Magnetic Tuning of Biquadratic Exchange Coupling in Magnetic Thin Films

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The effective interlayer coupling between antiferromagnetically coupled hard and soft ferromagnetic thin films is investigated as a function of the magnetic bit length in the hard layer, which is controlled using a magnetic recording system. The interlayer coupling is explored by studying the magnetization reversal of the soft layer. As the bit length decreases, the coupling evolves from antiferromagnetic to biquadratic to uncoupled. These results are reproduced using a micromagnetic model and determine the applicability range of Slonczewski's fluctuation model of biquadratic coupling.

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With the ability to atomically engineer thin magnetic films and multilayers, many new phenomena have been discovered, such as the oscillating interlayer exchange coupling between two ferromagnetic (FM) layers across thin metal layers [1]. In these systems, the magnetization of the FM layers is either parallel or antiparallel in remanence, depending on the sign of the coupling. Such coupling is often referred to as bilinear coupling and its areal energy density is described by $E_{BL} = -J_1(m_1 \cdot m_2)$, where m_1 and m_2 are the magnetization vectors in the two FM layers and J_1 is positive (negative) for ferromagnetic (antiferromagnetic) coupling. Upon close investigation of such an exchange-coupled Fe/Cr/Fe trilayer system, a second type of coupling was discovered, wherein the FM Fe layers are oriented orthogonally in remanence [2]. Referred to as biquadratic (BQ) coupling, it is described phenomenologically by the energy term $E_{BQ} = -J_2(m_1 \cdot m_2)^2$. Here, J_2 is a negative constant describing the strength of the BQ coupling.

Several mechanisms, which may lead to BQ coupling, have been identified and are reviewed in Ref. [3]. In the current work, we confine ourselves to the fluctuation mechanism, which was put forth to explain the observed BQ coupling in the Fe/Cr/Fe system [4]. In this theory, the BQ coupling energy is related to the thickness fluctuations of the Cr spacer layer, which in turn lead to a spatially alternating sign of J_1. To minimize E_{BL}, the Fe layers should be parallel in areas with FM coupling and antiparallel in areas with antiferromagnetic (AF) coupling. However, due to the high intralayer exchange coupling, such a configuration comes at a high cost of domain wall energy and often does not constitute an energy minimum. When the terrace size in the Cr spacer layer is not too large, the energetically most favorable state is aligning the average magnetizations of the FM layers perpendicular relative to each other with small angular fluctuations to locally reduce E_{BL}. When the domain wall width is larger than the layer thicknesses t, this fluctuation model yields the following relation [4]:

$$J_2 = -c\frac{(J_1)^2 L}{\pi^3 A}\coth\left(\pi\frac{t}{L}\right), \tag{1}$$

where A is the intralayer exchange coupling constant, L is the terrace size, and $c = 4$. Although this approach explains many experimental results, it is difficult to verify the underlying assumptions due to the lack of precise terrace size control and the poorly known local bilinear exchange coupling in many systems [5,6].

Experimental verification of BQ coupling generated by magnetic inhomogeneities was demonstrated in a Fe/SmCo bilayer system with uniaxial anisotropy [7]. In this system, it is not the physical structure of the interface, but the magnetization structure of the magnetically hard SmCo layer that generates fluctuations of the interfacial exchange coupling and is responsible for the appearance of BQ coupling. In contrast to the Fe/Cr/Fe system, this system allows the examination of different coupling states in one single sample. However, the intrinsic J_1 cannot be determined, because the exchange coupling is larger than the coercivity of either layer. Furthermore, the domain structure of the SmCo layer is only statistically controlled.

In the present Letter, we report on a different experimental approach, which closely imitates Slonczewski's model [4] and provides a model system for investigating BQ coupling. The samples consist of a thin magnetically soft Co layer that is AF coupled to a hard-magnetic recording media layer. Spatial fluctuations of the interlayer exchange field H_{Ex} are introduced by means of well-defined magnetic bit patterns that are written in the hard-magnetic recording layer. While the underlying mechanism for the appearance of BQ coupling remains unchanged, the presented experimental approach allows precise control of the magnetization structure using a magnetic recording system. Furthermore, all relevant experimental parameters are either known or can be extracted from measurements. The experimental results establish that BQ coupling can originate from fluctuations in the bilinear coupling and that the magnitude strongly depends on the spatial scale of these fluctuations. These results are quantitatively described by a micromagnetic model, which also allows evaluating the validity range of the fluctuation model and Eq. (1).

