Training-Induced Positive Exchange Bias in NiFe/IrMn Bilayers

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Positive exchange bias has been observed in the $Ni_{81}Fe_{19}/Ir_{20}Mn_{80}$ bilayer system via soft-x-ray resonant magnetic scattering. After field cooling of the system through the blocking temperature of the antiferromagnet, an initial conventional negative exchange bias is removed after training, i.e., successive magnetization reversals, resulting in a positive exchange bias for a temperature range down to 30 K below the blocking temperature (450 K). This new manifestation of magnetic training is discussed in terms of metastable magnetic disorder at the magnetically frustrated interface during magnetization reversal.

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The exchange bias in a ferromagnetic (FM)–antiferromagnetic (AFM) system was first discovered by Meiklejohn and Bean [1] in Co particles encapsulated by a shell of antiferromagnetic CoO. For more than 60 years this effect has been extensively studied, mainly due to the elusiveness of a fundamental understanding and its value for applications such as ultrahigh-density magnetic recording, giant magnetoresistance (GMR), and spin valve devices [2]. When a sample with a magnetically uncompensated FM/AFM interfaces is cooled through the Néel temperature (T_N) of the AFM, with the Curie temperature (T_C) of the FM being higher than T_N, an unidirectional exchange anisotropy, namely, exchange bias (EB) is induced in the system [3–5].

Usually the exchange bias direction is opposite (negative EB) to the FM magnetization direction during field cooling. The reverse situation, namely, a shift of the hysteresis loop to positive direction (positive EB) with respect to field cooling directions occurs too. Positive exchange bias (PEB) was first observed in FeF_2/Fe bilayers and was associated with antiferromagnetic interfacial coupling [6] which was recently observed experimentally [7,8]. In these systems the magnitude and sign of EB depend strongly on strength and direction of the cooling field (H_{CF}) [6]. This is in agreement with the results obtained by Leighton *et al.* for MnF_2/Fe bilayers [9]. Beckmann and Usadel [10] found using Monte Carlo simulations that a directional variation of H_{CF} can even result in different asymmetric magnetization reversal modes of the FM. It was also reported that a diluted AFM order at the interface may enhance the EB [11], and that the spin alignment at FM/AFM interfaces [12] depends on their roughness.

Another category of PEB has been observed for instance in $Cu_{1-x}Mn_x$/Co and CoO/Co bilayers, where PEB is established only in the proximity of the blocking temperature, T_B, [13–16] after field cooling. Further lowering of the sample temperature results in negative exchange bias. The microscopic mechanism of the PEB close to T_B is discussed on the basis of coexistence of FM and AFM interface coupling [14], interfacial RKKY coupling for the $Cu_{1-x}Mn_x$/Co bilayer [16], and unidirectional coercivity enhancement [13,15]. In general, the phenomena of PEB close to T_B is only observed for finite magnetic bilayer thicknesses while the microscopic origin, the impact of H_{CF} and competing coupling mechanisms at the interface are still debated.

Here we report on a new manifestation of exchange bias in IrMn-based EB bilayers, which is one of the most attractive AFM material for both device applications and fundamental research [17–23]. We show that PEB in the $Ni_{81}Fe_{19}/Ir_{20}Mn_{80}$ bilayers is induced in our samples only after several training cycles near T_B. This is different from all previous cases where PEB is already observed for the very first hysteresis loop after field cooling. Moreover, a new type of asymmetric magnetization reversal can be inferred by analyzing the shape of the hysteresis loop. The experimental results are discussed in the framework of frustration at the magnetically disordered AFM/FM interface.

A polycrystalline Si(100)/Cu(5 nm)/$Ni_{81}Fe_{19}$(7.5 nm)/$Ir_{20}Mn_{80}$(3.5 nm)/Cu(2.5 nm) sample was grown by magnetron sputtering on a Si wafer at a base pressure of 8.3×10^{-9} mbar. Ultraclean Ar gas was used as the sputtering medium. The partial Ar pressure during growth was 1.5×10^{-3} mbar. An uniaxial magnetic anisotropy was induced in the FM layer by applying an in-situ magnetic field of 2 KOe parallel to the sample surface. A Cu (5 nm) buffer layer was deposited to promote a smooth growth of the magnetic heterostructure. The subsequent $Ni_{81}Fe_{19}$ (Py) and $Ir_{20}Mn_{80}$ layers were capped with Cu (2.5 nm) to prevent oxidation of the heterostructure.

