Direct Observation of Exchange Bias Related Uncompensated Spins at the CoO/Cu Interface

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Magnetization-induced optical second harmonic generation (MSHG) from the exchange-biased CoO/Cu-(X)/Fe multilayer shows the presence of pinned uncompensated spins at the CoO/Cu interface. For increasing Cu spacer thickness, the exchange bias measured via the hysteresis loop shift diminishes and disappears at $X = 3.5$ nm, while the MSHG signal still shows a strong magnetic contribution from the CoO interface. This indicates that the magnetic interaction between Fe and CoO layers is sufficiently strong to induce order in the antiferromagnetic layer even at a spacer thickness for which there is no observable hysteresis loop shift.

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Despite its very important applications in spin valves, and therefore in storage and sensor devices, there is still no complete theoretical explanation for the exchange bias effect, which was discovered almost 50 years ago [1]. Recently, this phenomenon has attracted a lot of renewed research interest [2,3] as its understanding appears to be of prime importance for its tuning [4], reliability, and functioning.

Exchange bias is obtained when a ferromagnet (FM)/antiferromagnet (AFM) bilayer is cooled from above the Néel temperature but below the Curie temperature, in the presence of an external magnetic field. Its main characteristics are: a shift H_E of the hysteresis loop away from the zero field and, usually, an increase of the coercivity H_C. The temperature below which the effect occurs is called the blocking temperature T_B.

Recent research suggests that a small number of uncompensated AFM spins at the interface might be the origin of the loop shift [5]. On the other hand, it has also been suggested that the main part of those uncompensated spins will couple to the ferromagnet and rotate with it [6,7], thus contributing to the coercivity enlargement. These explanations present exchange bias as essentially an interfacial effect, having a next-neighbor range.

However, other studies revealed a long-range nature of exchange bias: upon insertion of a nonmagnetic spacer layer between the FM and the AFM, some authors observed an exponential [8] or even sharper [9] decrease of the effect, while others have reported an oscillatory behavior [10]; in some cases oscillations were shown to occur only at certain temperatures [11].

In order to elucidate the role of the interfaces and to understand the interplay between the short and long-range aspects of exchange bias, we have applied the interface-sensitive technique of magnetization-induced second harmonic generation MSHG in combination with the "bulk"-sensitive linear magneto-optical Kerr effect (MOKE) to investigate both the temperature dependence of exchange bias and its value as function of the distance (X) separating the FM from the AFM across a Cu spacer. We show that the appearance of exchange bias at the temperature T_B is accompanied by the formation of pinned uncompensated spins at the AFM/spacer interface, in agreement with observations made by other techniques [5–7,12]. Those spins are aligned under the influence of the FM interface and are directly responsible for exchange bias. To our surprise, we find that while the hysteresis loops measurement indicates an almost complete disappearance of exchange bias above $X = 3.5$ nm, MSHG reveals that, even at this thickness, there is still a good alignment of pinned uncompensated spins at the AFM/spacer interface below T_B.

Our results indicate that the range upon which the FM influences the magnetic order at the AFM interface extends even further than the distance determined from the hysteresis loop shift. In addition, we demonstrate the excellent sensitivity of the MSHG technique to probe these very important but buried interfaces.

Some work on detecting uncompensated spins in exchange-biased systems with MSHG has been done previously [13] on a different type of multilayer (without spacer); however, the authors concentrated on a single aspect of the MSHG dependence (the polarization rotation) and the observed effects were very subtle.

The basic structure of our layered samples was Si(111)/Fe/Cu/CoO/Au. Initially, 6 nm Fe was deposited by molecular beam epitaxy (MBE) on hydrogen-passivated Si(111), followed by a Cu layer with varying thickness. After the preparation of 2 nm CoO [14,15], the sample was covered by a 6 nm Au cap layer to prevent contamination from the atmosphere. The Cu thickness was varied both on a single sample, in the form of a stepped wedge, and as a series of separate samples. The Fe film possesses a single crystalline bcc(110) surface orientation [16] while the CoO consists of densely packed roundly shaped particles [15]. Transmission electron microscopy showed sharp interfaces for all the discussed Cu thicknesses (for Cu thicknesses appreciably below 2 nm, pinhole effects play a major role). Because the CoO does not reveal any x-ray diffraction peaks, it is assumed to be amorphous.