The investigated samples are based on the design of antiferromagnetically coupled (AFC) magnetic recording media [8]. They consist of two FM granular layers, separated by a 0.6-nm Ru layer to maximize the AF exchange coupling (Fig. 1). While in AFC media, both FM layers typically are made from similar, granular CoPtCrB alloys, the lower layer of the films used in this study is pure Co. The alloy choice and the relatively large thickness of the Co layer greatly enhance the effect of A. Magnetic bits of lengths $L = 250$, 111, 71, 53, and 42 nm are written into the CoPtCrB layer of the sample using a 6-μm wide write head. The transitions between two bits are along the y direction and the average magnetization of the grains in the CoPtCrB layer is parallel to the x axis (Fig. 1). The existence and length of the bits are verified by monitoring the read back signal and imaged using magnetic force microscopy [Fig. 1(c)]. It is important to note that the bits are written into the CoPtCrB layer, whereas the magnetically soft Co layer responds to the coupling across the Ru layer after the write process [9]. Two samples are investigated, which have slightly different Co layer thickness of $t_{Co} = 1.41$ and 1.27 nm. The CoPtCrB layer of both samples is 13.5 nm thick with a coercivity of ~4000 Oe.

The deposition processes promote in-plane magnetic anisotropy and granular growth with an average grain diameter of ~8 nm. The anisotropy axes of the grains are distributed randomly in the film plane leading to local grain-to-grain variation of the magnetization from the average. However, the grains in the Co layer are highly exchange coupled, yielding a relatively soft-magnetic layer with no preferred in-plane anisotropy direction [10]. The high intergranular exchange prohibits the Co layer from locally responding to the magnetization of each individual grain in the CoPtCrB layer. Instead, it responds to the magnetization averaged over many grains [11] and it is the bits as imaged in Fig. 1(c) that are the magnetic fluctuations of interest. This conclusion is supported by micromagnetic modeling described below.

The write procedure is repeated until a ~6-mm wide area is covered with equally sized bits. To characterize the effective interlayer exchange coupling, we measure the magnetization reversal of the Co layer using an alternating gradient magnetometer. The maximum applied field is chosen to be sufficient to saturate the Co layer but small enough to not disturb the CoPtCrB layer. Magnetic hysteresis loops of the Co layer are carried out in both the x and y directions. This procedure is repeated for each bit length and sample.

Examples of the resulting minor loops of the sample with $t_{Co} = 1.41$ nm are plotted in Fig. 2. The top row [2(a), 2(c), and 2(e)] shows the minor loops captured with the field applied along the x direction, while the magnetic field is applied along the y direction for the bottom row [2(b), 2(d), and 2(f)]. All loops are normalized to the saturation magnetization of the Co layer. Figures 2(a) and 2(b) show the minor loops for $L = 250$ nm. The loop in Fig. 2(a) is split into two separate loops, each loop showing the signature of an easy-axis loop. This shape is observed as a result of the separate responses of the Co layer to the positive and negative bits in the CoPtCrB layer. If no external field is present, the sample is in the AF ground state, as illustrated in Fig. 1(a), with the magnetization of the Co layer being antiparallel to that of the CoPtCrB layer. Since the bit cells in the CoPtCrB layer are of equal length, the average magnetization of the Co layer is zero. When a magnetic field is applied in the positive x direction, the domains in the Co layer, in which the magnetization points in the negative x direction, are destabilized and eventually switched. The resulting loop

