Field Induced Biquadratic Exchange in Hard/Soft Ferromagnetic Bilayers

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The appearance of biquadratic exchange coupling between soft Fe and hard SmCo thin layers is found. The remanent magnetization in the Fe layer reorients from parallel to perpendicular with respect to the SmCo easy axis after application of large enough negative field. To explain such an unexpected behavior in contacting ferromagnetic layers a model is proposed based on Slonczewski's fluctuating exchange mechanism. In our samples a partial remagnetization of the hard layer creates fluctuations of the bilinear interactions. The intralayer exchange averaging of the resulting magnetization fluctuations in the soft layer causes the observed biquadratic coupling.

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Exchange interactions between the layers is the dominating factor determining the physical properties of magnetic thin film nanostructures [1,2]. Depending on the magnetic ordering in the layers and the nature of a spacer between them, these interactions can vary from ferromagnetic to antiferromagnetic and in some cases generate structures with perpendicular orientation of spins. Such a perpendicular coupling of magnetic moments, first observed in Fe/Cr/Fe trilayers with varying Cr thickness [3,4] and Co/Cu/Co structures [5], is usually associated with coupling of ferromagnetic (FM) films through a nonmagnetic or antiferromagnetic (AFM) layer of corresponding thickness. It is described phenomenologically by the second order term in the exchange Hamiltonian $H_2 = -j(\mathbf{M}_1 \cdot \mathbf{M}_2)^2$, known as the biquadratic exchange [1,2,6]. Here $\mathbf{M}_1$ and $\mathbf{M}_2$ are magnetic moments in the layers so that the negative exchange constant j will favor the perpendicular coupling of spins. In principle, the biquadratic coupling is allowed by the symmetry of the microscopic Hamiltonian for pairs of spins but it is usually much smaller than the conventional Heisenberg exchange $H_1 = -J\mathbf{M}_1 \cdot \mathbf{M}_2$ (see, e.g., [7]). Under the conditions of reduced bilinear exchange the biquadratic term could become dominating. However, there is no experimental proof of such a situation in any homogeneous magnetic system. For magnetic layers separated by a nonmagnetic spacer the biquadratic exchange can appear due to different reasons [8]. There are intrinsic mechanisms caused by the interference of electrons on the spacer thickness but they result in a small biquadratic term $j \ll J$. A larger term appears due to extrinsic mechanisms associated with inhomogeneities of the spacer: thickness variations, loose spins (magnetic impurities in the spacer matrix), and proximity magnetism [1,8]. Slonczewski showed [9] that in FM/AFM/FM trilayers a biquadratic energy contribution can appear due to spatial fluctuations of the bilinear interlayer coupling J which induce magnetization fluctuations that relax by *intralayer* exchange. The sign of this energy contribution is always positive (j is negative) for any distribution of J.

In the present paper we report on the observation of an unusual perpendicular coupling between two ferromagnetic layers in direct contact. Such a junction is conventionally believed to have only ferromagnetic (parallel) arrangement of magnetic moments across the interface. However, as shown below the magnetic inhomogeneity of the hard layer which arises during its partial remagnetization can induce the fluctuating magnetization in the soft layer and result in the observed noncollinear coupling. The mechanism is similar to that suggested by Slonczewski and the formulas of [9] are relevant even though the physical reason for fluctuations is different.

We studied junctions of $SmCo_7$ (hard) and Fe (soft) layers. The structures (with top Fe layer) were grown by dc magnetron sputtering onto MgO substrates coated with an epitaxial Cr buffer layer (see [10,11] for details). The results presented here are obtained on a bilayer of Fe(20 nm)/SmCo(20 nm) on MgO(110) with uniaxial magnetic anisotropy. Its easy magnetization axis is along the in-plane **c** axis of SmCo. 100 μm holes were made at several locations in the samples using optical lithography and Ar ion etching. Fringing fields at the hole edges allow the orientation of magnetic moments to be monitored as discussed below. The macroscopic easy-axis magnetization loops were measured by SQUID magnetometry. They show a typical double kink appearance with one kink at the exchange bias field H_E and another one at the remagnetization field of the hard layer (see Fig. 2b). Cycling around H_E at moderate field amplitudes reveals small coercivity and an unchanged remanent magnetization. A high resolution magneto-optical (MO) imaging technique [12] was used to directly image the evolution of magnetic patterns during remagnetization.