X-ray resonant magnetic scattering measurements were performed at BESSY II using the ALICE diffractometer [24] installed at the BESSY II PM3 bending magnet beamline. Magnetic hysteresis loops for the ferromagnetic layer were measured by tuning the x-ray energy at the Ni L_3 absorption edge and monitoring the specularly reflected x-ray intensity as a function of magnetic field applied parallel to the sample surface and in the scattering plane.

Maximum magnetic sensitivity, i.e., asymmetry, $(I^+ - I^-)/(I^+ + I^-)$, (where $I^\pm$ are reflected intensities for opposite magnetic field directions) was achieved by utilizing 80% circularly polarized x rays in specular condition at an incident angle equal to $\theta = 9.9°$.

Figure 1(a) shows the magnetic training effect observed for the Py(7.5 nm)/$Ir_{20}Mn_{80}$(3.5 nm) sample at 10 K after field cooling from $T = 470$ K through the blocking temperature, $T_B = 450$ K. The first hysteresis loop exhibits a sharp reversal at H_{c1} (the coercive field at the very first reversal) and a more rounded reversal at H_{c2} (the coercive field at the second reversal). By measuring a second hysteresis loop we observe a decrease of the exchange bias field, $H_{\rm EB} = (H_{c1}\text{-}H_{c2})/2$, which is characteristic for a training effect. Strikingly, the second hysteresis loop exhibits the same steep/rounded characteristic features at H_{c1}/H_{c2} as the first one. Even after 14th hysteresis a sharper reversal at H_{c1} as compared to H_{c2} is preserved. This training effect is different as the one observed for the archetypal Co/CoO EB bilayer [13,25–27] and also for IrMn/CoFe bilayers [23]. There, at the very first reversal after field cooling a transition from an essentially single AFM domain to a multidomain state occurs which leads to a transition from a pronounced asymmetric hysteresis to an essentially symmetric one [25]. The shape of the hysteresis at the second reversal was more rounded, and in contrast to our current observation, the consecutive hysteresis loops remain essentially rounded at both hysteresis loop branches. Such a transition to a symmetric hysteresis behavior is characteristic for changes in the bulk AFM domain structure. Since we do not observe this behavior in our system we believe that the AFM bulk spin structure is robust, lacking a dramatic change of the spin configurations which would naturally lead to a pronounced change of loop asymmetry. Also, higher orders anisotropy of the AFM layer [28,29] may not be the main origin of this type of training effect, since the AFM layer exhibits an uniaxial anisotropy, as confirmed by azimuthal dependence of the coercive fields (data not shown).

