta + J_{SB}M_S M_B\cos\theta. \quad (1)$$

Here, the coefficients $K_{n,S}$ are the surface anisotropy constants and J_{SB} is the exchange interaction between the surface and the bulk magnetizations, M_S and M_B, respectively. This is depicted in Fig. 1(a). Note that the figure is drawn to

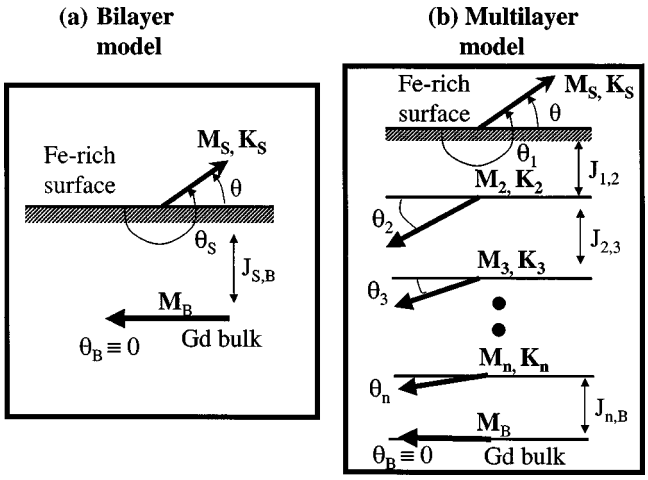

FIG. 1. Diagrams of the two approximations used for modeling the behavior of the Fe/Gd magnetic multilayer system. Panel (a) shows the magnetic bilayer approach, and panel (b) shows the multilayer approach, where the Gd is allowed to break up into discrete magnetic layers. This simulates the formation of a domain wall in the Gd layers between the canted FeGd surface layer and the in-plane magnetized Gd bulk.

show that the Fe is antiferromagnetically aligned to the bulk (i.e., $J_{SB}<0$), as occurs in the actual system. This does not change the energy minimization of the system, but it is important later for calculating the MOKE signal.

To illustrate the full multi-step spin reorientation with temperature we introduce the parameter $m \equiv M_B(T)/M_B(0)$, the reduced magnetic moment of the bulk Gd. The *decrease* in m from one to zero corresponds to an *increase* in temperature from zero to the Gd Curie point $T_{C,\mathrm{Gd}} \equiv T_{CB}$. Normalizing Φ_S with the positive, temperature independent coefficient $-J_{SB}M_S M_B(0)$ gives the formula for the reduced thermodynamic potential φ:

$$\varphi = \frac{\Phi_S}{-J_{SB}M_S M_B(0)} = \alpha\cos^2\theta + \beta\cos^4\theta + \gamma\cos^6\theta - m\cos\theta. \qquad (2)$$

The minimization of φ with respect to the angle θ gives the following solutions: the in-plane state ($\theta=0$), the perpendicular state ($\theta=\pi/2$) and the canted state ($0<\theta<\pi/2$) with θ determined by

$$m = 2\alpha\cos\theta + 4\beta\cos^3\theta + 6\gamma\cos^5\theta. \qquad (3)$$

To obtain a double well of the anisotropy energy with a global minimum at $\cos^2\theta=0$ and a local minimum at some intermediate angle between zero and $\pi/2$, one must investigate Eq. (3) for all different signs of the reduced anisotropy constants α, β, and γ. We find that the case with $\alpha\neq 0$, $\beta=\gamma=0$ cannot give rise to a first-order transition. On the other hand, it can be shown that the case $\alpha>0$ and $\beta<0$, $\gamma=0$, gives rise to a first-order transition for $m\neq 0$ only if the additional requirement $1/\sqrt{2}<-\alpha/2\beta<\sqrt{3}$ is satisfied; otherwise the transition is second order. The plot of $m(\cos\theta)$ is presented in Fig. 2. Arrows show the cooling and heating

