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Can uniaxial anisotropy be responsible for training in exchange coupled system?

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Training in exchange bias can be related to the field cooled state of the ferromagnet—*multidomain* or *single domain*. In a system of coexistent states, realized by coercive field cooling, we could observe usual training for the single domain portion while the multidomain portion remains untrained. This crossover state indicates to the fact that antiferromagnetic uniaxial anisotropy can be made responsible for the training in exchange coupled system where no biaxial anisotropy could exist. © *2010 American Institute of Physics*. [doi:10.1063/1.3475699]

I. INTRODUCTION

Interfacial exchange coupling between a ferromagnet (FM) and an antiferromagnet (AF) can "lock" the magnetization into the FM in a well-defined direction. This effect, which in a phenomenological picture takes the form of a unidirectional magnetic anisotropy, is known as exchange bias.[1] The locking predominantly depends upon the state of the interface at which the AF-FM spins are frozen in, as they are field cooled (H_{FC}), provided that the AF domain size remains unchanged with respect to its initial state. An irreversible (pinned) AF interface magnetization arising from the volume domains stabilized by the defects in the AF has been a crucial ingredient in explaining the origin of the effect.[2] While the coercivity enhancement has been attributed to uncompensated spins in the AF grains that are with rotatable magnetization within the AF (ferromagnetically coupled),[3,4] the nonrotatable moments (antiferromagnetically coupled) leads to exchange bias field along with small vertical shift in magnetization axis (indicating a very small fraction of uncompensated spins involved for bias field). Mysteries of symmetry and asymmetry (during magnetization reversal) is also now understood in terms of interplay of different anisotropic and Zeeman energies in the system.[5]

Another important aspect of exchange bias is the difference in subsequent (partial) magnetization reversal loops which is called the "training effect." It can be classified into two types:[6] one between the first and second loop and another one involving subsequent higher number of loops. The first type has been proposed to arise from the AF magnetic symmetry.[7] Whereas the second type has been demonstrated experimentally as the $H_{EB} \sim (n)^{-1/2}$ (for $n > 2$), where n is the number of loops. This has been suggested to arise from the reconfiguration of the AF moments or domains during the field cycling.[8]

We concentrate here on the AF anisotropy that has been identified as the primary cause of training[7] and was also suggested to be responsible for the usually observed asymmetry in magnetization reversal. The free energy per unit area of the AF-FM bilayer can be written as

$$E = -K_F t_F \sin^2\theta_F - K_{AF} t_{AF} M_{AF} (\sin^2\theta_{B1} + \sin^2\theta_{B2}) - J_E M_{AF-(B1,B2)} \cdot M_{FM} - J_{AF} M_{AF-B1} \cdot M_{AF-B2} - H \cdot M_{FM} t_F - H \cdot M_{AF} t_{AF-(B1,B2)}, \quad (1)$$

where the first term represents the uniaxial anisotropy energy of the FM layer with the anisotropy constant K_F. The second term is the *uniaxial* anisotropy of the AF layer with the anisotropy constant K_{AF}. The third term is the *unidirectional* anisotropy energy characterized by the exchange coupling constant J_E and the fourth term is the coupling constant of the AF sublattice B1 and B2. Finally, the last terms stand for the respective Zeeman energies for filed H. The angles are between the easy-axis directions and FM magnetization and each AF sublattice magnetization, respectively. Due to the exchange coupling, the FM uniaxial anisotropy may also be affected by the AF uniaxial anisotropy in-spite of different ordering temperatures.

Coherent rotation model calculations show training to be originating from symmetries higher than the uniaxial one (biaxial anisotropy) even in highly uniaxial anisotropic systems (e.g., CoO). Biaxial anisotropy can be due to an inherent frustration in the system coupling the two AF sublattice resulting in perpendicular configuration. Uniaxial anisotropy, on the other hand, can be induced during growth which let the AF sublattice parallel to each other. The perpendicular orientation was believed to be recompensable for the training effect which was relaxed after one field cycle.[7]

This scenario was somewhat contradicted by micromagnetic calculations that predicted training (the effect of which is much smaller in magnitude) and symmetric magnetization reversal even for uniaxially anisotropic system.[9] Note that the same calculations—considering biaxial anisotropy—predicted asymmetric reversal and large training (in agreement with Hoffmann[7]).

