Unidirectional coercivity enhancement in exchange-biased Co/CoO

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A unidirectional coercivity enhancement, exhibiting the same behavior as a positive exchange bias, has been discovered in a temperature range below the blocking temperature in Co/CoO bilayers. Below this temperature range, the usual shift of the center of the \( M - H \) loops to the negative or antiparallel to the cooling field direction is found. This behavior is observed in both magnetic hysteresis loops and transport properties. The positive exchange bias can be explained by reversible changes in the interfacial pinning by the antiferromagnet causing an asymmetric magnetization reversal and a unidirectional coercivity enhancement along the cooling field direction. © 2002 American Institute of Physics. [DOI: 10.1063/1.1498505]

Exchange bias refers to the shift of the magnetic hysteresis loop along the field axis, when a ferromagnet (F) is coupled to an antiferromagnet (AF).\(^1,2\) When cooled through the Néel temperature in an applied field sufficient to saturate the magnetization of the ferromagnet, the F–AF exchange coupling usually leads to a shift of the hysteresis loop in the direction opposite to the cooling field. Such a shift is referred to as a negative exchange bias. Although not understood microscopically, the unidirectional anisotropy arises from the direct exchange interaction between the ferromagnet and the antiferromagnet. In two systems, Fe/FeF\(_2\) and Fe/MnF\(_2\) bilayers,\(^3,4\) a positive exchange bias has been observed. In these samples, a relatively large cooling field (\( > 1 \) T) reversed the easy direction of the usual negative exchange bias, producing a positive shift. For this system, it was proposed that the additional Zeeman energy provided during the cooling led to the positive exchange bias shift.\(^5\)

Here, we report that positive exchange bias may also be found in a narrow temperature range just below the blocking temperature (\( T_b \)) of Co/CoO bilayers. Although we will refer to this effect as a positive exchange bias, in fact, our discovery is a unidirectional coercivity enhancement. The shift crosses over to the usual negative exchange bias at a temperature, \( T_F \), below the blocking temperature. The positive shift can be as large as one fourth of the coercivity, when measured with a vibrating sample magnetometer (VSM). The VSM loop results are further compared to anisotropic magnetoresistance (AMR) studies of the magnetization reversal process. In both cases, a noticeable asymmetry is present near the blocking temperature, where the positive exchange bias is measured. At lower temperatures, when the shift is in the negative direction, the asymmetries are different and disappear in trained films.\(^6\)

In what follows, we first demonstrate the directional dependence of the exchange bias with temperature as measured with a VSM. Similar hysteresis measurements with AMR highlight the asymmetry that occurs in the temperature range of positive exchange bias. As detailed in the last section, this coercivity asymmetry can be understood as originating from changes in the magnetic disorder at the F–AF interface.

The seven investigated samples consisted of Ta/Co/CoO/Ta thin films. After a 5 nm Ta seed layer was dc sputtered on a Si(100) substrate, the antiferromagnetic CoO was reactively sputtered in an oxygen–argon mixture of 3 mTorr with a partial oxygen pressure of 16 \( \mu \)Torr at a rate of 16.8 nm/min. This AF layer thickness was varied between 2 and 9 nm. Subsequent to the CoO deposition, a Co layer with a thickness between 10 and 20 nm was next sputtered. The Co layer was followed by a 5 nm Ta layer overcoat for oxidation protection. All the samples exhibited qualitatively the same behavior in their magnetic measurements.

The magnetic properties of the samples were determined with a vibrating sample magnetometer, and four-point-probe resistance technique to measure the anisotropic magnetoresistance. For all measurements, the samples were cooled with an in-plane magnetic field from 300 K to a specific measurement temperature, measured, and then warmed back to ambient temperature to reset the exchange bias. Various cooling fields, from 0.2 to 6.5 kOe, yielded identical results within the measurement error. As expected for thin AF films, the observed blocking temperature for the samples was lower than the Néel temperature (\( T_N \)) for bulk CoO (291 K).\(^7\)

At ambient temperature, above \( T_N \), the angular AMR response followed the expected cosine squared law.\(^8\) Since the AMR was used as a probe of the magnetization direction,\(^9\) the exact current direction in the thin film was first determined at 300 K. The sample was then field cooled in 2 kOe with the field perpendicular to the current (the field sweeps were measured collinear with the cooling field).

Typical results from a VSM measurement illustrate the positive exchange bias shift shown in Fig. 1(a) for a sample with Co (13 nm)/CoO (6 nm) that was field cooled in 2 kOe from 300 to 245 K; observation of this effect requires only modest cooling fields since the origin of the positive exchange bias is very different from the previously mentioned case.\(^3\) Note that the loop is shifted in the direction of the cooling field, indicating larger fields are required to magnetize the sample in the same direction as the cooling field. A mirror image was obtained for a cooling field with opposite sign. As shown in Fig. 1(b), when the sample is measured at a lower temperature, 220 K in this case, the familiar negative exchange bias is observed.