Exchange bias was induced by cooling the sample from a temperature of 300 K, in the presence of an external magnetic field of 2.5 kOe. Hysteresis loops extended from −4.5 to +4.5 kOe.

MSHG measurements were performed using a Ti:sapphire laser at 800 nm wavelength with a pulse width of ~100 fs and a repetition rate of 82 MHz. The laser power was 5 mW and the light was focused to a spot with diameter of ~100 μm. The angle of incidence was 30° and the magnetic field was applied in the longitudinal configuration [for further details see Ref. [17]].

For intense electromagnetic fields, such as those generated by a pulsed laser beam $\mathbf{E}(\omega)$ incident on a thin multilayer film, the polarization at the harmonic frequency 2ω is given by

$$\mathbf{P}_i^{(l)}(2\omega) = \chi_{ijk}^{(l)} \mathbf{E}_j^l(\omega) \mathbf{E}_k^l(\omega), \tag{1}$$

where χ_{ijk} is a third order polar tensor describing the nonlinear second-order optical susceptibility at the symmetry breaking interface between the centrosymmetric films and l numbers the interfaces in our sample [17]. We can separate two types of contributions to the susceptibility: the "magnetic" (χ_m) and "nonmagnetic" (χ_{nm}), depending on whether the tensor elements associated with them change sign upon reversal of the magnetic moment; see inset in Fig. 1. Note that the nonmagnetic part also includes defects or microstructure effects. It should be understood here that any such effects do not have a magnetic orientation, and, in particular, that they will not reverse after field cooling in an opposite external magnetic field.

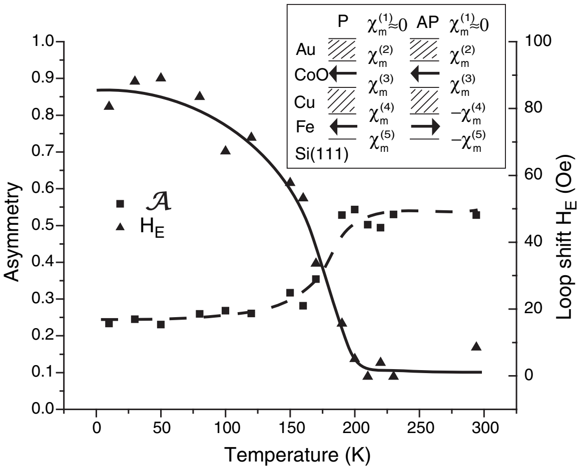

FIG. 1. Temperature dependence of the asymmetry $\mathcal{A}$ (squares) and of the loop shift H_E (triangles) measured with MSHG from a sample with Cu thickness 2.5 nm. The lines are guides to the eye. Inset: effective second-order susceptibilities from different interfaces of the sample, in the case of negative exchange bias, below T_B. $\mathcal{A}$ is determined by reversing the Fe layer magnetization with an applied external field. The arrow in the CoO symbolizes the pinned uncompensated spins in the AFM.

The nonzero net magnetic moment at the CoO interfaces is related to the exchange bias and its sign can be reversed if the sample is field cooled in an opposite magnetic field. We can distinguish two types of configurations: parallel (P) and antiparallel (AP), which is related to the relative orientation of the FM layer; see inset in Fig. 1.

In this Letter we use the model based on the work of Sipe [18] and applied among others by Wu *et al.* [19] which does not include the contribution of quadrupolar terms from the bulk. Therefore, the tensor elements that we use are "effective" and may contain a contribution from these terms.

