Positive exchange biasing in GdFe/NiCoO bilayers with antiferromagnetic coupling

D. Z. Yang, 1, 4 J. Du, 2, 3 L. Sun, 3 X. S. Wu, 3 X. X. Zhang, 2 and S. M. Zhou 1, 3, *

1 Surface Physics Laboratory (National Key Laboratory) and Department of Physics, Fudan University, Shanghai 200433, People’s Republic of China
2 Department of Physics, Hong Kong University of Science & Technology, Kowloon, Hong Kong, People’s Republic of China
3 State Key Laboratory of Solid State Microstructures, Nanjing University, Nanjing 210093, People’s Republic of China
4 Department of Physics, Southeast University, Nanjing 210096, People’s Republic of China

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For GdFe/NiCoO bilayers after field cooling, hysteresis loops at low temperatures are shifted along both the horizontal and vertical axes. The exchange field \( H_E \) changes from negative values to positive values with increasing cooling field \( H_C \) and the coercivity \( H_C \) acquires a maximum near the crossover of \( H_E \). At 5 K, \( H_E \) and \( H_C \) at \( H_C = 3 \) T and the peak height \( \Delta H_C \) are proportional to the inverse GdFe layer thickness. At \( H_C = 3 \) T, \( H_E \) is always positive in the temperature region from 5 to 350 K. At low \( H_C \), however, \( H_E \) is negative at low temperatures and becomes positive at high temperatures. These results can be ascribed to antiferromagnetic coupling between GdFe and NiCoO layers.

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Positive exchange biasing (EB) has first been observed in a variety of antiferromagnet (AF)/ferromagnet (FM) bilayers with AF materials fluorides, in which the FM and AF layers were argued to have antiferromagnetic coupling at the interface.\(^1\) It has been detected recently in ferrimagnet/ferrimagnet, FM/ferrimagnet bilayers with antiferromagnetic coupling and other systems.\(^2\)–\(^7\) These bilayers have two additional distinguished features. First, the exchange field \( H_E \) has a crossover from negative values to positive values at a critical value of the cooling field \( H_C^{\text{sat}} \). Second, the coercivity \( H_C \) has a maximum at \( H_C^{\text{peak}} \), in addition to the normal enhancement due to the EB.\(^8\) Apparently, the EB strongly depends on the magnitude of the cooling field \( H_C \). It is quite different from conventional FM/AF bilayers with ferromagnetic coupling, in which \( H_E \) and \( H_C \) are almost independent of \( H_C \) if it is larger than the saturation field of the FM layer.\(^9\)\(^,\)\(^10\) In the strict sense, the positive EB has been found only in a few FM/AF systems.\(^2\)–\(^7\) Therefore, extensive studies on the mechanism of the positive EB have been hindered and more experiments are required.

In this paper, we will study the EB phenomena in FM/AF bilayers by using GdFe\((\text{Gd}_{15}\text{Fe}_{55})/\text{NiCoO}(\text{Ni}_{30}\text{Co}_{50})\) bilayers, where NiCoO and GdFe are typical AF material and ferrimagnetic alloys, respectively.\(^2\)\(^,\)\(^11\) The atomic magnetic moment of Gd comes from spin and orbital angular momentums and both of them are parallel to the atomic magnetic moment. The atomic magnetic moment of Fe is contributed only from the spin angular momentum because the orbital angular momentum is almost quenched. Due to antiferromagnetic coupling between the spins of Gd and Fe, the atomic magnetic moment of Gd is aligned antiparallel to that of Fe and the macroscopic magnetization of Gd\(_{15}\)Fe\(_{55}\) alloys is parallel to that of Gd atoms since the latter is dominant. More remarkably, the magnetizations of NiCoO and GdFe layers can have antiferromagnetic coupling at the interface when the contribution of Gd atoms is larger than that of Fe atoms. This is because the atomic magnetic moment of Gd should also be coupled to Co and Ni atoms in AF bilayers antiferromagnetically at the interface. The compositions of the NiCoO and GdFe layers are selected so that the Néel temperature of the AF layer is lower than the Curie temperature of the GdFe layer and FM and AF layers are coupled antiferromagnetically, where the Néel temperature of the NiCoO layer is 400 K and the Curie temperature of the GdFe layer is 430 K.\(^11\)\(^–\)\(^13\)