Hard/soft ferromagnetic bilayers are usually referred to as *exchange springs*. With increasing external field $H > H_E$ opposite to their initial polarization the magnetization of the soft layer twists along its thickness (as a spring) and then untwists under decreasing field due to the "elastic" intralayer exchange torques. At the interface the spin orientation of the soft layer is captured by the interlayer

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exchange coupling with immobile spins of the hard layer. The MO contrast at the edges of the etched holes where in-plane magnetic moments produce fringing fields reveals the moment orientation in the soft Fe layer [10]. Figure 1a illustrates the initial state of the uniaxial sample after reducing H from 7 T to 0. Here both, $\mathbf{M}_{\mathrm{Fe}}$ and $\mathbf{M}_{\mathrm{SmCo}}$, follow the same easy axis parallel to the direction of the polarizing field. The MO pattern forming a half dark/half bright circle (dark and bright reveal up and down directions of perpendicular components of the fringing field) corresponds to the orientation of $\mathbf{M}_{\mathrm{Fe}}$ shown by arrows.

Upon application of a negative field, this contrast starts to rotate when the field value exceeds $H_E \sim 700$ Oe. The magnetization rotates gradually with increasing H towards the applied field direction (Figs. 1b–1e). If the maximum negative field is not very large, the magnetization in the Fe layer springs back upon reduction of H, and at remanence the initial homogeneous polarization of the sample is restored (Figs. 1f–1h).

The field dependence of the magnetization angle for $\mathbf{M}_{\mathrm{Fe}}$ is shown in Fig. 2a. It corresponds to the macroscopic magnetization curve for -1.7 kOe presented in Fig. 2b confirming that the magnetization is mainly due to the rotation of $\mathbf{M}_{\mathrm{Fe}}$.

Unexpectedly, when the maximum negative field exceeds ~ 2 kOe, the subsequent remanent state ($H = 0$) reveals a distinctive change in the average magnetic moment direction of $\mathbf{M}_{\mathrm{Fe}}$ with respect to the initial polarization. This unusual behavior indicates the appearance of the biquadratic coupling $\sim(\mathbf{M}_1 \cdot \mathbf{M}_2)^2$ favoring perpendicular orientation of magnetic moments in the hard and soft layers.

Figures 3a–3h present images of remanent states after application of different negative magnetic fields H showing the increase of the remanent $\mathbf{M}_{\mathrm{Fe}}$ angle with H. This increase indicates the growing contribution of the biquadratic coupling. Note also that there are small-scale magnetic inhomogeneities observed as local variations of the contrast in the film around the hole. To explain the observed rotation of the average remanent magnetic moment of the Fe layer we suggest the appearance of an effective biquadratic exchange due to the inhomogeneous state of the partially remagnetized hard layer (see Fig. 3i). Because of the large uniaxial anisotropy of SmCo and because of the presence of grains one can expect that at high enough negative fields the SmCo magnetization starts inverting in small domains that are randomly scattered in the initially polarized matrix. Initial remagnetization is thought to occur in domains $\sim$50 nm in size (this is the grain diameter in the SmCo film [11]). These domains remain inverted when