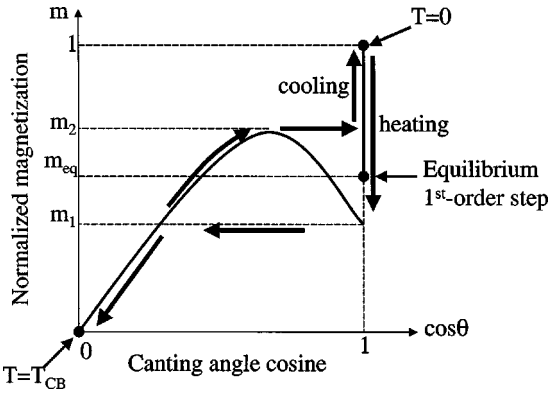

FIG. 2. Evolution of the surface Fe-Gd magnetization orientation ($\cos\theta$) with reduced Gd magnetization, m, using only linear, K_1, and quadratic, K_2, anisotropy constants.

cycles and confine the area of the maximal hysteresis loop. This loop simulates the magnetization angle jumping up discontinuously at m_1 as the sample is heated, and back down at m_2 during cooling. For a computer simulation, however, we will observe the jumplike behavior at some intermediate magnetization, denoted in Fig. 2 as m_{eq}. This is because the simulation will always find the lowest energy equilibrium angle rather than the intermediate metastable ones.

However, the problem with this solution is that the requirements for the continuous transition, as described in Sec. II and Ref. 17, cannot be met for this range of parameters. Therefore, in the absence of a cubic term in the energy expansion ($K_{3S}, \gamma=0$), it is possible to obtain *either* a first- or second-order transition *but not both*. Consideration of canted states in an arbitrarily large number of atomic Gd layers does not change this result because at each layer the same considerations apply.

With further analysis, it can be shown that only the case $\alpha>0$, $\beta<0$, and $\gamma>0$ allows for both first- and second-order steps in the spin reorientation transition. The plot of m vs $\cos\theta$ (solid line) for this case is presented in Fig. 3, where

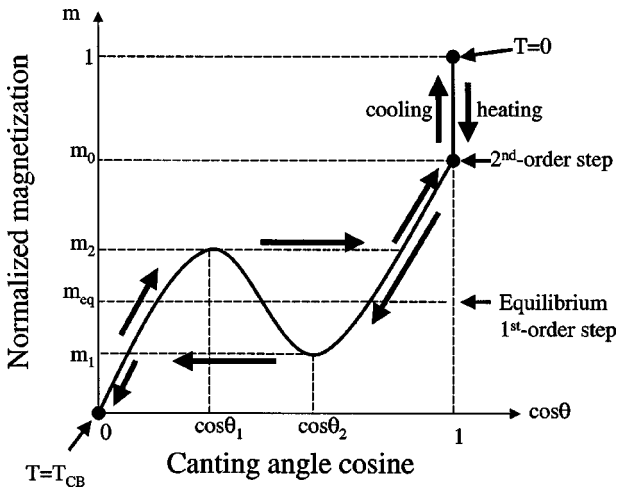

FIG. 3. The evolution of the surface Fe-Gd layer magnetization orientation ($\cos\theta$) with reduced Gd magnetization, m, using linear, K_1, quadratic, K_2, and cubic, K_3, anisotropy constants.

$m=m_0$ and $m=m_{eq}$ correspond to the second and the first-order equilibrium steps, respectively. Arrows show the cooling and heating cycles and define the boundary of the maximal hysteresis loop. The spin reorientation is essentially multistepped, with the second-order transition preceding the first-order one only if the requirement $m_0 \equiv 2\alpha+4\beta+6\gamma > m_{eq}$ is satisfied. Therefore, within the surface driven model, the description of the multistep spin reorientation transition is possible only if $K_{1S}>0$, $K_{2S}<0$, and $K_{3S}>0$ and they satisfy further stringent conditions for their relative magnitudes.[18]