A large specimen of GdFe/NiCoO (20 nm) bilayer was deposited on Si(100) at ambient temperature by magnetron sputtering system. The base pressure was 2 \times 10^{-7} Pa and the Ar pressure 0.33 Pa during deposition. GdFe and NiCoO layers were made from GdFe and NiCoO composite targets by dc and rf sputtering, respectively. In experiments, small Gd pieces were put on Fe target and small pieces of CoO on NiO target to form GdFe and NiCoO composite targets. The growth rates of GdFe and NiCoO layers were 0.1 and 0.04 nm/s, respectively. The FM layer has a wedge shape to alleviate the run-to-run variation. During deposition, a magnetic field of about 130 Oe was applied parallel to the film plane and along the wedge direction to induce an in-plane uniaxial anisotropy in the FM layer. To analyze the magnetic properties, in-plane hysteresis loops were measured by superconductivity quantum interference device. Before measurements, a large specimen was cut into small pieces along the wedge direction. Each sample was heated to 370 K and cooled to 5 K under an external magnetic field and hysteresis loops were then measured during warming up. During field cooling and measurements, the external magnetic field is aligned along the wedge direction. The compositions of NiCoO and GdFe layers were analyzed by x-ray fluorescence. Structural characterization was carried out by x-ray diffraction. GdFe layers are amorphous and NiCoO layers polycrystalline with preferred (002) orientation.

Figure 1 shows typical hysteresis loops of GdFe(15 nm)/NiCoO(20 nm) bilayers at 5 K, where \( H_{CF} = 3 \) T, 0.5 T, and 150 Oe. One can find that hysteresis loops are shifted away from the zero magnetic field. \( H_E \) is positive for \( H_{CF} = 3 \) T and negative for \( H_{CF} = 150 \) Oe. Moreover, \( H_C \)
for \( H_{\text{CF}} = 150 \text{ Oe} \) is larger than that of \( H_{\text{CF}} = 3 \text{ T} \). Remarkably, the hysteresis loop has a prominent asymmetry for \( H_{\text{CF}} = 3 \text{ T} \) and the asymmetry becomes weak as \( H_{\text{CF}} \) is decreased. Figure 2(a) shows the variation of \( H_E \) at 5 K as a function of \( H_{\text{CF}} \) for GdFe/NiCoO bilayers with different FM thickness \( t_{\text{FM}} \). With increasing \( H_{\text{CF}} \), \( H_E \) for all samples changes sharply from negative to positive at small \( H_{\text{CF}} \) and finally approaches to saturate. The critical value \( H^0_{\text{CF}} \) for the crossover does not significantly with \( t_{\text{FM}} \). Figure 2(b) shows that for all \( t_{\text{FM}} \), \( H_C \) increases sharply with initially increasing \( H_{\text{CF}} \) and reaches a maximum. Finally, it decreases to reach a constant as \( H_{\text{CF}} \) is further increased. Fortunately, the maximum \( H_C \) is located near \( H^0_{\text{CF}} \). At \( H_{\text{CF}} = 3 \text{ T} \), \( H_E \) and \( H_C \) increase with decreasing \( t_{\text{FM}} \) as a result of the interfacial nature of the exchange biasing in the GdFe/NiCoO bilayers.

Figure 3 shows \( H_E \) and \( H_C \) at 5 K and \( H_{\text{CF}} = 3 \text{ T} \) for GdFe/NiCoO (20 nm) bilayers as a function of \( t_{\text{FM}} \). \( H_E \) is approximately proportional to \( 1/t_{\text{FM}} \), demonstrating an interfacial nature of the EB, as shown in Fig. 3(a). According to the slope of the curve \( H_E \) vs \( 1/t_{\text{FM}} \), the exchange coupling energy can be calculated. For GdFe/NiCoO bilayers, it is 0.06 erg/cm². Since \( H_C \) of corresponding free single layer films is as small as few oersteds (not shown), the enhanced \( H_C \) of bilayers is proportional to \( 1/t_{\text{FM}} \), as shown in Fig. 3(a). From Fig. 3(a), one can know that the right coercivity of the hysteresis loop \( H_{C1}(=H_E + H_C) \) changes sharply with \( t_{\text{FM}} \) while the left coercivity \( H_{C2}(=H_E - H_C) \) is almost independent of \( t_{\text{FM}} \). In this way, one can know that for the ascent branch of the wedged FM layers the magnetization reversal process is accompanied by a motion of a single domain wall while for the descent branch it is accompanied by the nucleation of the multidomains and motion of the domain walls. The right and left branches of the loop are thought to have different magnetization reversal process.\(^{14,15}\) In Fig. 2, we define the \( \Delta H_C = H_C^{\text{max}} - H_C^{\text{min}} \), where \( H_C^{\text{max}} \) and \( H_C^{\text{min}} \) are the coercivity of the sample near \( H_{\text{CF}} \) and at \( H_{\text{CF}} = 3 \text{ T} \), respectively. At 5 K, \( \Delta H_C \) is found to decrease as \( t_{\text{FM}} \) is increased and the dependence can be approximately fitted by the scale of \( \Delta H_C \approx 1/t_{\text{FM}} \), as shown in Fig. 3(b).