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As seen in Fig. 1, the magnitude of the exchange bias depends upon whether it is the first or second loop measured after field cooling. This behavior is summarized in Fig. 2 where the exchange bias for the first and second loops are shown as a function of temperature for the same sample. The sample exhibits a positive exchange bias from the blocking temperature of about 260 K to below 240 K. It should be noted that in the approximate temperature range of 235–240 K, the first loop has a negative bias whereas the second loop has a positive shift. Illustrated in the inset of Fig. 2 is the nonmonotonic behavior of the right coercive field (along the field cooling direction), which results in an enhancement of the coercivity\(^{10,11}\) in the temperature range of positive exchange bias. As discussed below, the asymmetry in this temperature regime arises from the change in disorder of the AF pinning layer as the magnetization reverses.

The magnetization reversal was further studied using AMR to determine the average direction of the magnetization. A representative AMR hysteresis loop is shown in Fig. 3 at a temperature where positive exchange bias is observed for a sample with Co (15 nm)/CoO (2 nm). For the current-field geometry used, the resistance increases with an increasing magnetization component perpendicular to the applied field direction.\(^6\) As in the VSM data, the resistance peaks are shifted positively just below the blocking temperature. Also note, as shown in Fig. 3, the left peak (antiparallel to the cooling field) is sharper than the right peak (parallel), which rises slowly and drops quickly. This asymmetry in the reversal process persists into the second measured AMR hysteresis loop. At temperatures either below the disappearance of the positive exchange bias, \(T_P\), (see the inset of Fig. 3) or above the blocking temperature, \(T_B\), the trained AMR curves are symmetric; i.e., the magnetization reversal processes on either side of the trained loop can be taken to be the same.

We attribute this positive exchange bias or anisotropic coercivity to a distribution of blocking temperatures in the AF grains, as would occur with the expected variation in the grain sizes of polycrystalline CoO\(^{12,13}\). In order to explain how this distribution can effect the observed anisotropic coercivity, we will consider a model system with equal numbers of two types of AF grains. Different blocking temperatures as a result from the different volumes distinguish the two types of grains.\(^{14}\) In the following discussion we will assume the system is at a temperature between the blocking temperatures for the two types of AF grains.

Upon field cooling the sample to this intermediate temperature, the exchange field from both types of AF grains is in the field cooling direction. After a negative magnetic field, applied opposite to the cooling field, reverses the magnetization, the torque from the ferromagnet is sufficient to rotate the low anisotropy AF grains’ sublattices (as the temperature is above their blocking temperature). When a magnetic field is next applied in the field cooling direction and the magnetization rotates back to its original state, the low anisotropy AF grains’ sublattices rotate back to their original orientation, again due to the torque exerted by the ferromagnet.

In the cooling field direction and all subsequent times the F magnetization is in this direction, the net magnetization of each AF grain is aligned with the F magnetization, corresponding to the most ordered interface. This is to be contrasted with the negative field direction, where the rotated AF grains have their net magnetization opposite to that of the larger, stable AF grains. In this state, the sublattice magnetization of AF grains is disordered. The above two distinct states are the starting states for the ferromagnetic reversal. Thus, when the magnetization is being rotated to the negative (positive) field direction, the interface is disordered (or-
dered. Consistent with the observation that the coercivity is enhanced with increased disorder via pinning of domain walls, the positive coercivity is expected to be larger than the negative. In other words, the rotation of the magnetization from the disordered state to the more ordered state along the cooling direction is hindered by domain wall pinning, so that the positive or right coercive field is enhanced by $\Delta H$ and the loop is shifted along the cooling direction. This unidirectional coercivity enhancement is consistent with our observations of a coercivity bump only in the positive coercive fields, as shown in the inset of Fig. 2. If this $\Delta H$, due to the local interfacial inhomogeneities created by the switching of the AF grains, is bigger than the induced unidirectional anisotropy field from the large grains, then the resulting hysteresis loop shift will be positive. At temperatures below the lower blocking temperature all the AF sublattices at the interface are frozen from any rotation and the usual negative exchange bias is observed.

Summarizing, we have observed a positive exchange bias or anisotropic coercivity in Co/CoO bilayers in a temperature range below the blocking temperature. The origin of this effect is due to cyclical changes in the magnetic order at the F–AF interface. These changes come from the nearly reversible switching of low anisotropy AF grains. This switching adds disorder to the F–AF magnetic interface, thereby altering and impeding the magnetization reversal along the cooling field direction. The AMR magnetization reversal curves demonstrate such an asymmetry in the reversal process in the regime when the loop has a positive bias. Finally, the unidirectional coercivity enhancement in the Co/CoO samples should be distinguished from the positive exchange bias found in Fe/FeF$_2$ samples. In the present work, the cooling field is not strong enough to induce a unidirectional anisotropy in the opposite direction, but rather at specific temperatures ($T_P<T<T_B$), the induced unidirectional anisotropy is too small to compete with the dominating asymmetry in left and right coercive fields.

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1 W. H. Meiklejohn and C. P. Bean, Phys. Rev. 102, 1413 (1956); 105, 904 (1957).
14 Since the crystalline anisotropy energy of a grain is an extensive quantity, the two types of grains are most easily viewed as grains with two different volumes. In a polycrystalline film there is a distribution of grain volumes. However, we are considering only two for illustrative purposes.