The question that remains therefore is this: whether

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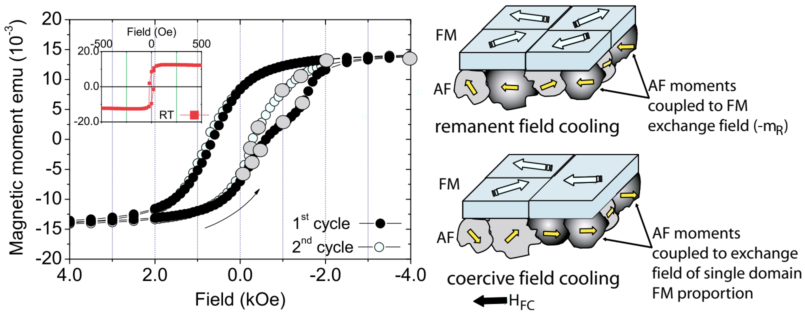

FIG. 1. (Color online) (a) SQUID hysteresis loops of $[Co/CoO/Au]_{16}$ ML during the first (solid symbol) and second field cycles (open symbol) for $H_{FC}=+20$ Oe. The loops correspond to a coercive field cooling state after positive saturation at RT and the arrow along the curve indicates the field sweeping direction. The inset shows the hysteresis loop at RT. The gray dots are the points of field along the curve used in neutron measurements. Schematics represent the state of the FM-AFM layer magnetization after being field cooled at remanent cooling field ($-m_R$) and at $H_{FC}\simeq H_c$.

uniaxial anisotropy can exclusively contribute to training? There are some recent experimental reports on the effect of a crossover from uniaxial to biaxial anisotropy and its effect on training.[10] However, no specific experimental study, which uniquely defines the role of uniaxial anisotropy in training, is known thus far.

It is well known that normal field cooling renders *single domain* state (from saturation state of the FM) and exhibits clear training response. Interestingly, polarized neutron measurements by Paul *et al.* have shown that cooling a system in a remanent state (rendering *remanent* exchange bias), can lead to *microscopic* suppression of training effect. Note that remanent field cooling is different from demagnetized state cooling, even though *multidomain* FM state is realized in both cases. Suppression of training was attributed to poorly defined AF uniaxial anisotropy within the AF layer due to remanent field cooling.[11]

We have considered such a remanent field cooled state as the state where one can freshly introduce an uniaxial anisotropy. This can be done if we chose to cool the system in a small external field which would be close to the coercive field of the specimen. For $H_{FC}\simeq$coercive field of the FM (H_c), a coexistence of two different FM micromagnetic configurations can be realized within the same system: *multidomain* and *single domain*.[12] The portion of the AF layer that is exchange coupled to the multidomain configuration can show suppression of training whereas that coupled to the single domain configuration is expected to show training as usual.

In this study, using depth sensitive polarized neutron scattering, we report on the investigation of such complex magnetic state of the interface (by *simultaneously* analyzing transversal as well as longitudinal components of magnetization) on a microscopic scale. With our unique methodology we could unravel the basic relation between training and uniaxial anisotropy within the AF layer at their onset.

II. EXPERIMENTAL TECHNIQUE

Depth sensitive neutron scattering under grazing incidence with polarization analysis has been proven decisive in the identification of reversal mechanisms in exchange bias systems.[13,14] Due to the interaction between polarized neutrons and magnetic moments in the specimen, polarized neutron reflectivity is sensitive to the in-plane magnetization for a homogeneous film on a *microscopic* scale. Neutron scattering with polarization analysis can discriminate the longitudinal and transverse components of the magnetization. In the experiment four different cross sections can be distinguished, namely, nonspin-flip (NSF): (R_{++} and R_{--}) and spin-flip (SF) channels (R_{+-} and R_{-+}). Here + and − signs are used to distinguish the intensity contributions R representing a polarization component ∥ or anti-∥ to the guiding field, respectively. $R_{++/--}$ contains the sum/difference between the nuclear and magnetic scattering, whereas the SF signal contains only the magnetic information. A magnetization rotation is identified by a significant increase in the specular SF reflectivities, which corresponds to the formation of in-plane magnetization components developing perpendicular to the guiding field H_a, applied collinear to H_{FC}. On the other hand, magnetic reversal by domain nucleation and propagation does not provide enhanced SF specular intensities, as long as the domain sizes are smaller than the lateral projection of the neutron coherence length $l_{\parallel}$ (but can produce off-specular signal in the SF channels), because the local magnetization M is always collinear to H_a. In the specular scattering geometry, normal wave vector transfers $q_{\perp}=2\pi/\lambda[\sin(\alpha_i)+\sin(\alpha_f)]$ are probed while off-specular scattering contributions along the in-plane momentum transfer vector $q_{\parallel}=2\pi/\lambda[\cos(\alpha_f)-\cos(\alpha_i)]$ arise, when the in-plane translational symmetry is broken by interface waviness (roughness) or by magnetic domains on a length scale shorter than $l_{\parallel}$ along $q_{\parallel}$.[15]