Figure 4 shows the temperature dependence of \( H_E \) and \( H_C \) for GdFe(15 nm)/NiCoO(20 nm) bilayer with two different \( H_{\text{CF}} \). For \( H_{\text{CF}} = 3 \text{ T} \), \( H_E \) is always positive at all temperatures, as shown in Fig. 4(a). It decreases monotonically with increasing temperature and approaches zero near 350 K. \( H_C \) changes with temperature in a similar way. Apparently, the large \( H_C \) is caused by the establishment of the EB. It is of particular interest to find different temperature dependence for small \( H_{\text{CF}} \). As shown in Fig. 4(b), at low temperatures, \( H_E \) is negative and becomes positive at temperatures above

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**FIG. 1.** Typical in-plane hysteresis loops of GdFe(15 nm)/NiCoO(20 nm) bilayers at 5 K, with \( H_{\text{CF}} \) of 3 T (a), 0.5 T (b), and 150 Oe (c).

**FIG. 2.** Dependence of \( H_E \) (a) and \( H_C \) (b) at 5 K on \( H_{\text{CF}} \) for GdFe/NiCoO (20 nm) bilayers with a variety of \( t_{\text{FM}} \), as denoted by the inset numbers.

**FIG. 3.** The \( t_{\text{FM}} \) dependence of \( H_E \) and \( H_C \) (a) and the additional coercivity enhancement \( \Delta H_C \) (b) for GdFe/NiCoO (20 nm) bilayers, where \( H_{\text{CF}} \) is 3 T and temperature is 5 K.
Actually, at negative saturation field \( H \) of 0.25 cm\(^2\), which is equivalent to the magnetic moment of GdFe\(_s\)lar to the observed phenomena in fluoride based bilayers and 1 ML FM or AF layers. 

At least, one can draw two conclusions from the results in Fig. 5. First, \( \Delta m_{AF} \) is not equal to zero. Otherwise, \( m_{AF}(15 \text{ kOe}) \neq m_{AF}(50 \text{ Oe}) \) with FM domain wall or without any FM domain wall. The results in the inset are in agreement with the fact that \( \Delta m_{AF} \) is positive for \( H_{CF} = 15 \text{ kOe} \) and negative for \( H_{CF} = 50 \text{ Oe} \). Second, no domain wall parallel to the film plane exists in the AF layers. This is because \( \Delta m \) is about the magnetic moment of 1 ML AF layer and equivalent to \( \Delta m_{AF} \). Therefore, \( \Delta m_{FM} \) must be equal to zero and thus no FM domain wall parallel to the film plane exists in the GdFe layer. In a word, the so-called hybrid domain wall model can be excluded in the explanations of the present results.\(^2\)

The evolution of the exchange field with \( H_{CF} \) in Fig. 2 can be explained as a result of the competition between antiferromagnetic coupling energy of FM and AF spins and the Zeeman energy of the AF spins with the external magnetic field \( H_{CF} \). For a large/small \( H_{CF} \), the AF spins are aligned parallel/antiparallel to \( H_{CF} \) and the FM spins. After field cooling procedure, the AF spins at low temperatures are of a meta-stable/stable state, i.e., in high/low energy state. Therefore, the positive/negative \( H_E \) can be induced and there should be a critical value \( H_{CF}^0 \) for the crossover of \( H_E \). It can be estimated as follows:\(^3\)

- \( \Delta m = \Delta m_{AF} + \Delta m_{FM} \)

250 K. \( H_C \) increases slightly with rising temperature and acquires a maximum near 250 K, i.e., the same temperature for the crossover of \( H_E \).