For a fixed polarizer-analyzer combination, the second-order susceptibility χ at any given interface can be described by a single number. In the limit of ultrathin films we can combine those susceptibilities:

$$\chi_m^{\mathrm{P}} = (\chi_m^{(2)} + \chi_m^{(3)}) + (\chi_m^{(4)} + \chi_m^{(5)}) \tag{2a}$$

$$\chi_m^{\mathrm{AP}} = (\chi_m^{(2)} + \chi_m^{(3)}) - (\chi_m^{(4)} + \chi_m^{(5)}) \tag{2b}$$

$$\chi_{nm} = \chi_{nm}^{(1)} + \chi_{nm}^{(2)} + \chi_{nm}^{(3)} + \chi_{nm}^{(4)} + \chi_{nm}^{(5)}. \tag{2c}$$

Note that here we have included a possible contribution of the Au/CoO interface $\chi_m^{(2)}$. A potential contribution from the AFM ordering in CoO [20] is incorporated in the nonmagnetic part as it should be symmetrical with respect to the direction of the field cooling.

The second harmonic intensity for the parallel and antiparallel configurations is then given by

$$I^{\mathrm{P/AP}} = |\chi_{nm} + \chi_m^{\mathrm{P/AP}}|^2 I^2, \tag{3}$$

where I is the intensity of the incoming fundamental light.

Further, we can define the asymmetry $\mathcal{A}$ (or relative magnetic contrast) as

$$\mathcal{A} = \frac{I^{\mathrm{P}} - I^{\mathrm{AP}}}{I^{\mathrm{P}} + I^{\mathrm{AP}}} = \frac{2R}{1 + R^2} \cos\phi, \tag{4}$$

where $R = |\chi_m^{(4)} + \chi_m^{(5)}| / |\chi_{nm} + \chi_m^{(2)} + \chi_m^{(3)}|$ and ϕ is the phase angle between numerator and denominator. For $R^2 \ll 1$ $\mathcal{A}$ is proportional to R, whereas for $R^2 \gg 1$, it is proportional to $1/R$. For the intermediate case, $R \approx 1$, $\mathcal{A}$ is constant and equal to $\cos\phi$.

Above T_B, exchange bias and thus the possible magnetic contributions of the CoO interfaces disappear, i.e., $\chi_m^{(2)} = \chi_m^{(3)} = 0$. In that case, R can be written simply as the ratio between the magnetic and nonmagnetic tensor elements:

$$R = |\chi_m^{(4)} + \chi_m^{(5)}| / |\chi_{nm}|. \tag{5}$$

Below T_B the presence of pinned uncompensated spins at the CoO interfaces may lead to $\chi_m^{(2)}$ and $\chi_m^{(3)} \neq 0$, resulting in a decrease in R and consequently, for $R^2 \ll 1$, a decrease in $\mathcal{A}$. This is illustrated in Fig. 1, which shows the temperature dependence of both $\mathcal{A}$ and the exchange bias value measured with MSHG for a sample with $X = 2.5$ nm. These results clearly demonstrate the appearance of $\chi_m^{(2)}$ and/or $\chi_m^{(3)} \neq 0$.

To investigate the decrease of exchange bias as function of the distance separating the AFM and FM layers, we used a "wedge" sample with six different thicknesses of Cu spacer. In Figs. 2(a) and 2(b) the loop shift H_E and the asymmetry $\mathcal{A}$ of this sample are plotted as a function of temperature showing an almost opposite behavior of these two quantities: while H_E decreases strongly with thickness, the temperature dependence of $\mathcal{A}$ increases. Note that in Fig. 2(b) the thickness dependence is visible only below T_B; from this we can conclude that the observed effect is of magnetic and not optical nature. Despite some scatter in the data, the trends are absolutely clear: for $X = 1.5$ nm, there is no pronounced change in the asymmetry with decreasing temperature, whereas for larger thickness the asymmetry diminishes gradually and clearly with decreasing temperature.

The asymmetry for $X = 0$ nm is given as a reference. Note that for the absence of a spacer the optical response is very different since the number of layers and interfaces is smaller. Though the loop shift values are almost the same as those for $X = 1.5$ nm, this was not found to be the case for the coercivity with $X = 0$ nm exhibiting twice the coercivity values of $X = 1.5$ nm.