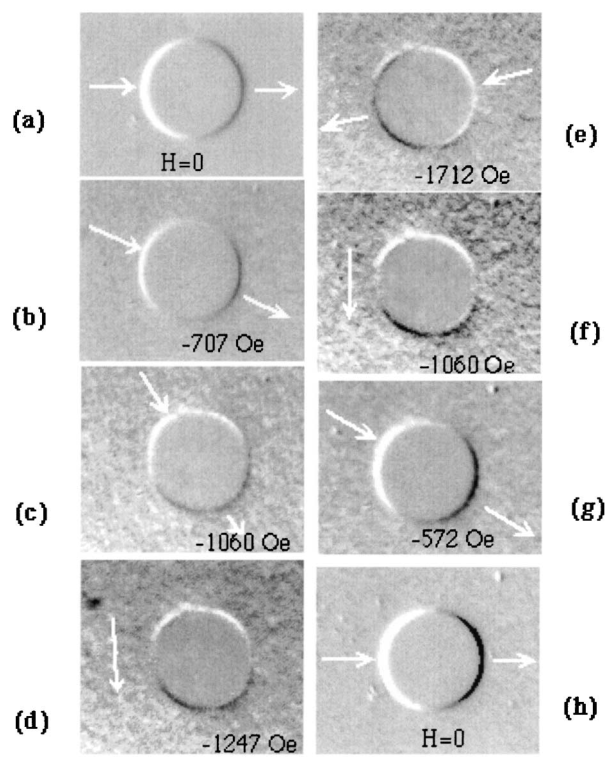

FIG. 1. Magneto-optical patterns around a hole during application of a negative field (H values shown on the pictures) to the SmCo/Fe structure polarized horizontally by $+7$ T.

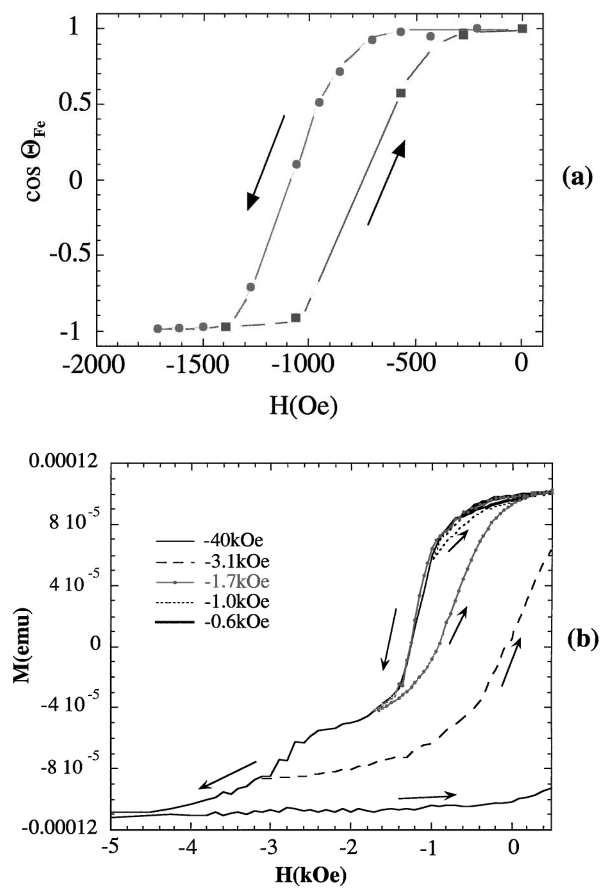

FIG. 2. (a) Field dependence of the cosine of angle between $\mathbf{M}_{\mathrm{Fe}}$ and the initial polarization direction measured during application of the field. (b) Macroscopic magnetization loops for different maximum negative fields. The remagnetization of Fe starts at $\sim H_E \sim -700$ Oe and the remagnetization of SmCo sets in below ~ -2.6 kOe.