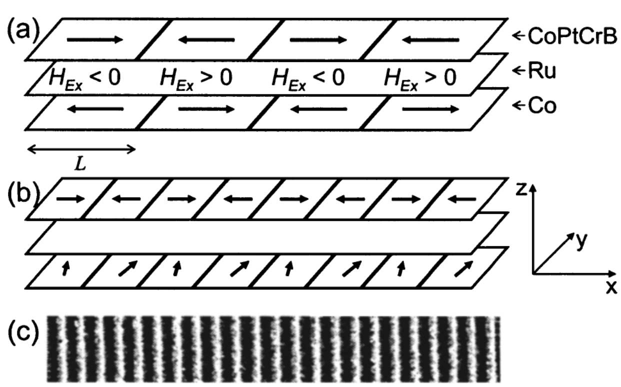

FIG. 1. Structure of the investigated samples. The arrows show the magnetization direction. (a) Magnetization state for large bit lengths L in the CoPtCrB layer with the Co layer antiparallel to the CoPtCrB layer. H_{Ex} represents the antiferromagnetic exchange coupling field between the magnetic layers. (b) Magnetization state for small bit lengths in the CoPtCrB layer with the average Co layer magnetization perpendicular to the CoPtCrB layer. (c) A 12-μm wide magnetic force microscope image of the sample with 250-nm bit lengths.

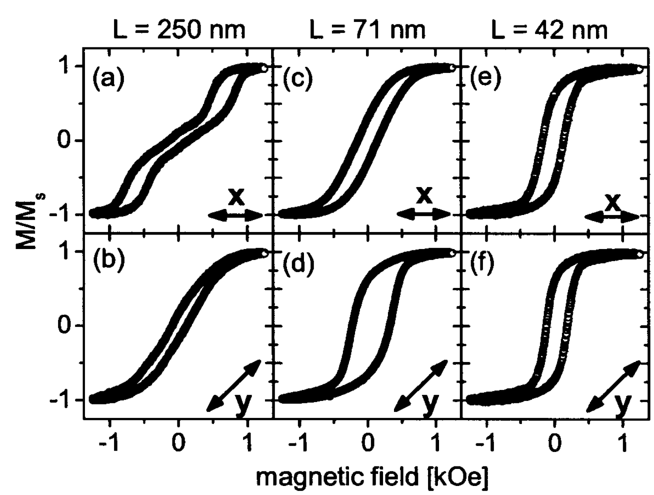

FIG. 2. Magnetic hysteresis loops of the Co layer of the sample with $t_{Co} = 1.41$ nm for different bit lengths L. (a), (c), and (e) are taken along the x axis, and (b), (d), and (f) are taken along the y axis, as defined in Fig. 1.

is shifted by the AF exchange field, which is on the order of 660 Oe for this sample. Similarly, upon application of a negative magnetic field, the remaining domains are switched. Consequently, two shifted loops are observed corresponding to positive and negative bits. When the magnetic field is applied along the y direction, no exchange bias is observed and the magnetization loop is a hard-axis loop [Fig. 2(b)]. Thus, in the presence of large bits in the CoPtCrB layer, the easy-axis of the Co layer is parallel to the x direction.

When the bit length in the CoPtCrB layer is reduced to $L = 71$ nm, the hard-axis loop is along the x direction [Fig. 2(c)] and the easy-axis loop is along the y direction [Fig. 2(d)]. In remanence, the average Co layer magnetization points in the y direction, which is perpendicular to the magnetization direction of the CoPtCrB layer, as illustrated in Fig. 1(b). This suggests the presence of BQ coupling, which is a direct result of the magnetization structure in the CoPtCrB layer. For even smaller bit lengths ($L = 42$ nm), both loops [Figs. 2(e) and 2(f)] are similarly shaped, suggesting that the effect of the interlayer exchange coupling disappears within the experimental error. Since the loops for $L = 71$ nm [Figs. 2(c) and 2(d)] show the clearest difference between the easy- and hard-axis loops, we use these to estimate the BQ coupling coefficient. Accounting for the coercivity and an estimated exchange field, we arrive at a saturation field of 115 Oe. It follows that $J_2^{\rm exp} = 0.023 \pm 0.005$ erg/cm^2 [solid square in Fig. 3(b)]. The solid circle in Fig. 3(b) shows $J_2^{\rm exp}$ for the sample with $t_{\rm Co} = 1.27$ nm.

