Origin of the Training Effect and Asymmetry of the Magnetization in Polycrystalline Exchange Bias Systems

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The training effect and asymmetry in exchange-coupled polycrystalline CoO/Co bilayers with in-plane magnetization has been investigated. This system is selected for its large training effect and initial asymmetry of the magnetic hysteresis after field cooling, which is removed after training. Applying an in-plane magnetic field perpendicular to the cooling field largely restores the untrained state with its pronounced asymmetry. The possibility to reinduce the asymmetry strongly depends on the magnitude of the perpendicular field, providing the key to identify the physical origin of training and removal of the asymmetry. These effects result from misalignment between the ferromagnetic magnetization and the uncompensated magnetization of the granular antiferromagnet.

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Coupling between an antiferromagnetic (AF) and a ferromagnetic (FM) layer, referred to as exchange bias [1], results in a shift and a broadening of the hysteresis loop of the ferromagnet and received much attention because of its importance for technological applications.

Here, we focus on the training effect [2] and the symmetry of the magnetization loop [3], which can be clearly observed in the intensively studied CoO/Co bilayer system. It is generally accepted that the origin of the training effect is related to a change in the state of the AF layer compared to the original state after field cooling. Since Néel's original explanation of the training effect [4], several models with different physical backgrounds have been put forward to explain the asymmetry and training effect. Hoffmann [5] pointed out that the specific anisotropy of the AF layer plays a crucial role in understanding of the training effect. The model confirmed the experimental finding that training effects are not present in EB systems with uniaxial magnetic anisotropy in the AF layer. In a different approach Suess *et al.* [6] associated exchange bias with domain wall formation between weakly exchange-coupled grains in the antiferromagnet with a perfectly compensated interface. The storage of unidirectional anisotropy energy in lateral domain walls between the AF grains leads to a training effect and asymmetry when cycling through consecutive hysteresis loops [7]. An earlier approach, based on the Fulcomer and Charap model [8], considered an AF layer made up of an assembly of noninteracting grains with different sizes and anisotropy orientations. This model explained the training effect as the result of a changed spin orientation in the uncompensated AF grains [2,9]. Binek *et al.* [10] proposed to link the training effect to a rearrangement of the AF spin structure towards equilibrium. Relying on domain wall formation parallel to the interface, Radu *et al.* [11] ascribed the training effect and asymmetry to the formation of interfacial domains, while Hauet *et al.* [12] explained the training effect as the irreversible change of a frozen domain wall near the interface.

It is clear that additional experimental results are necessary to identify the physical mechanism responsible for the training effect in the CoO/Co system. We performed a detailed study of the training effect in CoO/Co bilayers with four-point high-resolution measurements of the anisotropic magnetoresistance (AMR) effect. The AMR effect is caused by the spin-orbit scattering. For a saturated ferromagnet, it depends on the relative orientation of current, and magnetization and can be expressed as [13]

$$R(\beta) = R_\perp + R_0\cos^2\beta, \tag{1}$$

where β is the angle between the current and the magnetization direction, $R_\perp$ is the resistance with the magnetization perpendicular to the current, and R_0 is the difference in resistance with the magnetization parallel and perpendicular to the current, respectively. As a result, the contribution of pure domain wall motion to the AMR effect is negligible during the magnetization reversal, while pure rotation of the magnetization changes the AMR effect between a minimal and a maximal resistance value.

The CoO/Co bilayers are prepared at room temperature by dc magnetron sputtering a 20 nm thick Co layer at a rate of 0.5 Å/s on top of an oxidized Si wafer. After deposition, the Co layer is oxidized *in situ* in a partial oxygen pressure of 10^{-3} mbar [14]. Simulations of the x-ray reflectivity profile show that the oxidation results in the formation of a 2–3 nm thick CoO top layer [11] with an average rms roughness of 0.6 nm. X-ray diffraction measurements further confirm that the Co layer is polycrystalline. The exchange coupling between both layers is established by cooling the CoO/Co bilayer in a field of 400 mT parallel to the CoO/Co interface to 10 K, which is well below the temperature [15] where the exchange bias shift first appears. Figure 1(a) shows the hysteresis loop measured at

