Perpendicular exchange bias and magnetic anisotropy in CoO/permalloy multilayers

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For permalloy/CoO multilayers, after perpendicular field cooling, perpendicular exchange bias has been observed below room temperature. For out-of-plane hysteresis loops, the exchange field and the coercivity at fixed temperatures are proportional to $1/\Delta H_{FM}$ and $1/t_{FM}^{3/2}$ ($t_{FM}$=permalloy layer thickness), respectively. At the same time, the remnant ratio increases with decreasing $t_{FM}$. The induced effective uniaxial anisotropy changes as a linear function of $1/t_{FM}$, as a symbol of an interface anisotropy. The interface anisotropic energy increases with decreasing temperatures. For samples with small $t_{FM}$, perpendicular anisotropy films can be established at low temperature. The established perpendicular magnetic anisotropy and its remarkable temperature dependence might facilitate perpendicular magnetic recording and magneto-optical storage techniques.

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I. INTRODUCTION

Exchange bias between ferromagnet (FM) and antiferromagnet (AFM) has been studied extensively due to recent interest in magnetoelectronics devices.1–3 In most cases, exchange bias is established through field cooling in the film plane (longitudinal) where the magnetic easy axis of soft ferromagnetic materials normally lies in plane due to the shape anisotropy. In comparison to a free FM layer, exchange biased FM/AFM layers can be characterized by a coercivity enhancement accompanied with a shift of the hysteresis loop from zero field.4 For the longitudinal exchange bias, the dependence of the exchange field and the coercivity enhancement on constituent layer thickness is well known. For example, the exchange field is proportional to the inverse FM layer thickness and the coercivity generally decreases with increasing FM layer thickness, as a symbol of an interface nature of the exchange bias.

Recently, perpendicular exchange bias has been observed in ferromagnetic Co/Pt multilayers with perpendicular magnetic anisotropy when they are in contact with an antiferromagnetic layer such as CoO, FeMn, and FeF2 after field cooling perpendicular to the film plane.5–7 The coercivity and the perpendicular anisotropy are enhanced, in contrast to corresponding free magnetic multilayers. This will be very helpful in fabrications of recording media in ultrahigh-density perpendicular magnetic recording technique because high perpendicular anisotropy is required to overcome superparamagnetism in perpendicular magnetic recording media.8 Some issues, such as the temperature dependence of the perpendicular exchange bias and the magnetization reversal process, have been studied. For example, the exchange field and the coercivity for the perpendicular exchange bias have been found to change as a linear function of the temperature in [Co/Pt]/NiO structures.9 Asymmetric magnetization reversal has been observed in [Co/Pt]/CoO system.10 Up to now, however, the primary characteristic, such as the FM layer thickness dependence of the exchange field and the coercivity, has not been revealed yet. Therefore, the interface nature of the perpendicular exchange bias has not been addressed systematically. This is possibly because perpendicular films of Co/Pt multilayers can only be established with very small Co layer thickness and limited numbers of bilayer. More remarkably, in most studies the exchange bias was still established along the magnetic easy axis. In this paper, we report on perpendicular exchange bias in FM/AFM layers with intrinsic in-plane ferromagnetic anisotropy. Experiments were performed on FM/AFM multilayers with permalloy (Py) and CoO as the ferromagnetic and antiferromagnetic layers, respectively. The dependence of the exchange field and the coercivity on the FM layer thickness and the temperature is studied. Perpendicular field cooling introduces strong uniaxial anisotropy which increases with decreasing temperature and is of the nature of the positive interface anisotropy. For small FM layer thickness, the induced uniaxial anisotropy can overcome the demagnetization energy to establish perpendicular anisotropy films.