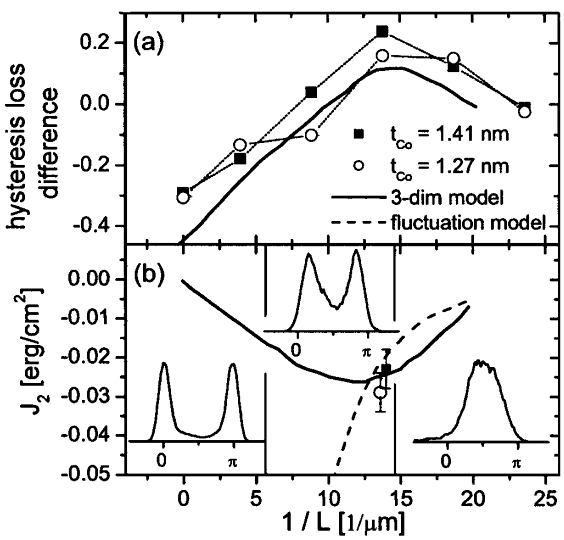

FIG. 3. (a) Normalized hysteresis loss difference calculated from the experimentally measured loops as a function of the inverse bit length L. Negative (positive) values of the coupling direction indicate bilinear (biquadratic) behavior. The dotted lines are a guide to the eye. The solid line represents micromagnetic modeling results for the sample with $t_{\rm Co} = 1.41$ nm. (b) Comparison of the biquadratic coupling constant J_2 estimated from the experiment, the closed-form fluctuation model (dashed line), and the micromagnetic model (solid line). The insets show modeled distributions of the magnetization angles in the Co layer for $L = 250$, 71, and 42 nm.

Because of the complex nature of the loops, in particular, the split loop type shown in Fig. 2(a), conventional measures, similar to the one used for the loops at $L = 71$ nm, do not allow the quantification of the induced anisotropy. We have therefore devised a method based upon the hysteresis loss for our quantitative analysis [12]. The normalized hysteresis loss difference $d = (A_y - A_x)/(A_y + A_x)$ is plotted in Fig. 3(a) for both samples. Here, A_x and A_y are the areas enclosed by the loop, when the field is applied along the x and the y directions. In this notation, $d < 0$ is indicative of a system dominated by bilinear coupling. In contrast, $d > 0$ suggests that the magnetization of both FM layers is aligned perpendicular and BQ coupling is dominating. Model calculations based on the Stoner-Wolfarth model show the same behavior of d as the easy axis is rotated. If the CoPtCrB layer of the sample with $t_{\rm Co} = 1.41$ nm (filled squares) is uniformly magnetized, d is negative, i.e., the Co layer is AF coupled to the CoPtCrB layer. At $L \sim 125$ nm, d crosses through zero and the system starts to be dominated by the BQ coupling. After peaking at $L \sim 71$ nm, d decreases and vanishes at $L \sim 42$ nm. The curve for the sample with $t_{\rm Co} = 1.27$ nm (open circles) shows a similar trend, except that it is slightly shifted to smaller L, as predicted by the fluctuation [4] and the micromagnetic model discussed below. Thus, our data verify the existence of BQ coupling and determine the fluctuation length scale for which it occurs. Note that these experiments are performed for isotropic Co layers. Further experiments and modeling show that an only modest uniaxial anisotropy in the Co layer may prohibit the observation of the BQ coupling.

A three-dimensional micromagnetic model similar to the one in Ref. [13] is used to help understand the experimental data. Two two-dimensional planes of macroscopic spins, with each spin representing a single grain in the CoPtCrB layer and the Co layer, respectively, model the sample. Each pair of corresponding spins in the two layers is AF exchange coupled. The easy axes of the spins of the CoPtCrB layer are randomly distributed in the x-y plane. Since no other field is applied on the spins, the magnetization of each spin in the CoPtCrB layer is aligned along the easy axis. The transitions between two bits in the CoPtCrB layer are approximated by an *atan* transition of 15-nm width [14]. At small L, the width of the transition becomes comparable to the bit length, leading to a reduction of the average magnetization amplitude in the CoPtCrB layer. In this case, a reduction of the AF exchange coupling is observed. The soft-magnetic Co layer consists of exchange-coupled spins, which are free to rotate in the x-y plane. Therefore, the energy terms considered in this model are AF coupling energy, intergranular exchange energy in the Co layer, and the averaged magnetostatic energy. A quasi-Newton method is used to minimize the sum of these energies by varying the rotation angles of the individual spins in the Co layer after a fixed pattern of bit length L was created in the upper CoPtCrB layer.