We have carefully compared the hysteresis loops of GdFe(6.2 nm)/NiCoO(20 nm) bilayers at different \( H_{CF} \) in Fig. 5. One can find that when \( H_{CF} \) is 50 Oe and 15 kOe, the exchange field is \(-14\) and 39 Oe, respectively. More remarkably, the inset shows \( m_{AF}(+H)(15 \text{ kOe}) > m_{AF}(+H)(50 \text{ Oe}) \). Actually, at negative saturation field \( m_{AF}(-H)(15 \text{ kOe}) < m_{AF}(-H)(50 \text{ Oe}) \), that is to say, the hysteresis loop with positive EB is shifted towards positive magnetization axis, in comparison with negative EB. For \( H_{CF} = 15 \text{ kOe} \), the positive shift amount \( \Delta m \) is about \( 3 \times 10^{-6} \text{ emu} \) for a sampling area of 0.25 cm\(^2\), which is equivalent to the magnetic moment of 1 ML FM or AF layers.

All the above results for GdFe/NiCoO bilayers are similar to the observed phenomena in fluoride based bilayers and those of GdFe/TbFe bilayers.\(^1,2,8,16,17\) Below we will analyze the above experimental results and reveal the mechanism behind them. Few models were proposed to explain the positive EB in Fe/FeF\(_2\) and Fe/MnF\(_2\) bilayers.\(^18–21\) The evolution of \( H_E \) and \( H_C \) with \( H_{CF} \) was thought to originate from the competition between the Zeeman energy of the AF spins in an external magnetic field and antiferromagnetic coupling. In Gd–Fe/Tb–Fe bilayers with antiferromagnetic coupling, however, the positive \( H_E \) was explained in terms of the hybrid domain wall near interface.\(^2\)

It is instructive to first analyze the vertical magnetization shift at different \( H_{CF} \) for GdFe/NiCoO bilayers. In general, the hysterisis loops of free FM layer films should be centered about the horizontal axis. For the present GdFe/NiCoO bilayers, however, the hysterisis loop is shifted along the vertical axis because of two possible reasons. First, a domain wall might be formed in the GdFe layer parallel to the film plane, which was argued to exist in ferrimagnet/ferrimagnet bilayers.\(^2\) For positive EB, the domain wall will be formed at large positive magnetic field and thus the magnitude of the magnetization at positive saturation magnetic field is reduced, that is to say, \( |m_{FM}(+H)| < |m_{FM}(-H)| = m_{FM}(\text{saturation}) \) and vice versa for negative EB, \( |m_{FM}(-H)| < |m_{FM}(+H)| = m_{FM}(\text{saturation}) \). Therefore, the vertical shift \( \Delta m_{FM} \) is negative and positive for positive and negative EB, respectively. Second, an additional small magnetic moment \( \Delta m_{AF} \) might be contributed from the AF layers, which cannot be altered within the measuring magnetic fields.\(^17\) For positive and negative EB, \( \Delta m_{AF} \) has positive and negative signs and thus the hysteresis loop should be shifted towards positive and negative magnetization axis, respectively. The total vertical shift of the hysteresis loops consists of two parts and \( \Delta m = \Delta m_{AF} + \Delta m_{FM} \).

At least, one can draw two conclusions from the results in Fig. 5. First, \( \Delta m_{AF} \) is not equal to zero. Otherwise, \( m_{AF}(+H)(15 \text{ kOe}) = m_{AF}(+H)(50 \text{ Oe}) \) with FM domain wall or without any FM domain wall. The results in the inset are in agreement with the fact that \( \Delta m_{AF} \) is positive for \( H_{CF} = 15 \text{ kOe} \) and negative for \( H_{CF} = 50 \text{ Oe} \). Second, no domain wall parallel to the film plane exists in the FM layers. This is because \( \Delta m \) is about the magnetic moment of 1 ML AF layer and equivalent to \( \Delta m_{AF} \). Therefore, \( \Delta m_{FM} \) must be equal to zero and thus no FM domain wall parallel to the film plane exists in the GdFe layer. In a word, the so-called hybrid domain wall model can be excluded in the explanations of the present results.\(^2\)

The evolution of the exchange field with \( H_{CF} \) in Fig. 2 can be explained as a result of the competition between antiferromagnetic coupling energy of FM and AF spins and the Zeeman energy of the AF spins with the external magnetic field \( H_{CF} \). For a large/small \( H_{CF} \), the AF spins are aligned parallel/antiparallel to \( H_{CF} \) and the FM spins. After field cooling procedure, the AF spins at low temperatures are of a meta-stable/stable state, i.e., in high/low energy state. Therefore, the positive/negative \( H_E \) can be induced and there should be a critical value \( H_{CF}^0 \) for the crossover of \( H_E \). It can be estimated as follows:\(^3\)
where $S_{Fe}$ is the spins of Fe atoms near interface, $S_{Gd}$ the spins of Gd atoms, and $S_{AF}$ the AF spins. $J_1$ and $J_2$ are the exchange interaction constants of Fe and Gd spins with AF spins, respectively. $M_{AF}$ is the net magnetization of the AF material, which was observed before in fluoride based FM/AF bilayers.\textsuperscript{17} Apparently, above equation shows that $H_{CF}$, is independent of the FM and AF layer thickness, as observed in Fig. 2. Since $J_1$ and $J_2$ have opposite signs, $H_{CF}$ in the GdFe/NiCoO and GdFe/TbFe bilayers is much smaller than those of Fe/FeF$_2$ and Fe/MnF$_2$ bilayers.\textsuperscript{1,2}