We have investigated multilayers (MLs) of the composition $SiO_2/[Co(9.0\ nm)/CoO(7.0\ nm)/Au(25.0\ nm)]_{16}$ with coercive field cooling. Details on similar sample growth and sample characterization (structural and magnetic) has been published earlier.[16] The neutron scattering experiments were performed at the polarized neutron reflectometer with polarization analysis TREFF at the FRM-II for a wavelength of 4.73 Å. In specular geometry, the angle of incidence α_i equals the final angle α_f, and λ is the wavelength from the monochromator. All measurements have been done after the sample was cooled to 10 K from room temperature (RT) by a continuous flow cryostat in presence of a cooling field provided by an electromagnet.

III. RESULTS

A. Hysteresis loops

Superconducting quantum interference device (SQUID) hysteresis loops of the ML during the first and second field cycles for $H_{FC}=+20$ Oe after negative saturation ($-m_R$) are shown in Fig. 1. Schematics show the state of the AF-FM interface after being field cooled at H_c as compared to that at remanent field cooling. At $H_a\approx-500$ Oe, the hysteresis loop

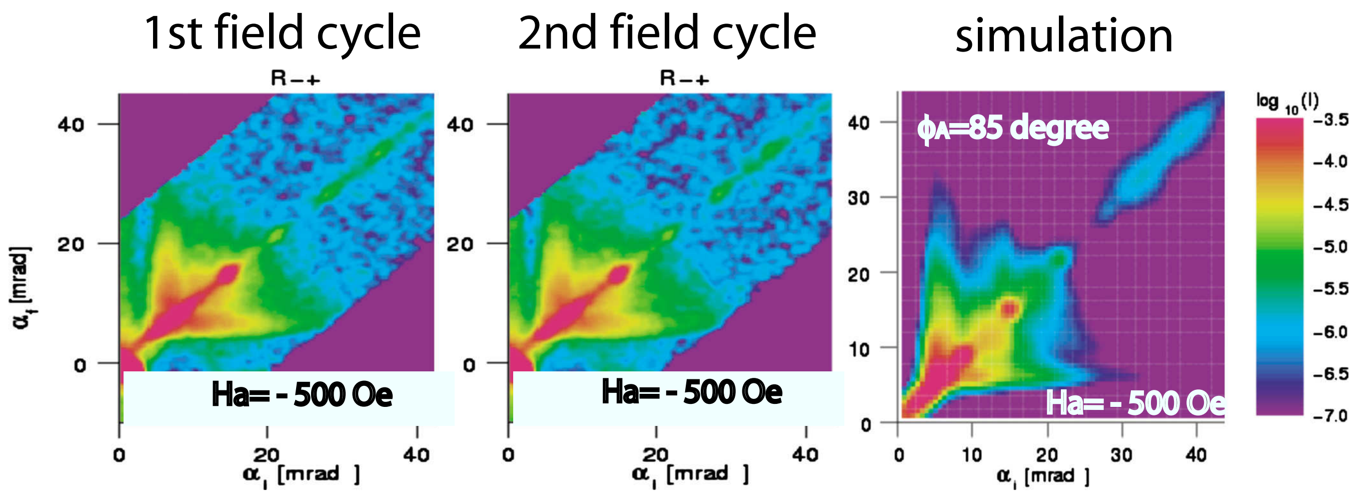

FIG. 2. (Color online) SF intensity maps $[R_{-+}]$ from Co/CoO/Au ML measured at $H_a=-500$ Oe and at 10 K after *positive* saturation. The color bar encodes the scattered intensity on a logarithmic scale. The simulated SF intensity map within DWBA is shown alongside for $\phi_A=85°$.