2. Surface driven model-multilayer approach and simulation

In order to describe the MOKE data, it is necessary to account for the deviation of the subsurface Gd atomic layers from the in-plane direction. Therefore, within the first model the thermodynamic potential of the system is given by

$$\Phi = K_{1S}M_S^2\cos^2\theta_1 + K_{2S}M_S^4\cos^4\theta_1 + K_{3S}M_S^6\cos^6\theta_1 - J_{12}M_S M_B\cos(\theta_1-\theta_2) + \sum_{n=2}^{\infty}[-J_{n,n+1}\times\cos(\theta_n-\theta_{n+1}) + K_{1B}\cos^2\theta_n]M_B^2. \quad (4)$$

Here $J_{12}<0$ is the exchange interaction between the surface and subsurface atomic layers, $J_{n,n+1}$ $(n>1)$ is the Gd-Gd interlayer exchange interaction, K_{1B} is the Gd layer anisotropy constant, $K_{1B}<0$, and the θ_n are the orientation angles for surface $(n=1)$ and bulk atomic layers $(n=2,3,....)$, as shown in Fig. 1(b). The minimization of Eq. (4) results in a dependence of the angles θ_n on the layer index and temperature via the temperature dependence of the Gd magnetization, $M_{Gd} \equiv M_B$. The surface magnetization, M_S, was assumed to be temperature independent in the temperature interval 270–290 K.

Equation (4) was solved using an iterative computer program to find the minimum energy for any given combination of anisotropy and exchange parameters. With this program we confirmed that the results of the bilayer approach discussed in Sec. III A 1 hold for the multi-layer approach. We were then able to simulate the temperature dependence of both the SPSEES and MOKE signals. These results are presented in Fig. 4.

For the SPSEES signal, we plot the magnetization direction of the topmost surface layer. The MOKE signal, on the other hand, is simulated by summing the contributions of each layer with an exponentially decreasing contribution as a function of depth. This simulation incorporates the fact that Fe and Gd give opposite rotations of light. These contributions therefore add because the Fe and Gd are antiferromagnetically coupled. We find that the best fit to the data occurs when the exponential attenuation constant is comparable to the depth of the magnetization profile. This is consistent with the fact that the domain wall width and the exponential attenuation of light in Gd are of similar magnitude.

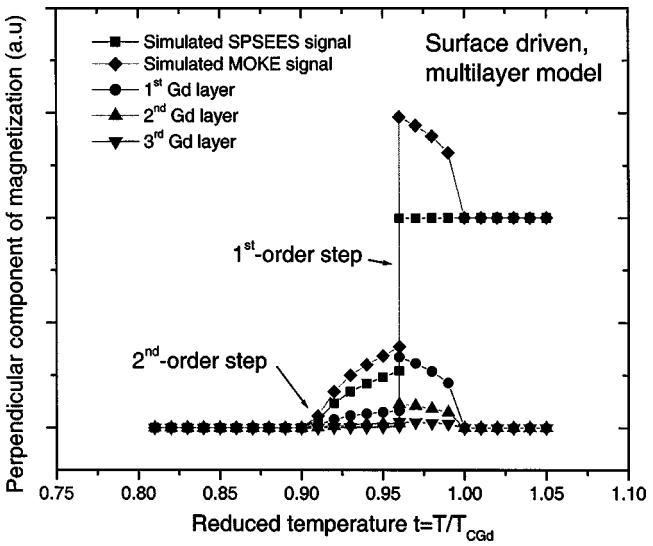

FIG. 4. Simulation of the surface driven model for the two-step spin reorientation transition of Fe/Gd.

B. Bulk driven model of the discontinuous phase transition

In this section we take the opposite approach to Sec. III A, and assume that the discontinuous transition is driven only by higher order anisotropies that exist in the Gd bulk. We first describe the physical mechanism within a simplified bilayer approach, Sec. III B 1, and then extend to a full multilayer approach to simulate the experimental data in Sec. III B 2.