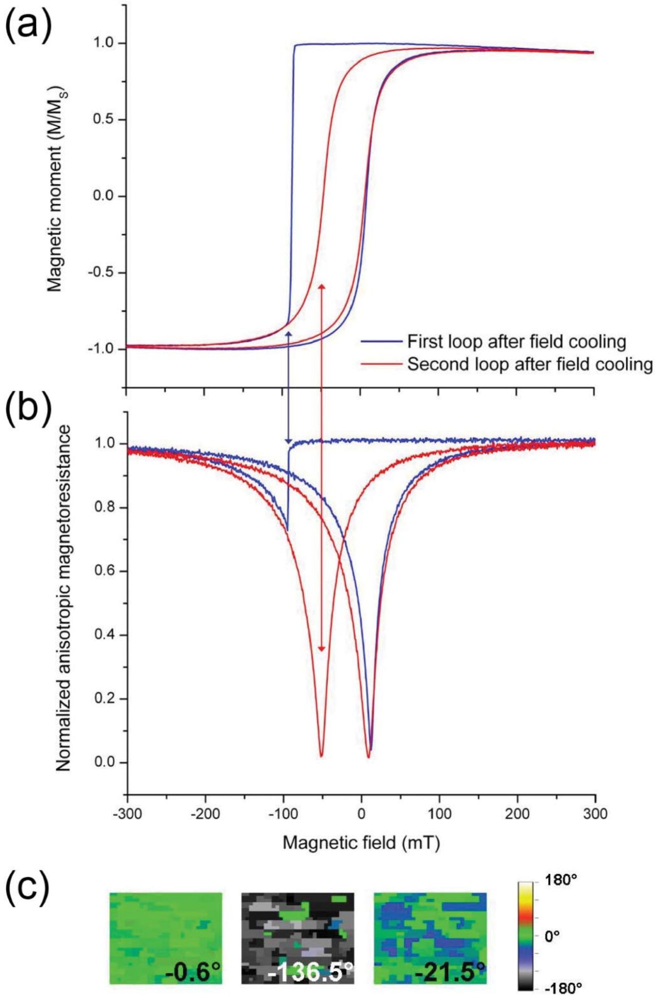

FIG. 1 (color). Magnetization (a) and magnetoresistance (b) measurement of a CoO/Co bilayer at 10 K after cooling in a field of 400 mT. In (c) we present the calculated distribution of the interfacial magnetization vectors of the AF CoO grains after field cooling (left), after the first reversal (middle), and after a complete hysteresis loop (right). The indicated angles represent the average interfacial AF magnetization direction with respect to the cooling field.

10 K with a vibrating sample magnetometer and Fig. 1(b) shows the corresponding AMR signal. The magnetization measurements confirm that the first magnetization reversal after field cooling is abrupt, while subsequent reversals are more rounded [11]. The smaller AMR change during the first reversal confirms that this reversal is dominated by domain wall motion, while the following reversals mainly result from a rotation of the magnetization. Previous experiments indicated that, once training has occurred, it is not possible to get back the asymmetry of the initial hysteresis loop without heating the sample when the magnetic field is applied along the cooling field direction. Figure 1(c) will be explained later.

Recently, we reported on the surprising possibility to largely reinduce the untrained state and asymmetry by performing a hysteresis measurement with an in-plane external field perpendicular to the cooling field direction, without changing the temperature of the bilayer [16]. The next hysteresis loop along the cooling field obtained after the perpendicular hysteresis loop resembles the initial asymmetric hysteresis loop with a reduced amount of spin rotation occurring at the first coercive field.

Our detailed AMR measurements reveal that the reversal of the training effect and asymmetry is strongly dependent on the magnitude of the applied perpendicular magnetic field. This remarkable new result is summarized in Fig. 2. For each data point in Fig. 2, the CoO/Co bilayer is cooled from room temperature to 10 K in a field of 400 mT. Subsequently, two hysteresis loops are measured along the cooling field to remove the asymmetry. Next, a hysteresis loop is measured with the magnetic field still in the sample plane but perpendicular to the cooling field direction. For this perpendicular hysteresis loop, the maximum applied field is given on the horizontal axis of Fig. 2. The following hysteresis loop is measured with the magnetic field again applied along the cooling field direction, and some of the latter measurements are shown in the insets of Fig. 2. Each point of the graph in Fig. 2 represents the degree of asymmetry, which is defined as the difference in height of the AMR peaks at both coercive fields divided by the change of AMR at the second coercive field. If the perpendicular field has a maximum value around 150 mT, the initial situation after field cooling (large training effect and asymmetry) is largely recovered. If the perpendicular magnetic field is small, the AF layer remains unaffected. On the other hand, the asymmetry can no longer be recovered if the perpendicular field is increased above 400 mT.