II. EXPERIMENTS

Multilayers were deposited onto Si (100) substrates in a computer-controlled multisource deposition system, at a base pressure of 10−8 Torr. The CoO layers were deposited from a composite target by rf sputtering and the Py layers were deposited from an alloy target by dc magnetron sputtering. The deposition rates for both components were 0.1–0.2 nm/sec with the Ar pressure of 5 mTorr during deposition. Three types of samples were fabricated. Uniform [CoO(4.0 nm)/Py(4.0 nm)]15 multilayered samples were fabricated for structural investigation. Samples for magnetic measurements have fixed CoO thickness but variable Py thickness with a structure of Cu (30.0 nm)/[CoO (4.0 nm)/Py(0–x nm)]15/CoO (4.0 nm)/Cu (30.0 nm). The Py wedge thickness in the two samples was 0–4 nm and 0–20 nm. Wedge samples allow us to study the FM layer thickness dependence of the perpendicular exchange bias under identical growth conditions. The additional CoO layer was deposited to ensure that each Py layer was sandwiched between two AFM layers.
III. RESULTS AND DISCUSSION

Figure 1 shows x-ray-diffraction patterns for the uniform [CoO(4.0 nm)/Py(4.0 nm)]$_{15}$ multilayer in both low and high angle regions. At low angles, eight main diffraction peaks from the whole multilayer diffraction can be identified. The value of the bilayer period calculated from the main peak separation is 8.1 nm, which is in good agreement with the designed layer thickness. At high angles, the strong 111 peaks for fcc CoO and fcc Py indicate that the multilayers are highly textured. The superlattice satellite peaks indicate that the as-deposited films have very good layered structures.

For magnetization measurements, the multilayers with wedged Py layers were cut into same size pieces along the wedge direction. Magnetization data in the range from 100 K to room temperature and below 100 K were obtained using a vibrating sample magnetometer (ADE model 10) with vector measurement capability and superconductor quantum interference device, respectively. Since the Néel temperature of CoO is 292 K, exchange bias was established by cooling from room temperature to low temperature under an external field.

Figure 2 shows hysteresis loops for a [CoO(4.0 nm)/Py(1.76 nm)]$_{40}$/CoO(4.0 nm) multilayer measured at room temperature (a), (b) and at 100 K (c), (d) after perpendicular field cooling. The left and the right columns refer to in-plane (a), (c) and out-of-plane (b), (d) hysteresis loops, respectively.

In experiments, we found that upon decreasing temperature, the coercivity, the exchange bias field, and the squareness all increase. At 100 K, there is no exchange field for the in-plane hysteresis loop although the coercivity is enhanced, compared to the room-temperature in-plane hysteresis loop. Since the CoO layers have (111) orientation along the film normal direction, in the case of perpendicular field cooling, the spins of Co atoms in CoO layers are located on the surface with half-apex angle. This distribution of AFM spins will induce an enhancement of the coercivity and zero exchange field. This phenomenon is quite different from the longitudinal exchange bias. For example, in the case of longitudinal exchange bias in CoO/Py bilayers, the coercivity is zero along the direction perpendicular to unidirectional axis. In other words, compared to the out-of-plane hysteresis loop at room temperature, the magnetic easy axis of the multilayer with perpendicular exchange bias has been changed from in plane to perpendicular to the film plane with decreasing temperature.

The magnetic anisotropy in the multilayered samples after perpendicular field cooling also shows a strong dependence on the FM layer thickness. Figure 3 shows typical hysteresis loops for [CoO(4.0 nm)/Py]$_{40}$/CoO(4.0 nm) multilayers with various Py layer thickness at 100 K. For thick Py layer thickness, the out-of-plane hysteresis loop is slanted and becomes squared with decreasing Py layer, and vice versa for in-plane hysteresis loops. Apparently, at fixed temperatures below room temperature, the magnetic easy axis of the CoO/Py multilayer is switched from in-plane to out-of-plane with decreasing Py layer thickness. As shown in Fig. 4, for multilayers after perpendicular field cooling, the squareness at 100 K has the opposite FM layer thickness dependence for
the hysteresis loops measured along the in-plane and out-of-plane directions. Note that the value of the squareness was taken by averaging the values of the descending and the ascending branches at the central fields of the hysteresis loops. The effect of perpendicular field cooling is more prominent in samples with thin FM sublayers where perpendicular anisotropy film can be established so that the magnetic easy axis changed from the in plane at room temperature to the out of plane at low temperature.