(first cycle) is divided into upper and lower components—coexistence of two different FM configurations. The lower component resembles the loop after remanent field cooling (multidomain) whereas the upper component resembles the loop after field cooling (single domain). The distinction between upper and lower part of the loop is particularly valid as we see the upper part (component) of the loop is shifted opposite to the cooling field while the lower part is almost unaffected (note that the lower part is similar for the first and second field cycles while the upper part is not).

In the first half of the first field cycle (closed circle), a considerable shift in the upper component of the hysteresis loop ($H_{EB}\sim-700$ Oe) is observed. This is analogous to the usual pinning of the AF spins to the exchange field of the partially saturated FM (but this shift is significantly smaller than that which would have been observed after cooling in a saturated field).[12] The lower component of the loop, on the other hand, shifts toward the opposite side ($H_{EB}\sim -570$ Oe). Here, the AF spins are locally coupled to the initial magnetization ($-m_R$) of the multidomain FM. While the shift of the upper part of the first cycle disappears during the second cycle, the shift in the lower part remains almost similar. As we start our field sweep from the positive saturation, the state of the system during its first half of first field cycle, is not trained while the loop during its second cycle (open circle), is obviously trained.

B. Neutron scattering measurements

The buried domain structure formation after field cooling is evident from the off-specular and specular SF scattering signals shown in Fig. 2.[17] These domains are on a length scale smaller than $l_\parallel$. The intensity maps (concentrated near the total reflection plateau) are simulated within the supermatrix formalism under the distorted wave born approximation (DWBA), considering the magnetization to fluctuate randomly ($\Delta\phi\approx 30°$) from domain to domain around their respective mean magnetization angle ϕ_A obtained from the fitted values of the extracted specular patterns.[15] The domains are estimated as $\approx 1.2\pm 0.2$ μm and are found to be vertically uncorrelated for both field cycles. However, changes in their correlation lengths (for the respective field cycles) are found to be within the errors of estimate.

Specular reflectivity patterns are extracted from the intensity maps. They are shown (closed circle) in Fig. 3 for $H_{FC}=+20$ Oe [taken at seven different H_a values along the decreasing branch of the hysteresis loop indicated by closed gray circles in Fig. 1(a)] together with their least-square fits (open circle) for two consecutive cycles of field.[13,14] The four peaks of the ML in the NSF channels are the respective order of Bragg reflections of the ML. R_{++} dominates over R_{--} for all fields before the reversal. This is related to a net magnetization, which is collinear to H_a during the first and second field cycles.

The angular variation (ϕ_A) in the net magnetization, as deduced from the fits to the specular reflectivity patterns for the first and second field cycles is shown in Fig. 4(a). It is clearly evident that there exists (unlike the remanent field cooled case) substantial training for the system and the reversal mechanism is predominantly via uniform rotation of magnetization. In case there has been a magnetization reversal by domain nucleation and propagation, the magnetic moment would have been drastically reduced. For a reduced magnetic moment, the SF signals which depict the transverse component of magnetization would have been reduced accordingly. The complete set of data was fitted without any change in the magnitude of the magnetic moment. The only parameter that varies in our routine is the magnetization angle. In Fig. 4(b), we plot the differences in ϕ_A for the respective two field cycles $\delta\phi_A=[\phi_A(1^{st}_{cycle})-\phi_A(2^{nd}_{cycle})]$, corresponding to the remanent field cooing (reconstructed from Ref. 11) as well as to the coercive field cooling (H_{FC} $=+20$ Oe). This plot of $\delta\phi_A$ with H_a, eventually gives a measure of the degree of training for $H_{FC}\simeq H_c$. One may recall that for remanent field cooling, we observed suppression of training ($\delta\phi_A\simeq 0$).[11] Here, we observe a clear evidence of training ($\delta\phi_A\neq 0$) beyond 1.0 kOe. This training can therefore be attributed exclusively to the induced anisotropy in the AF during the process of field cooling ($H_{FC}=$ $+20$ Oe).[9]