1. Bulk driven model—bilayer approach using surface anisotropy vs bulk exchange and anisotropy

The bulk driven model assumes that the discontinuous transition originates from the intrinsic properties of the Gd interfacial layers. Due to the predominant role of the shape anisotropy, the well-known second-order spin reorientation of the bulk Gd (Refs. 19 and 20) is suppressed due to the shape anisotropy of the samples.[21] The in-plane anisotropy energy of Gd therefore decreases to zero at the Curie point of Gd. On the other hand, the perpendicular Fe-Gd surface anisotropy energy is almost constant because it's magnetization does not change significantly in this range. This change in the balance between these two energies leads to a transition into the perpendicular state of the surface as the temperature is ramped up. To describe the first-order step, it is necessary to assume that the dependence of the Gd subsurface anisotropy energy is a non-monotonic function with a local minimum. The Gd anisotropy energy is expanded in $\cos^2\theta$, and the total energy is given by the equation

$$\Phi = -J_{SB}M_S M_B\cos(\theta) + K_S M_S^2\cos^2\theta + K_{1B}M_B^2\cos^2\theta + K_{2B}M_B^4\cos^4\theta + K_{3B}M_B^6\cos^6\theta. \quad (5)$$

The analysis of various cases corresponding to all possible signs of the bulk anisotropy constants K_{1B}, K_{2B}, and K_{3B} in Eq. (5) is similar to that for the first model. We find that the multi-step spin reorientation with both first- and second-order steps is possible only in the case where K_S, $K_{3B}\neq 0$,

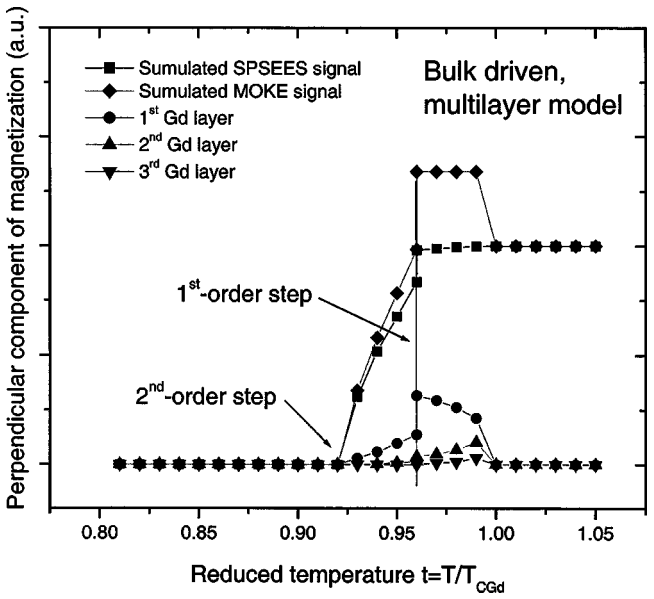

FIG. 5. Simulation of the bulk driven model for the two-step spin reorientation transition of Fe/Gd.

$K_{1B}<0$, $K_{2B}>0$, and $K_{3B}<0$, with restrictions on their relative magnitudes analogous to those described in Sec. III A 1.

2. Bulk driven model—multilayer approach and simulation

In order to describe the MOKE data, again we consider the deviation of the magnetization from the in-plane orientation in each Gd atomic layer. Within this second model the thermodynamic potential is given by

$$\Phi = -J_{12}M_S M_B \cos(\theta_1-\theta_2)+K_{1S}M_S^2\cos^2\theta_1 + \sum_{n=2}^{\infty}[-J_{n,n+1}M_B^2\cos(\theta_n-\theta_{n+1})+K_{1B}M_B^2\cos^2\theta_n + K_{2B}M_B^4\cos^4\theta_n+K_{3B}M_B^6\cos^6\theta_n]. \quad (6)$$

The minimization of Φ in Eq. (6) results in the dependence of angles θ_n on the layer index and on temperature via the temperature dependence of $M_{\mathrm{Gd}}\equiv M_B$, with M_S temperature independent.

Again, an iterative computer simulation of the magnetization profile and resulting temperature dependent surface and MOKE data were calculated in the same manner as Sec. III A. The results are presented in Fig. 5.