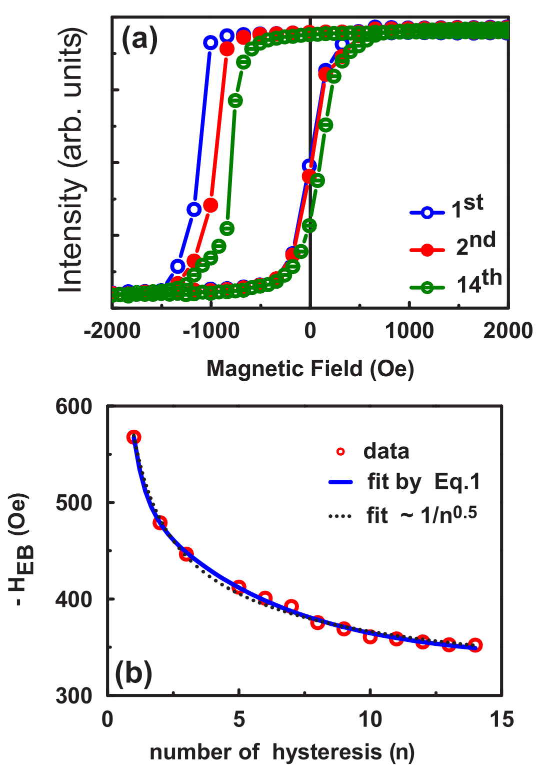

FIG. 1 (color online). (a) Magnetization curves measured at 10 K after field cooling ($H_{\rm FC} = 2$ K Oe) the system from 470 K through the blocking temperature. The 1st (blue), 2nd (red) and 14th (green) hysteresis loops are shown. (b) $H_{\rm EB}$ as a function of the loop index (n) extracted from individual hysteresis loops. Open circles are the experimental data, the line represents a model, and the dotted line is a $1/\sqrt{n}$ functional fit.

In order to explain the asymmetry of the magnetization curve in Fig. 1(a) we assume that the AFM layer behaves virtually as predicted by the Meiklejohn and Bean (MB) model. Only when the AFM thickness is slightly larger than a critical value, the AFM spins rotate reversible away from their stable angular orientations set by a field cooling procedure. During the magnetization reversal, the AFM spin direction acquires a maximum value of 45° at the critical thickness [5]. When this angular deviation is significant, an asymmetric magnetization reversal occurs [5]. Note that the condition of being close to the critical thickness regime is realized in our system. The critical AFM thickness for exchange bias was measured (data not shown) and is about 2 nm. Using a modified MB model named spin glass (SG) model [5] this asymmetry of the hysteresis loop is reproduced numerically at reduced thicknesses. An enhanced coercivity is also accounted for by assuming a magnetically disordered interface (see Fig. 3.39 in [5]).

The stability of the bulk AFM structure is seen also when plotting the exchange bias field, $H_{\rm EB}$, as a function of hysteresis loop index n in Fig. 1(b). We find that the EB field as a function of n can be described by the following empirical law [30]: $|H_{\rm EB}| = |H_{\rm EB}^\infty| + k/\sqrt{n}$, with $H_{\rm EB}^\infty = 272 \pm 4$ Oe and $k = 297 \pm 7$ Oe. A less significant decrease of the EB takes place between the first and the second hysteresis cycles, suggesting that no AFM domains (rearrangements) occur. A monotonous evolution of the coercivity and $H_{\rm EB}$ as a function of n appears due to the interfacial spin rearrangement at the magnetically disordered FM/AFM interface. The presence of interfacial spin frustration can enhance the interface area remarkably while keeping the total spin number preserved. At the FM/AFM interface the AFM magnetic anisotropy is assumed to be modified, leading to essentially two different types of AFM uncompensated spins after field cooling: namely, frozen and rotatable AFM uncompensated spins being rigidly exchange coupled to the AFM and FM layers, respectively [5].

With each cycle a spin rearrangement takes place and this modifies the coercive and exchange bias fields. Note that our approach is different as the one of Binek [31], although the main concept of interfacial magnetic instabilities is preserved. While Binek considers only a change of the interfacial AFM magnetization, we suggest that both components, frozen and rotating are affected by the FM magnetization reversals. Moreover, mixed ferromagnetic and antiferromagnetic coupled components will contribute distinctively, through different relaxations rates, to the training effect.

Additional evidence for this scenario can be obtained by describing the data in Fig. 1(b) with the following expression to simulate the relaxation of the exchange bias as a function of n:

$$H_{\rm EB}^{n} = H_{\rm EB}^{\infty} + A_f \exp(-n/P_f) + A_i \exp(-n/P_i), \quad (1)$$

We note that it is not possible to describe the curve in Fig. 1(b) by only one exponential. Here, $H_{\rm eb}^{n}$ is the exchange bias of the nth hysteresis loop, A_f and P_f are parameters related to the change of the frozen spins, A_i and P_i are evolving parameters of the interfacial magnetic frustration of the bilayer. The A parameters have dimension of magnetic field (Oersted) while the P's are dimensionless parameters and resemble a relaxation time, where the continuous variable is replaced by a discrete variable, namely, the hysteresis index n. The parameters obtained from fit to the $H_{\rm EB}$ data are $H_{\rm EB}^{\infty} = 335 \pm 5$ Oe, $A_f = 641 \pm 527$ Oe, $P_f = 0.44 \pm 0.18$, $A_i = 199 \pm 13$ Oe, and $P_i = 5 \pm 0.6$.