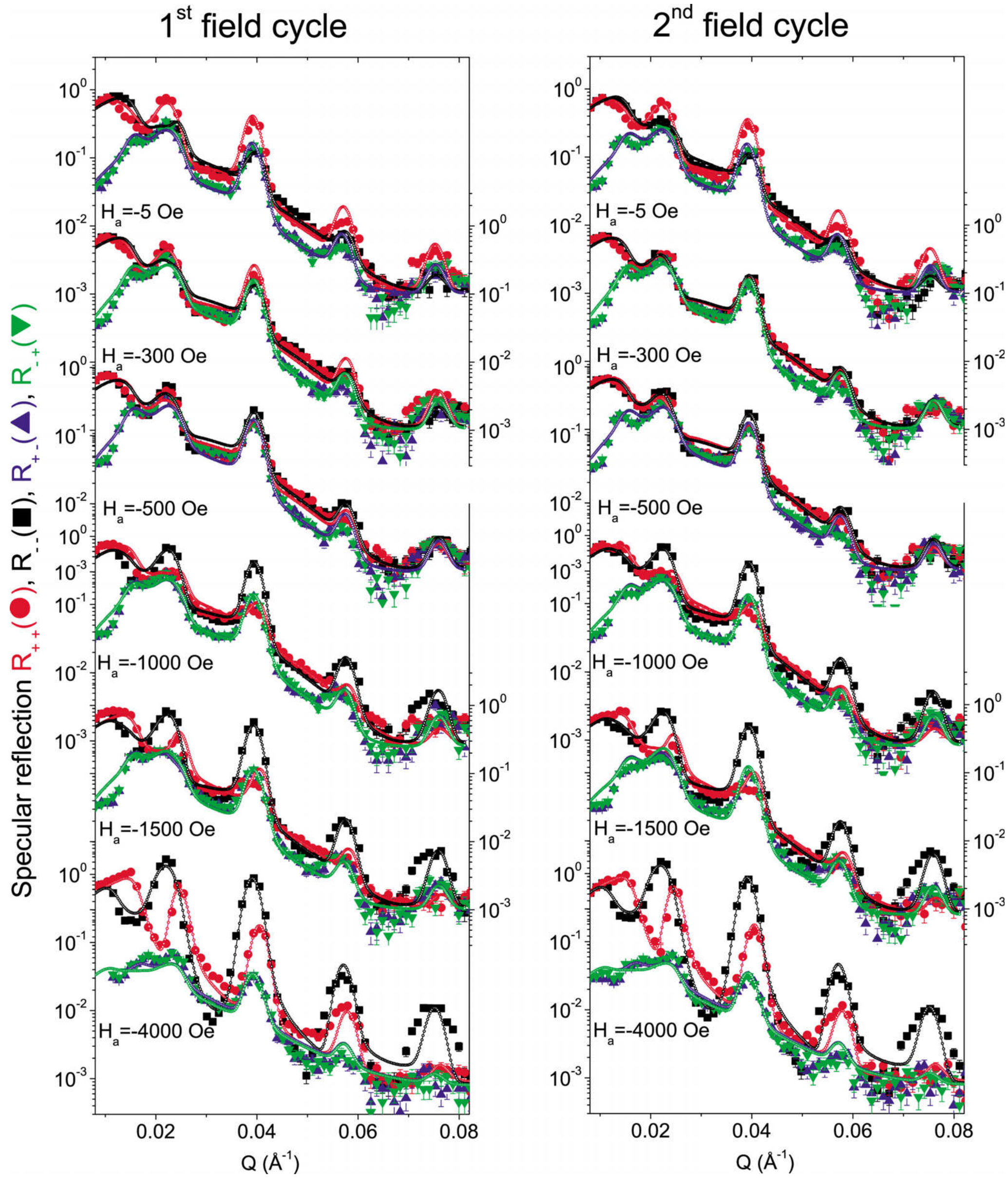

FIG. 3. (Color online) Specular reflectivity patterns for the NSF [R_{++} (red) and R_{--} (black)] and SF [R_{-+} (green) and R_{+-} (blue)] channels measured from Co/CoO/Au ML at different H_a as indicated after *positive* saturation. Solid symbols are data and open circles are fits to a model.