It is well known that for the longitudinal exchange bias the field cooling will induce an additional uniaxial anisotropy and unidirectional anisotropy at the FM/AFM interface. The unidirectional anisotropy results in the hysteresis loop shift and additional uniaxial anisotropy causes an enhancement of the coercive field (within uniform rotation model). This is also true for the perpendicular exchange bias. Figure 5 shows the dependence of the out-of-plane exchange field and the coercivity on the Py layer thickness at 5 K and 100 K for CoO/Py multilayers after perpendicular field cooling. Apparently, the exchange field is approximately proportional to the inverse Py layer thickness. This means that the perpendicular exchange bias is also of the interface nature. The exchange coupling energy is 0.12 erg/cm² at 5 K, smaller than the value of 0.25 erg/cm² at 10 K for [Co/Pt]/CoO multilayers with perpendicular exchange bias. The difference of the exchange coupling energy might be caused by the difference in the magnetization between Co and Py. The coercivity at 5 K and 100 K is proportional to $1/t_{FM}^{3/2}$, which was also observed for longitudinal exchange bias of Py/CoO bilayers and explained as a result of the random-field model at the FM/AFM interface. Figure 6 shows a typical temperature dependence of the out-of-plane exchange field and the coercivity. For a specific CoO/Py multilayer with perpendicular exchange bias, both the exchange field and the coercivity decrease with increasing temperature. The exchange field is equal to zero near room temperature, that is to say, the blocking temperature is close to the Neél temperature of CoO bulk.

As discussed above, both anisotropies can also be established perpendicular to the film plane. The uniaxial anisotropy energy per unit volume in the FM layer after field cooling along the surface normal can be written as

$$K_\perp = K_U - 2\pi M_S^2 + \frac{K_S}{t_{FM}},$$

where $K_U$, $M_S$, and $t_{FM}$ are the uniaxial anisotropy constant, the saturation magnetization, and the FM layer thickness, respectively.
where $t_{FM}$ is the FM layer thickness. The first term $K_U$ is the intrinsic anisotropy of bulk Py, the second term $-2\pi M_S^2$ is the demagnetization energy due to the shape anisotropy, and the third term is the induced uniaxial anisotropy by the perpendicular exchange bias. Since the last term is of the nature of the interface anisotropy as demonstrated below, it is expressed as $K_S/t_{FM}$. We can use effective anisotropy energy $K_{eff}$ to evaluate the magnetic anisotropy energy difference along the in-plane and out-of-plane orientations. Neglecting the bulk anisotropy energy difference between the in-plane and the out-of-plane orientations, $K_{eff}$ can be written as

$$K_{eff} = K_{||} - K_{\perp} = -2\pi M_S^2 + \frac{K_S}{t_{FM}}.$$  (2)

The sign of $K_{eff}$ determines the orientation of the magnetic easy axis. A negative $K_{eff}$ corresponds to the in-plane anisotropy films and a positive $K_{eff}$ corresponds to perpendicular anisotropy films, respectively. Since the demagnetization energy term $-2\pi M_S^2$ is always negative, strong positive interface anisotropy $K_S$ and small FM layer thickness $t_{FM}$ are required to establish the perpendicular anisotropy film ($K_{eff} > 0$). The anisotropic energy can be experimentally determined from the energy difference to magnetize the sample along the in-plane and the perpendicular directions, thus we can quantify the interface anisotropy at different temperatures for different Py thickness. The area between the virgin curve, the magnetization axis, and the horizontal line of $M = M_1$ determines the work done by the field to bring the sample to the state of magnetization of $M_1$. The energy can be expressed by integration as follows:

$$E = \int_{0}^{M_1} H(M) dM.$$  (3)

During the numerical calculations, the virgin curve approximation for the in-plane magnetization hysteresis loop was directly taken by averaging the upward and downward branches of the loop. For the perpendicular measurements, the loop shift was first accounted for before taking the average of the upward and downward branches. The energy difference calculated from the two approximate virgin curves based on the in-plane and out-of-plane magnetization equals $K_{effFM}$. In principle, the effect of irreversible magnetization reverse induced by AFM spins should be considered. However, it is a reasonable approximation to ignore the irreversible magnetization reverse such as in calculations of the uniaxial anisotropy for the longitudinal exchange bias in many polycrystalline FM/AFM bilayers, such as NiFe/PtMn and FeNi/FeMn bilayers. This can also be seen from the following calculated dependence of the effective uniaxial anisotropy on the FM layer thickness.