Indeed, within the SG approach, we distinguish a sharp contribution due to uncompensated spins at the interface and a much weaker decrease from the frozen uncompensated spins. The frozen component appears to relax about 10 times slower as compared to the other one.

For the remainder of this Letter we demonstrate the importance of magnetic training for establishing PEB. The temperature dependence of the $H_{\rm EB}$ and H_c for the bilayer is shown schematically in Fig. 2. The data were obtained by field cooling the sample in an external magnetic field of about 2 KOe from $T = 470$ K through the blocking temperature at each temperature shown in Fig. 2. Even up to the highest available temperatures in our experimental setup (470 K) the measured coercivity values were higher than that of a single Py layer [$H_c(P_y) \approx 1$ Oe] [see Fig. 3(a)]. This indicates that the Néel temperature of the AFM film was not reached. The data displayed in Figs. 2(a) and 2(b) were obtained from hysteresis loops of the freshly biased system (red lines and solid symbols) and after training via 30 hysteresis sweeps (blue lines and open symbols). The hysteresis loops in Fig. 2(a) display a roughly linear increase of $H_{\rm EB}$ with decreasing temperature.

Interestingly, the exchange bias after training is almost rigidly shifted to lower values over the whole measured temperature region. This can be clearly seen in Fig. 2(c) where the difference in exchange bias, $\Delta H_{\rm EB} = H_{\rm EB}(n = 1) - H_{\rm EB}(n = 30)$, before and after training is shown (blue lines and solid symbols). This decrease of the EB after training appears to contribute to the PEB near T_B [see the inset of Fig. 2(a)]. However, the size of the PEB is actually larger than that expected from the rigid shift at lower temperatures. Another new observation here is that the PEB occurs only after training. For instance, at $T = 435$ K a typical negative exchange bias extracted from the very first hysteresis cycle after cooling has changed sign, resulting in PEB after training.

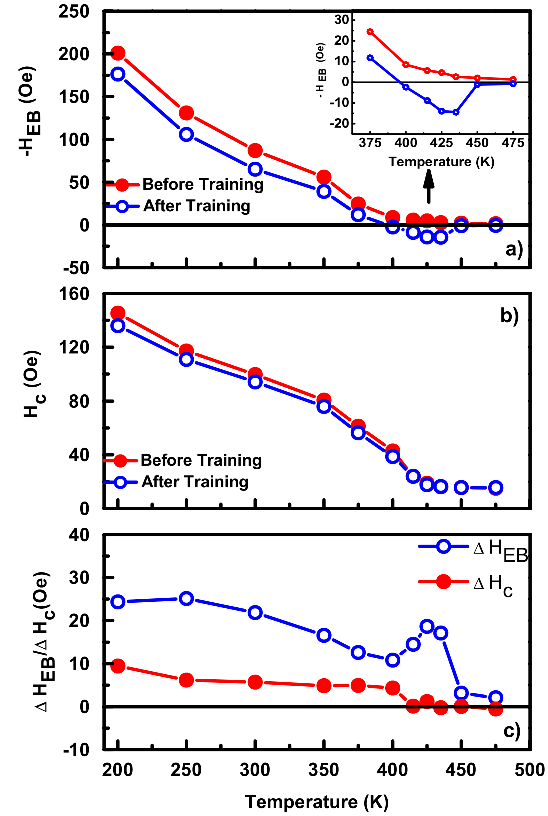

FIG. 2 (color online). Temperature dependence of the (a) $H_{\rm EB}$ for the first (red filled circles) and the last loop (blue open circle), (b) H_c for the first (red filled circles) and the last loop (blue open circles), and (c) difference of magnitude (before and after training) for $H_{\rm EB}$ and H_c fields of the FM layer. The first loop is measured right after field cooling whereas the last loop was measured after fast flipping of the magnetic field. The inset in (a) is an enlargement at the blocking temperature showing the positive exchange bias.