Since a multi-domain structure in the AF layer is formed and then fixed after field cooling procedure, an additional pinning effect of the AF domain walls on the motion of the FM domain wall exists,\textsuperscript{8,22} and $\Delta H_C$ can be expressed in terms of the domain size in FM and AF bilayers as follows:

$$\Delta H_C = \frac{J_{eff} - L_{FM}}{t_{FM} M_{FM} d_{AF} d_{FM}^2},$$

where $J_{eff}$ is the effective interaction between FM and AF layers and $a_0$ is the interfacial atomic separation in the FM layer. $L_{FM}$ is the length of the FM domain wall under consideration and $d_{AF}$ the domain size of the AF layer. Since the number of the AF domain has a maximum and thus $d_{AF}$ has a minimum at $H_{CF}^0$, the $\Delta H_C$ has a maximum near the crossover. As the FM layer has a wedge shape, the motion of a single domain wall occurs during magnetization reversal process, which is perpendicular to the wedge direction. Therefore, $L_{FM}$, taking the sample size perpendicular to the wedge direction (about 3 mm), is independent of $t_{FM}$. $\Delta H_C$ should be inverse proportional to $t_{FM}$. In a word, one can find that Eq. (2) can be used to explain the results in Figs. 2(b) and 3(b).

The evolution of $H_E$ with temperature at different $H_{CF}$ in Fig. 4 can be explained as follows. First, in FM/AF bilayers with antiferromagnetic coupling, the change in the sign of $H_E$ comes from the competition of the Zeeman energy of the AF spins in the external magnetic field and the antiferromagnetic coupling between the FM and AF layers.\textsuperscript{21} Second, at low temperatures the exchange biasing can be produced not only by the effect of the field cooling through the Néel temperature of the AF layers, but also by the accumulation effect of field cooling from temperatures lower than the Néel temperature to the sampling temperature.\textsuperscript{24} This is a so-called memory effect. In the present work, the exchange biasing was established by a field cooling procedure from 370 K, which is lower than the the Néel temperature of the AF layer. At very small $H_{CF}$, the Zeeman energy cannot overcome the antiferromagnetic coupling at any temperature in the region presented here, and thus the AF and FM spins are aligned antiparallel to each other. $H_E$ is negative at all temperatures. Similarly, one can understand the reason that $H_E$ is always positive at the whole measuring temperature region for high $H_{CF}$. At an intermediate $H_{CF}$, the coupling energy is larger than the Zeeman energy at low temperatures and the former one is smaller than the latter at high temperatures since the interfacial exchange coupling energy weakens at high temperatures. Therefore, $H_E$ is negative and positive at low and high temperatures, respectively.

In summary, we have prepared GdFe/NiCoO (20 nm) bilayers with a wedged FM layer and studied the EB as a function of $H_{CF}$, temperature, and the GdFe layer thickness. As $H_{CF}$ is increased, $H_E$ changes from negative values to positive values with a crossover at $H_{CF}^0$ and finally approaches saturation, and $H_C$ acquires an additional enhancement near $H_{CF}^0$. $H_E$ and $H_C$ for $H_{CF}=3$ T, and $\Delta H_C$ are proportional to the inverse GdFe layer thickness. At $H_{CF}=3$ T, $H_E$ is positive in the entire temperature region. At low $H_{CF}$, however, $H_E$ is negative at low temperatures and becomes positive at high temperatures. For GdFe/NiCoO bilayers, the magnetization shift is positive and negative for positive and negative EB, respectively. It is proposed that there is no FM domain wall parallel to the film plane during FM magnetization reversal process. The present results can be attributed to a competition between the antiferromagnetic coupling at the GdFe/NiCoO interface and the Zeeman energy of the AF spins in the $H_{CF}$.

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\textsuperscript{8} Electronic address: shimingzhou@yahoo.com