The effective anisotropy energy depends on the Py layer thickness and the temperature in CoO/Py multilayers with perpendicular exchange bias. Figure 7(a) demonstrates clearly that $K_{effFM}$ changes as a linear function of $t_{FM}$ at different temperatures. The typical linear dependence apparently shows the induced uniaxial anisotropy is of the nature of the interface anisotropy, similar to observed experimental results in Co/Pt multilayers. One can also find that the intercept of the curve $K_{effFM}$ versus $t_{FM}$, i.e., $K_S$ decreases with increasing temperature.

Figure 7(b) shows the temperature dependence of $K_{effFM}$ for multilayered samples with $t_{FM}$ = 1.3 nm and 2.8 nm. For the two samples, the anisotropy energy increases with decreasing temperature and has similar temperature dependence. However, for $t_{FM}$ = 1.3 nm, $K_{effFM}$ changes sign from negative to positive at 195 K upon cooling, indicating a switch in the magnetic easy axis change from in plane to out
of plane at lower temperatures. For $t_{\text{FM}} = 2.8$ nm, the magnetic easy axis remains in plane in the temperature of 100 K–300 K, as indicated by the negative value of $K_{\text{eff,FM}}$ over the measured temperature range. One can find at fixed temperatures below room temperature the magnetization easy axis is switched from in plane to perpendicular to the film plane when the Py thickness is smaller than an onset value. It can be understood that the onset value of the Py layer thickness increases with decreasing temperature.

Figure 7(c) shows the temperature dependence of the interface anisotropy $K_{S}$ for $t_{\text{FM}} = 1.3$ nm and 2.8 nm. First, one can find $K_{S}$ is independent of the FM layer thickness, which further confirms that the induced uniaxial anisotropy by perpendicular field cooling process exhibits an interface effect. Second, $K_{S}$ is positive below room temperature. Finally, such as $K_{\text{eff,FM}}$, it decreases with increasing temperature and approaches to zero near room temperature. Apparently, for the present FM/AFM multilayers, the temperature dependence of the interfacial anisotropy energy is determined by the AFM material if the Curie temperature of FM material is much higher than the Néel temperature of AFM material. Note that for conventional magnetic/nonmagnetic multilayers, such as Co/Pt ones, the temperature dependence of the interface anisotropy energy is governed by the Curie temperature of the constituent FM material. Positive interface anisotropy was also established in Py/FeMn multilayers by the same method in our group.19

IV. CONCLUSION

In summary, perpendicular exchange bias has been established in CoO/Py multilayers on application of an out-of-plane cooling field. The perpendicular exchange bias is found to exhibit an interface characteristic. The induced uniaxial anisotropy energy can be quantified from the magnetization hysteresis loops measured along the in-plane and out-of-plane directions. The calculated results show that the induced uniaxial anisotropy is of the nature of the interface anisotropy. The interface anisotropy, having the positive sign, increases with decreasing temperature. In Py/CoO multilayers with smaller FM layer thickness, perpendicular anisotropy films can be established along with perpendicular exchange bias. As pointed above, the perpendicular exchange bias is induced by cooling process under an external field perpendicular to the film plane, in analogy with common in-plane exchange bias phenomena in FM/AFM systems. Nevertheless, the results are helpful to understand the mechanism of the exchange biasing such as the effect of the orientation of the AFM spins on the exchange biasing. More importantly, the established perpendicular magnetic anisotropy and especially its distinguished temperature dependence might facilitate perpendicular magnetic recording and magneto-optical storage techniques, especially laser-assisted hybrid magnetic recording. For such applications, one should use an AFM which has high Néel temperature and can induce large interface anisotropy.

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