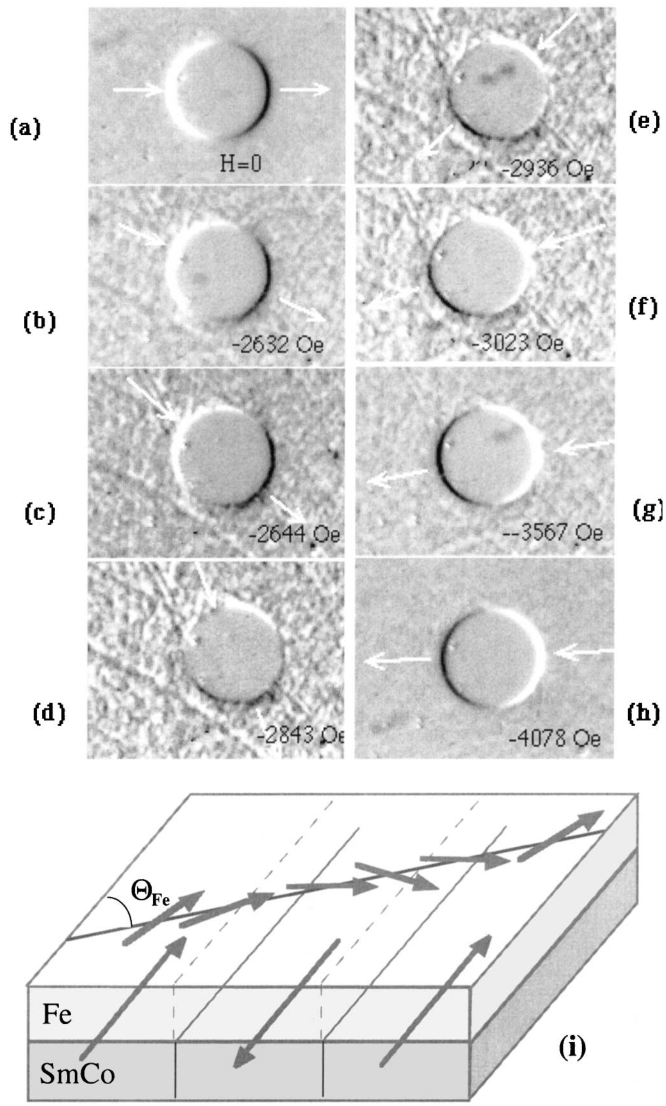

FIG. 3. (a)–(h) Remanent state after application of negative fields shown on the pictures. (i) Schematic of magnetization distributions in Fe and SmCo layers at the partial remagnetization of SmCo. Oscillations of $\mathbf{M}_{\mathrm{Fe}}$ around the average angle, Θ_{Fe} are shown.

the applied field is subsequently switched off. The inversion of **M** in these areas should cause oscillations of magnetic moments in the Fe film. These oscillations of $\mathbf{M}_{\mathrm{Fe}}$ around some average direction result in an increase of the exchange energy inside the Fe layer. The competition of this Fe *intralayer* exchange energy and the *interlayer* exchange energy arising from magnetization oscillations produces the biquadratic contribution and results in a net transverse moment in the Fe layer. For the compensated domain state in SmCo there will be essentially perpendicular average $\mathbf{M}_{\mathrm{Fe}}$ giving a zero remanence on longitudinal M(H) loops (Fig. 2b).

We adopt here a model by Slonczewski [9], who suggested an elegant explanation of the biquadratic exchange in a trilayer of Fe films separated by a Cr spacer with a stepwise modulated thickness. Steps in the thickness cause modulations $\pm\Delta J_1$ of the bilinear exchange between the magnetic layers to produce oscillations of the magnetic moments in the films (they tend to align for $+\Delta J_1$ and antialign for $-\Delta J_1$). The resulting contribution to the total exchange energy has a biquadratic shape $\sim \sin^2\theta$, where θ is the angle between the average magnetic moments in the layers. The coefficient of the biquadratic exchange is $J_2 = -[(\Delta J_1)^2 L/\pi^3] \cdot \Sigma\{A_1^{-1} \coth[\pi(2m-1)D_1/L] + A_2^{-1} \times \coth[\pi(2m-1)D_2/L]\}/(2m-1)^3 (m = 1 \text{ to } \infty)$ [9]. In this formula L is the width of the steps, A_i is the intralayer exchange stiffness, and D_i is the thickness of the ferromagnetic layers. J_2 must be multiplied by a function $N(f)$ describing the competition between opposite coupling in domains corresponding to $\pm\Delta J_1$. Here f is the fraction of domains with positive or negative coupling and $N(f)$ is maximum near $f \sim 0.5$ and decays to 0 at $f = 0$ and 1 [9]. Thus the biquadratic coupling should be most efficient when the mesas and the background ($+\Delta J_1$ and $-\Delta J_1$ regions) have the same area and disappear when the size of mesas (or background) is small.