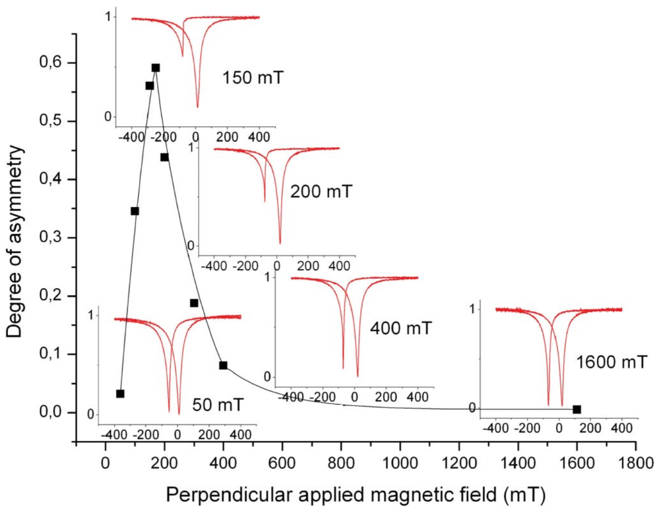

FIG. 2 (color). Degree of asymmetry (see text) of the AMR along the cooling field after performing a hysteresis loop perpendicular to the cooling field. The horizontal axis corresponds to the maximum value of the applied perpendicular magnetic field. The full curve is a guide to the eye. As illustrated by the different insets, dependent on the perpendicular field, the initial asymmetry of the hysteresis loop measured along the cooling field can be reinduced.

It is possible to explain the recovery and disappearance of the training effect and asymmetry within the framework of the model of Fulcomer and Charap [8], which was modified by Hou *et al.* [17]. This model corresponds to many AF grains with size G_i that are exchange coupled to a single FM domain with size $\sum G_i$. An FM type of interaction is considered between the uncompensated AF grains and the neighboring FM domain. After field cooling, the interfacial magnetizations of the AF grains are approximately oriented along the cooling field. If the FM magnetization is reversed, some of the interfacial magnetization vectors of the AF grains will change their orientation depending on their size and orientation with respect to the AF easy axis. The total energy of the model is

$$E = K_{\mathrm{AF}} t_{\mathrm{AF}} \sum_i G_i \sin^2[3(\varphi_i - \alpha_i)] - B \sum_i \sqrt{G_i} \cos(\theta - \varphi_i) - \mu H M_S t_F \cos(\psi - \theta) \sum_i G_i, \tag{2}$$

where t_{AF} (3 nm) and t_F (20 nm) denote the thickness of the AF and FM layer, respectively. The constants K_{AF} and B are the magnetocrystalline anisotropy of the antiferromagnet and the FM exchange coupling between both layers, respectively. The constant K_{AF} is 5×10^5 J/m^3 [18] and B (2×10^{-11} J/m) is fixed in such a way that the exchange bias shift after field cooling corresponds to the experimental value. The first term in the energy equation describes the magnetocrystalline energy of all the AF grains, the second term represents the exchange coupling between the AF grains and the FM domain, and the third term is the Zeeman energy. The angles φ_i, α_i, θ, and ψ are the angles between the interfacial magnetization of the AF grains, AF easy axis, FM magnetization, applied field, and a reference axis (chosen to be the cooling field direction), respectively [2]. Since the Co domain contains many differently oriented crystal grains, the magnetocrystalline energy of the ferromagnetic domain is omitted. The AF grains are supposed to not interact with each other because samples with distinct grain boundaries are considered, and grains are sufficiently small in order not to break up into domains. If there would exist a strong interaction between AF grains, clusters of AF grains would be formed that rotate coherently. In that case, the clusters of AF grains can be treated effectively as one grain in the model. The grain areas are distributed with a log-normal distribution with an average of 9 nm^2 and a standard deviation of 8 nm^2. In our calculations we used 100 AF grains that are coupled to one FM domain. Each grain is supposed to have 3 easy axes, and the orientations of the easy axes of the different grains are randomly distributed within the plane of the film. The starting conditions are such that all interfacial magnetizations of the CoO grains are directed along the easy axis closest to the cooling field direction, and the magnetization of the FM layer is oriented along the cooling field. The state after field cooling is searched by implementing a Newton minimization algorithm, which results in a domain map of the interfacial magnetization of the AF CoO grains where the average magnetization vector is directed $-0.6°$ away from the cooling field [left color map in Fig. 1(c)]. During the reversal of the magnetic field, some of the interfacial magnetization vectors of the CoO rotate, while others remain near the same position [middle color map in Fig. 1(c)]. In our calculations the FM magnetization rotates in the negative sense (from 0° to $-180°$) because the average orientation of the interfacial magnetization vectors of the CoO after field cooling is slightly negative and, as a result, some of the magnetization vectors follow this negative direction. If the magnetic field is changed back to its original value of +400 mT after field cooling, not all magnetization vectors rotate back to their initial position [right color map in Fig. 1(c)]. After going through a complete hysteresis loop, the average AF interfacial magnetization is directed $-21.5°$ away from the cooling field, which creates a torque acting on the ferromagnetic spins and triggers the transition from domain wall motion to rotation of the magnetization. Our model calculations are consistent with earlier calculations within the framework of the same model [2]. Both calculations confirm the experimental fact that the first magnetization reversal of the ferromagnetic Co layer is more abrupt, while subsequent hysteresis loops are dominated by a gradual rotation of the Co magnetization.