The inset in Fig. 2 shows that the H_E dependence on spacer thickness shows a sign of oscillation around $X = 2$ nm for $T > 100$ K. This interesting feature, which is possibly due to a RKKY-like coupling between the FM and the AFM, has been observed previously by Lin *et al.* [11].

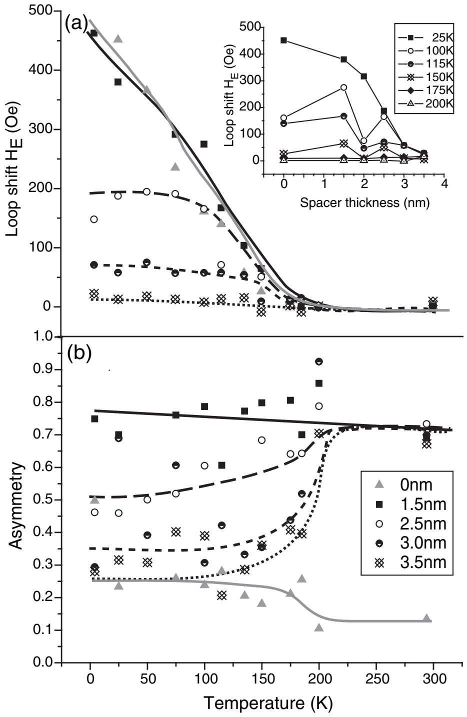

FIG. 2. Temperature dependence of the exchange bias loop shift measured with MOKE (a) and of the asymmetry measured with MSHG (b). The lines are guides to the eye.

For $T < T_B$, Fig. 2 shows that at $X = 1.5$ nm, the exchange bias is relatively large and increases strongly with decreasing temperature from T_B, while the MSHG asymmetry remains constant. On the other hand, for $X = 3.5$ nm, the exchange bias is almost zero, and does not change much with temperature, while the asymmetry decreases strongly below T_B.

Here we observe the dependence of $\mathcal{A}$ on the Fe interfaces: for $X = 1.5$ nm, $\mathcal{A}$ remains constant, indicating that here $R \approx 1$ (see discussion above). For $X > 1.5$ nm, the contribution of the Fe interfaces ($\chi_m^{(4)}$ and $\chi_m^{(5)}$) decreases, leading to $R \ll 1$ and thus $\mathcal{A} \approx R$. A calculation based on Fresnel coefficients and the refractive index of Cu gives indeed a diminishing of the SHG contribution from the Cu/Fe interface by almost 50% with increasing thickness of the Cu spacer from 1.5 to 3.5 nm.

In order to check the presence of a contribution of the Au/CoO interface, i.e., $\chi_m^{(2)}$, we studied the MSHG intensity as function of the analyzer rotation at different temperatures for all Cu spacer thicknesses. Figure 3(b) shows that for $X = 3.5$ nm, below T_B, the curves demonstrate a polarization rotation due to the appearance of new tensor components—those responsible for the pinned AFM spins at the interface(s). However, for $X = 0$ nm [Fig. 3(a)], there is no such rotation. Therefore, since the Au/CoO interface is common for these two cases, while the CoO/Cu appears only for $X > 0$ nm, we can conclude that $\chi_m^{(2)} \approx 0$ and that the observed spin order is located at the CoO/Cu interface. The ferromagnetic nature of this ordering can be clearly demonstrated with the polarization dependence of the MSHG signal. Figure 4(a) shows the MSHG response of the sample above T_B when the magnetization is saturated in both directions. After field cooling [Figs. 4(b) and 4(c)], new tensor components corresponding to the pinned uncompensated spins at the CoO/Cu interface appear. We can clearly distinguish the parallel and antiparallel configurations: when the CoO/Cu interfacial spins are parallel to the Fe ones [dashed line on Fig. 4(b) and solid line on Fig. 4(c)], the curves below T_B have the same shape as those above T_B. On the other hand, when the pinned uncompensated spins at the CoO/Cu interface and the Fe spins are opposing each other [solid line on Fig. 4(b) and dashed line on Fig. 4(c)], the