In Figs. 5(a) and 5(b) we show the SF specular intensities (on a linear scale) for the case of remanent field cooling and that for the coercive field cooling. The field value at which the spectra are compared also shows the maximum evidence of training for coercive field cooling. Further, in Fig. 5(c), we show the difference spectra (SF channels) for the two cases. Here the training is convincingly depicted as compared to the untrained case. The intensities are average intensities of the R_{+-} and R_{-+} channels.

IV. DISCUSSION

In explaining the role of AF moments[4,18] (during this crossover from untrained to trained state), we opt for the energy minimization calculations on spin-glass systems that were done by treating the spins as Ising or as Heisenberg types.[19] In case of Ising spin system, the variation in magnetization with field were shown to have a magnetic hysteresis, whereas for the isotropic Heisenberg spin system, no such hysteresis were predicted. With the introduction of a uniaxial anisotropy term within the Heisenberg spins, the loops resemble to that of an ordered Ising spin system. Thus, the irreversibility of spins (responsible for magnetic hysteresis), was obviously associated with the uniaxial anisotropy.

In exchange bias system, for *remanent field cooling*,[11] the AF grains remain largely isotropic—despite repeated field cycling. Thus, within the spin-glass model, they can be regarded as behaving like Heisenberg type of spins, i.e., spins with poorly defined uniaxial anisotropy (follow the magnetic field). On the other hand, for a *saturated field cooling*, the system can be similarly considered as behaving like Ising type of spins (trapped in a metastable state) with well-defined uniaxial anisotropy.

For the *coercive field cooling* case, the AF moments are coupled to the exchange fields of two different FM configu-

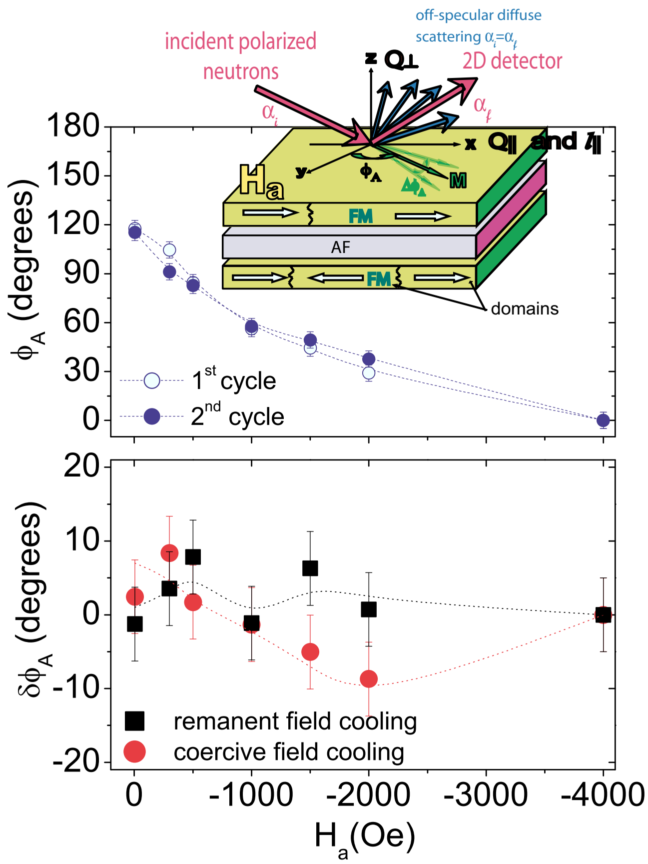

FIG. 4. (Color online) (a) Variation in the angle ϕ_A as a function of H_a for coercive field cooling. The dashed lines are guides to the eye. The schematic shows the scattering geometry. (b) $\delta\phi_A$ for the remanent field cooing (reconstructed from Ref. 11) as well as for +20 Oe (coercive field) cooling.