IV. DISCUSSION

The multi-step spin reorientation discussed here for the Fe/Gd system has not been observed in any other thin film or multi-layered system. We find that the explanation of the first order transition necessitates a nonlinear dependence of the anisotropy energy on the magnetization orientation. In addition, the parameter space available for the coexistence of both a first- and second-order phase transition in this system is limited.

In both models, it is necessary to have a surface that favors perpendicular magnetization, i.e., $K_{1S}>0$. This effect is to be expected because, according to experimental data available, both Fe-Gd (Ref. 22) and pure Fe (Refs. 7–9) films exhibit a magnetic anisotropy that favors a magnetization perpendicular to the plane of the film.

For the surface driven model, however, K_{2S} and K_{3S} have never been experimentally determined.[2] Existence of these terms is not necessarily expected because the long-range order of the Gd(0001) surface was reported to be broken by the deposition of the Fe over layer.[12] However, the sixth-order symmetry, C_{6v}, in the first few subsurface Gd layers might affect the surface magnetic anisotropy energy as well as the local structural symmetry seen by the individual Fe atoms. This effect could, therefore, contribute a cubic term to the Fe-Gd anisotropy energy expression.

For the bulk driven model, on the other hand, values for K_{1B} and K_{2B} have been reported from experiments on bulk Gd.[19,20] At low temperature, the sign of K_{1B} agrees with our model, while in the vicinity of the Gd Curie point it was reported to become slightly positive. Due to the negative contribution of the shape anisotropy of the Gd film substrate, however, K_{1B} is not expected to change sign in this experiment. On the other hand, the values for K_{3B} reported in the literature are positive and small. In-depth analysis of the data available brings us to the conclusion that K_{3B} is, in fact, not well established even for bulk Gd samples. Also, it is likely that the anisotropy constants of the Gd layers in the surface region are affected by the Fe overlayer, and therefore differ from the bulk values.

The comparison of our MOKE and surface magnetization computer simulations, presented in Figs. 4 and 5, shows that they are very similar. Both models demonstrate the two-step spin reorientation; the first step is a second-order transition from the in-plane state to a canted, out-of-plane state, and the next step is a first-order transition from the canted state to a nearly perpendicular state. The bump in the simulated MOKE signal just above the first order step matches well in shape and magnitude with experimental data. This supports the conclusion made in Ref. 13, and shows that the Gd interfacial layers take part in the formation of a canted magnetic structure. The main difference between Figs. 4 and 5 is rather subtle, in that the change in the surface *z*-component on the first-order step is larger within the first model.

Such uncertainty in the models for describing a spin reorientation transition in exchange coupled magnetic multilayers is not unique. Because most experiments register only the fact that a transition occurs, rather than directly measuring the anisotropy in each layer, it is often impossible to identify which layer is driving it. The exchange coupled layers in the super-resolution magneto-optical devices are a good example.[23] These devices are based on rare-earth and transition metals with competing in-plane and perpendicular anisotropy. Two possible mechanisms were found for that spin reorientation transition. The first mechanism is an intrinsic spin reorientation in the GdFeCo layer, and the second is the exchange coupling force between the perpendicularly magnetized TbFeCo and the in-plane GdFeCo films. These mechanisms are can be explained using only linear terms in the anisotropy and standard spin reorientation models.

In contrast, our analysis of the order of the transitions in the Fe/Gd system demonstrates that this spin reorientation is fundamentally different. This is because it relies on a nonlinearity in the anisotropy at or near the surface. The data available related to the first two anisotropy constants K_1 and K_2, for Fe, Gd, and Fe-Gd films required by each model appear to be of the appropriate sign, but a careful measurement of their magnitudes is needed to compare to simulations. However, the most important insight into the mechanism driving this two-step transition would be a experimental determination of the sign and magnitude of the cubic anisotropy constant K_3 in the bulk and at the interface for Gd and Fe-Gd thin films.

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