A simple physical picture of this mechanism was presented in [13] through the effect of exchange averaging. In general, one can suggest that spatial variations of coupling energy of any nature ΔE (exchange, magnetostatic, or magnetoelastic) will produce oscillating fields $H_M \sim \Delta E/M$ which after averaging by the intralayer exchange fields $H_{\mathrm{ex}} \sim Ak^2/M$ ($k = \pi/L$ and $2L$ is a characteristic period of oscillations) will give the biquadratic contribution $\sim(\Delta E) \cdot (H_M/H_{\mathrm{ex}}) \sim (\Delta E)^2 L^2/A$. Such a term is always positive and increases with the scale of oscillations L. Therefore, any inhomogeneity should assist formation of the noncollinear coupling.

In our case the reason for the exchange modulations is the inversion of the local SmCo spins rather than the value of J_1. However, the mechanism of the biquadratic coupling through the exchange averaging of magnetization oscillations in the soft layer remains similar. As the maximum negative field increases, the number and size of inverted domains in the SmCo increases. So the biquadratic exchange should increase ($\mathbf{M}_{\mathrm{Fe}}$ tilts more from the easy axis of SmCo) until the inverted area becomes dominant ($f > 1/2$) and the biquadratic coupling starts to decrease again ($\mathbf{M}_{\mathrm{Fe}}$ turns toward the easy axis of SmCo but opposite to the initial polarization). By characterizing a given state of SmCo by the relative fraction of inverted area f, it is possible to estimate the average angle of $\mathbf{M}_{\mathrm{Fe}}$. In the total energy expression one has to account for the biquadratic term as well as contributions from the bilinear exchange and anisotropy of the Fe film. The bilinear term should be multiplied by the difference of positively and negatively magnetized areas $1-2f$. In turn, the biquadratic coupling enters with Slonczewski's coefficient $N(f)$. So the total energy per unit area of the Fe film in zero field becomes

$$E_{\mathrm{tot}} = -(1 - 2f)J_1 \cos\Theta - N(f)J_2 \sin^2\Theta + K \sin^2\Theta .$$

Here Θ is the average angle of $\mathbf{M}_{\mathrm{Fe}}$ with respect to the easy axis coinciding with the initial polarization. The interfacial bilinear exchange constant is $J_1 = kA_{\rm int}/d$ with $k < 1, A_{\rm int} = 1.8 \cdot 10^{-6}$ erg/cm, and $d = 2$ Å the lattice parameter of Fe. The biquadratic exchange is

$$J_2 \approx [(J_1)^2 L/\pi^3] \cdot A_{\rm Fe}^{-1} \coth[\pi D_1/L],$$

with $L \sim 500$ Å (grain size), $D = 200$ Å (film thickness), and $A_{\rm Fe} = 2.8 \times 10^{-6}$ erg/cm. $K = K_{\rm Fe} D$ with $K_{\rm Fe} = 10^3$ erg/cm^3. The above values taken from fitting the magnetization curves of the SmCo/Fe bilayers [14] give $J_1 \sim 30$ erg/cm^2 and $J_2 \sim 61$ erg/cm^2 at $k \sim 1/3$. Minimization of $E_{\rm tot}$ with respect to Θ gives the remanent orientation of $\mathbf{M}_{\rm Fe}$ as