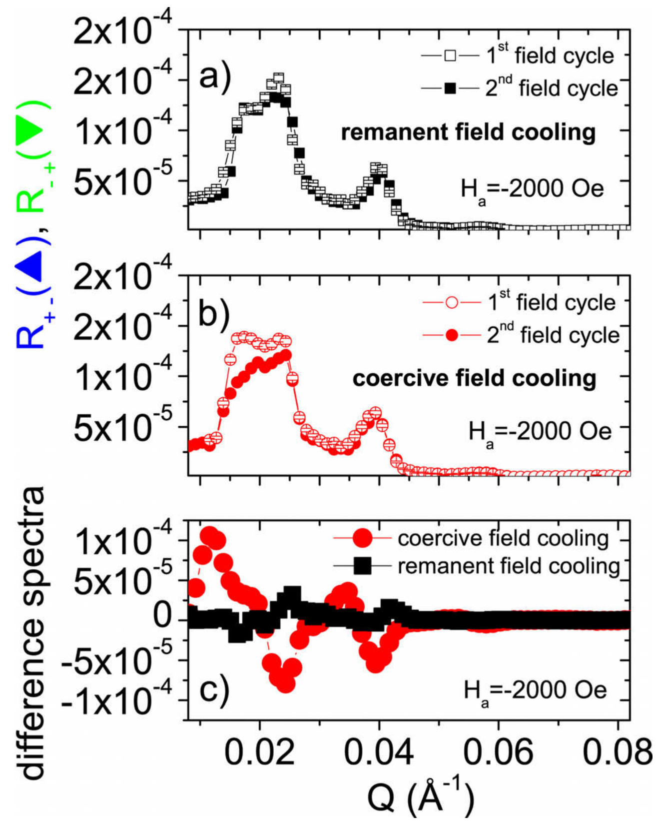

FIG. 5. (Color online) SF specular signal at H_a=−200 Oe for (a) remanent field cooling and (b) coercive field cooling. (c) Difference spectra of the SF channels corresponding to the two cases of field cooling has been compared to show the training for coercive field cooling.

rations: the lower components of the SQUID loop are due to Heisenberg type of spins (poorly defined uniaxially anisotropy), while the upper components of the loop are due to Ising type of spins (fairly defined uniaxially anisotropy). We argue that the irreversibility of spins that causes the training in this case originates exclusively from the freshly introduced uniaxial anisotropy. We rule out the possibility of any biaxial anisotropy axis as such a frustration was definitely not manifested even in the case of remanent field cooling.

In case of remanent field cooling, the system behaves as poorly defined uniaxial anisotropic system or as isotropic Heisenberg type of system. This leads to a reversible magnetization reversal (suppression of training). In case of such an isotropic system, it is only after the introduction of a uniaxial anisotropy term that the irreversibility (hysteresis) is manifested. One may recall that a biaxial term only increases such irreversibility due to increased frustration in the system. Thus, it is obvious to conclude that no anisotropic term (uniaxial or biaxial) existed for our remanent field cooled state as it showed a complete suppression of training.

Now for our coercive field case, we are dealing with (a) the same system (identical preparation conditions), (b) we follow the same procedure and conditions of field cooling (in the same direction with respect to the applied field), and (c) we use the same instrument to measure the specimen (identical environment). The only difference being the magnitude of the field (≈20 Oe) that was applied before it was cooled down below the blocking temperature. Thus, we can rule out the possibility of introducing an additional frustration (like biaxial anisotropy) within the system externally. Additionally, the training that we observe is only about 10% as compared to the untrained state. This is also in a way indicative of the fact that it is only uniaxial anisotropy that is induced. A biaxial anisotropy would have increased the training significantly. Moreover biaxial anisotropy is more common in epitaxial systems. It has been shown earlier that exchange coupling not only provides a unidirectional bias field but also induces a uniaxial anisotropy, breaking the biaxial symmetry of epitaxial Co layer.[20] Fourfold symmetry of anisotropy is rarely found in polycrystalline films (because the biaxial easy axes are randomly oriented) and that too under very special conditions (scratches or contamination on surface).[21]

Heisenberg type of spins can always form a local unidirectional anisotropy in a direction along the AF anisotropy, which can be away from the FM magnetization direction. Ising type of spins, on the other hand, would be restricted along the FM magnetization direction. Upon field cycling the magnetization for Heisenberg type of spins is independent of history, whereas Ising types are dependent.

V. CONCLUSION

In conclusion, the *suppression of training*—due to poorly defined uniaxial anisotropy, followed by the *introduction of training*—due to well-defined uniaxial anisotropy, clearly shows that training in exchange bias systems can originate exclusively out of uniaxial anisotropy within the AF layer. Monitoring training at its onset, has been particularly possible due to the unusual coexistence of two different FM configurations, realized within the same polycrystalline system by coercive field cooling.

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