We can discriminate between several possible mechanisms leading to PEB. We observe a rather constant temperature dependence for the $\Delta H_{\rm EB}$ in Fig. 2(c). A dominant RKKY origin [16] of the PEB would lead to a nonmonotonic temperature dependence for the $\Delta H_{\rm EB}$, which is not observed. Also, an unidirectional enhancement of coercivity as a main reason for an apparent PEB [13,15] is not fully supported by our data. A rotation of bulk AFM grains or domains would have to be suppressed at low temperatures. Our data shows no significant exchange bias and coercivity variation ($\Delta H_{\rm EB}$ and ΔH_c) across the T_B and closely below it. As a result we are lead to the conclusion

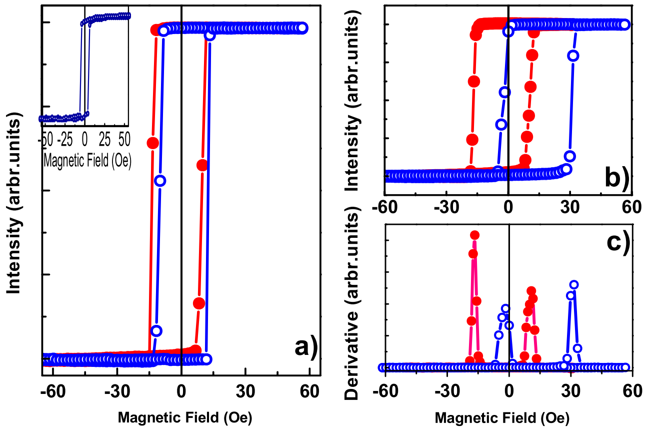

FIG. 3 (color online). The MH loops of the FM for two representative temperatures $T = 450$ K (a) and $T = 435$ K (b), respectively, measured before (red filled circle) and after (blue open circle) training. The system has been field cooled ($H_{FC} = 2$ KOe) from $T = 470$ K. The inset shows the hysteresis of a $Ni_{81}Fe_{19}$(7.5 nm) layer in the absence of a IrMn layer. In (c) the derivative of the hysteresis loops [of (b)]. The asymmetry of each hysteresis and its reversal are clearly seen as a different amplitude at the coercive fields when comparing the ascending and the descending branches.

that irreversible changes at the interface are responsible for the PEB.

In order to explain the occurrence of PEB we assume a simple model based on the previous description of PEB [6,14], where an uncompensated spin component exhibiting a fundamentally antiparallel coupling to the FM is needed. During the field cooling procedure a minority of the uncompensated interfacial AFM spins will prefer to align antiparallel to the direction of the FM layer defined by the cooling field. This situation would result in a typical negative EB [6]. After training, this minority component will rotate irreversibly due to consecutive magnetization reversals of the FM acting on a frustrated spin state. This frustrated spin state is a consequence of symmetry breaking at the FM/AFM interfaces [32,33]. As a result, a weak positive shift of the hysteresis loop will occur at all temperatures after training. When, cooling from above T_B, this component will dominate strongly right below the blocking temperature since it is anchored stronger to the bulk side of the AFM layer. Only for lower temperatures, the majority component providing negative exchange bias will lead to a robust exchange bias.

The rotation of this minority component is clearly seen in Figs. 3(b) and 3(c). The untrained hysteresis loop at $T = 435$ K is asymmetric, namely, the first reversal is steeper as compared to the second one (compare the density of field points across the reversals at the coercive fields). After training, this asymmetry reverses, namely, the second reversal becomes steeper. This is a direct proof of that a minority unidirectional anisotropy responsible for the PEB has rotated during field cycling.

In conclusion, we have observed a novel asymmetry of the hysteresis loop which was predicted numerically [5] at the critical region for exchange bias. The training effect leads to irreversible changes of an essentially frustrated interfacial spin state. At the blocking temperature a positive exchange bias occurs after training. A rotation of a minority antiparallel coupled spin component is clearly revealed through the asymmetry reversal of the hysteresis loops. The experimental data allow us to discriminate between different models for the newly observed positive exchange bias, supporting a mixture of antiferromagnetic (minority) a ferromagnetic (majority) coupling mechanism at the interface.

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