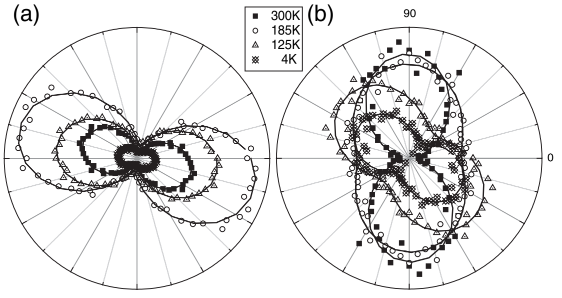

FIG. 3. MSHG intensity from a sample with Cu thickness 0 nm (a) and 3.5 nm (b) as function of analyzer rotation angle.

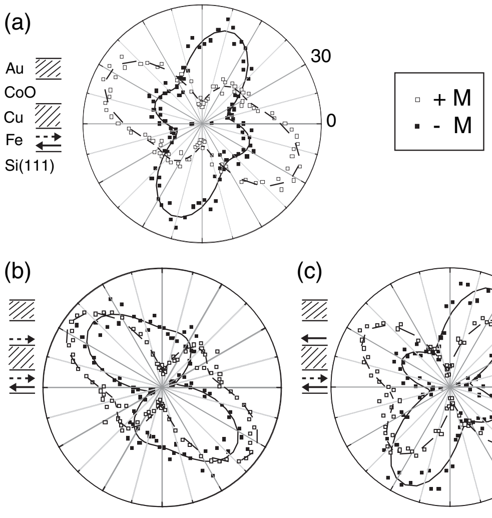

FIG. 4. MSHG intensity from a sample with Cu thickness 2.5 nm, as function of incoming polarization rotation angle. (a) For magnetization $+\mathbf{M}$ and $-\mathbf{M}$, above T_B, (b) for magnetization $+\mathbf{M}$ and $-\mathbf{M}$ below T_B and after positive field cooling, (c) for magnetization $+\mathbf{M}$ and $-\mathbf{M}$ below T_B and after negative field cooling.

shape of the polarization dependence is clearly modified due to the sign change of $\chi_m^{(3)}$.

Thus the asymmetry reveals the presence of ferromagnetically ordered AFM spins at the CoO/Cu interface. Of particular interest is the fact that for $X = 3.5$ nm, the loop shift is almost zero indicating the absence of exchange bias, while the anisotropy $\mathcal{A}$ and the polarization data on Fig. 4 clearly show the presence of pinned uncompensated spins at the antiferromagnet/spacer interface. It appears that for this thickness of Cu, the magnetic order of CoO is influenced by the Fe although the effect is not strong enough to induce measurable exchange bias effects in return. One should realize that for small spacer thickness, the FM layer plays a double role: it induces pinned AFM spins, but also diminishes the number of uncompensated spins available for pinning by strongly coupling to them and forcing them to reverse with the magnetization. On the other hand, for thicker spacer layers, the FM coupling to the uncompensated spins becomes weaker and therefore these are less affected by the magnetization reversal; i.e., they remain pinned in the direction of exchange bias. The distance across which the pinning process occurs extends until a limit that remains to be determined experimentally. In our case we can conclude that this limit is larger than 3.5 nm.

In conclusion, we have shown that the formation of pinned uncompensated spins at the CoO/Cu interface, which are rather difficult to detect by means of hysteresis loop measurements, can be observed with the asymmetry sensitivity of the nonlinear magneto-optical technique of MSHG. This new possibility allows us to study the limits upon which the FM layer affects the AFM magnetic ordering at the interface in exchange-biased multilayers. We have provided evidence that the range of this phenomenon is relatively large and that it extends beyond distances where effects in the hysteresis loop are observed. Our technique thus demonstrates its high sensitivity to the interfacial exchange coupling and its excellent potential as a tool for studying magnetic interface effects in general.

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