To compare the theoretical and experimental results, we have calculated the average effective anisotropy and its easy-axis angle relative to the x axis. Then, it is straightforward to calculate the energy barriers for switching in the x and y directions. The normalized energy barrier difference is related to the hysteresis loss difference and is plotted as a solid line in Fig. 3(a). For this calculation, the bilinear exchange constant is extracted from the experiment using $J_1 = -H_{Ex} M_{Co} t_{Co} = -0.134\ \mathrm{erg/cm^2}$, where $M_{Co} = 1422\ \mathrm{emu/cm^3}$ and $t_{Co} = 1.41$ nm. The grains are 8 nm in diameter with 0.9 nm spacing. Agreement between experiment and modeling results is achieved for the whole bit length range using $A = 2.0 \pm 0.2\ \mu\mathrm{erg/cm}$ for the intralayer exchange coupling strength, close to the bulk value of Co [15].

The solid line in Fig. 3(b) is a theoretical estimate of the BQ coupling coefficient J_2 as a function of the inverse bit length and is in quantitative agreement with the experimental data. The increase of $|J_2|$ starting from large L is dominated by the rotation of the Co layer magnetization into the y direction. In contrast, the decrease of $|J_2|$ for $L < 71$ nm is primarily caused by the reduction of the effective, induced anisotropy at small L. The insets of Fig. 3(b) show typical distributions of the magnetization angles in the Co layer for three different L, as determined by the micromagnetic model. They illustrate the tendency of the systems toward a bimodal distribution as L increases. The development of a bimodal distribution coincides with the appearance of discrepancies between micromagnetic and fluctuation model.

For comparison, Eq. (1) with $c = 2$ is also plotted in Fig. 3(b) (dashed line). Here, the intrinsic J_1 is reduced proportionally to the magnetization amplitude of the CoPtCrB layer, such that $J_1^{\mathrm{eff}} = kJ_1$ with $k = 0.6$ for $L = 71$ nm and $k = 0.4$ for $L = 42$ nm. We observe a strong discrepancy between our theoretical J_2 curve and the fluctuation model for $L > 71$ nm ($1/L < 14\ \mu\mathrm{m}^{-1}$). While Eq. (1) predicts an increasing $|J_2|$ with increasing L, the micromagnetic model predicts a maximum in $|J_2|$ and then a subsequent decrease. The differences are the result of assumptions made in deriving Eq. (1) [16] and extending it to large L, for which they no longer apply. Furthermore, the three-dimensional model possesses more degrees of freedom and, hence, is able to better minimize the total energy. In spite of the approximations, the experimentally measured BQ coupling is in quantitative agreement with Eq. (1) for $L < 71$ nm. This range, corresponding to $|J_2| < \sim 0.25 |J_1^{\mathrm{eff}}|$, provides an experimentally derived limit to where Eq. (1) is applicable and is considerably smaller than previously estimated [3,17].

In conclusion, our experiments on the effective interlayer exchange coupling between antiferromagnetically coupled hard and soft ferromagnetic thin films have observed biquadratic coupling for certain bit lengths. The experimental system allows the measurement and control of most parameters that affect biquadratic coupling. As a result, experiments can be quantitatively compared with models. We find that the fluctuation model is adequate only for describing experimental data with small bit lengths, where magnetic frustration dominates. A three-dimensional micromagnetic model is presented, which is able to reproduce the experimental results over the full range of measured bit lengths. This experimental system may have applicability to the study of other physical phenomena, such as spin-flop coupling [18] and coercivity enhancement [19] in exchange bias, where magnetic frustration is also believed to play a role.

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