$$\cos\Theta_{\rm rem} = (1 - 2f)J_1/2[N(f)J_2 - K].$$

Figure 4a shows the calculated rotation of the remanent $\mathbf{M}_{\rm Fe}$ with the degree of remagnetization f of the SmCo film. In the calculation the numerical function $N(f)$ taken from [9] was approximated by a polynomial of fifth power. The value of f grows with increasing maximum value of applied negative fields. The calculated $\Theta_{\rm rem}(f)$ can be compared with the measured dependence of the remanent $\mathbf{M}_{\rm Fe}$ angle on the negative field $\Theta_{\rm rem}(H)$ in Fig. 4b. This yields $f(H)$ characterizing a remagnetization curve for SmCo (see Fig. 4b). It shows the sharp inversion of the SmCo domains corresponding to the rotation of the remanent $\mathbf{M}_{\rm Fe}$. These results are in good agreement with macroscopic $M(H)$ curves (Fig. 2b) which show ~ zero remanence after application of -3.1 kOe.

Note, that the area in the plane around the hole in Figs. 3c–3f is strongly inhomogeneous, indicative of the underlying magnetization oscillations in the SmCo film. However, this area appears smoother at the beginning of the rotation of $\mathbf{M}_{\rm Fe}$ (Fig. 3b) and when it becomes nearly inverted (Fig. 3g). This supports the above mechanism for the biquadratic interactions in exchange coupled magnets due to the inhomogeneous remagnetization of the hard layer. The maximum inhomogeneity is expected at intermediate fields when areas of opposite magnetizations become equal. It should be less at smaller and larger fields. The partial inhomogeneous switching of SmCo seems to be also the main reason for the degradation of the exchange spring magnets (this problem is addressed as an important issue for both magnetic springs [15] and spin-dependent tunnel junctions [16]). However, the origin of the switching and the effects of the soft layer on this process need more careful investigation.

In conclusion, we observed noncollinear remanent magnetic configurations formed in SmCo/Fe exchange springs. Such configurations were not expected for two ferromagnetic layers in direct contact. They appear as a result of the partial remagnetization of the hard layer and can be explained by an adaptation of Slonczewski's fluctuation mechanism of the biquadratic exchange. The reported observations of the field induced biquadratic exchange open a possibility of tuning the exchange coupling between the ferromagnetic layers using fine changes of the magnetic structure of hard ferromagnetic films by an external magnetic field.

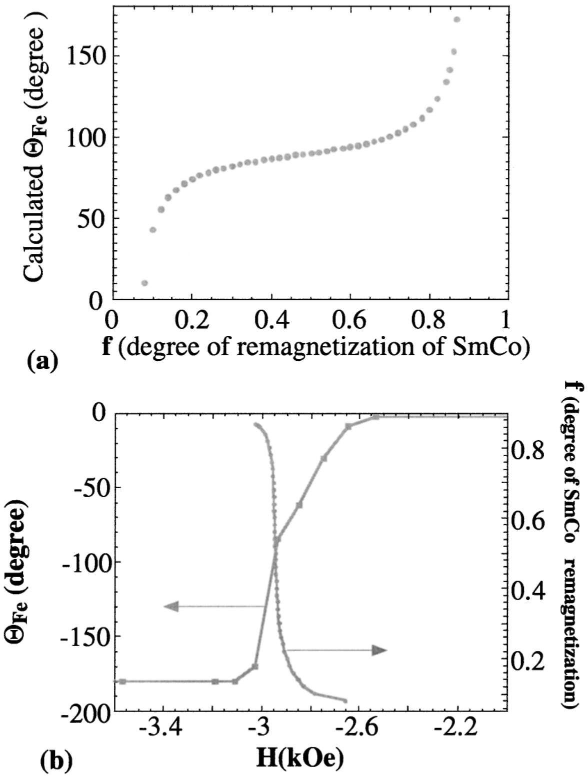

FIG. 4. (a) Calculated angle of remanent $\langle \mathbf{M}_{\rm Fe} \rangle$ as a function of the SmCo remagnetization degree f. (b) Measured angle of remanent $\langle \mathbf{M}_{\rm Fe} \rangle$ after different negative applied fields H and the remagnetization curve $f(H)$ for SmCo recalculated using (a).

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