The results of our calculations are shown in Fig. 3(a) where, similar to Fig. 2, the hysteresis loops in the insets are those along the cooling field after performing the perpendicular hysteresis loop. The corresponding interfacial magnetization vectors of the CoO at saturation are shown in Fig. 3(b). The different color maps on the left-hand side of each of the 6 panels in Fig. 3(b) illustrate the domain configurations if a field of 380 mT (field parallel to the 0° direction) is applied along the cooling field direction after performing a perpendicular hysteresis loop with a magnitude shown on the left-hand side of the color maps. The perpendicular field is varied along a direction parallel to the ±90° directions and a perpendicular hysteresis loop ends along the +90° direction. The color maps on the right-hand side in each of the panels in Fig. 3(b) illustrate the domain configuration at -380 mT (field along the $-180°$ directions), i.e., after the reversal of the FM magnetization. Figure 3(b) reveals that the AF grains are not influenced by a small perpendicular field [compare left color map in Fig. 3(b) at 50 mT and right color map in Fig. 1(c)]. The hysteresis loop along the cooling field is still symmetric and the FM magnetization rotates along the negative direction. The left domain configuration of Fig. 3(b) illustrates that if the maximum applied perpendicular field in the model is increased to 90 mT, the average interfacial magnetization vector of the CoO (2.8°) rotates slightly beyond the initial position after field cooling ($-0.6°$) and the FM magnetization now rotates in positive direction [domain configuration on the right-hand side of the panel for 90 mT in Fig. 3(b)]. At this perpendicular field value, the initial situation after field cooling with a larger asymmetry is largely recovered. If the magnitude of the

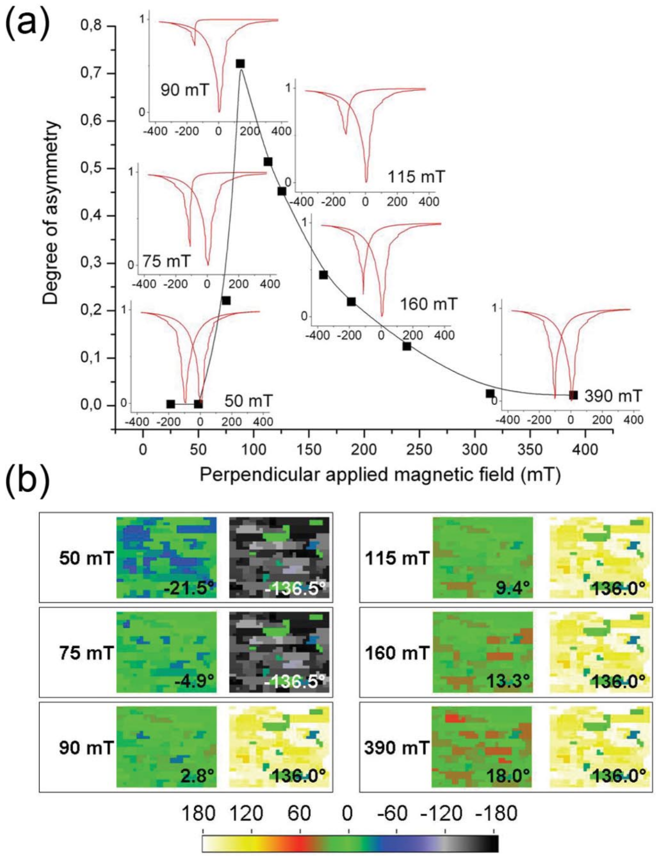

FIG. 3 (color). Calculated AMR effects (a) based on the extended Fulcomer and Charap model. The color maps in (b) correspond to the calculated domain configurations in the AF layer at saturation of the FM layer for a magnetic field along the cooling field direction and after applying a perpendicular magnetic field. The magnitude of the perpendicular field is indicated on the left-hand side of the color maps in each of the 6 panels. The left color map in each panel shows the calculated configurations for positive saturation at 380 mT, while the right map shows the domain configurations after the reversal of the FM magnetization at −380 mT.

perpendicular field is further increased, the interfacial magnetization vectors of the CoO rotate more and more beyond their initial field cooling position. As a result, the asymmetry is absent and the first reversal is dominated by a rotation of the magnetization. Here, we only want to obtain qualitative agreement for the magnetic field dependence of the degree of asymmetry. Since the model only considers the most dominant energy terms, a perfect agreement between theory and experiment cannot be achieved.

The model clearly reveals that the orientation of the interfacial magnetization of the AF grains can be largely rotated back to the initial orientation after field cooling by the application of the appropriate perpendicular external field. If the perpendicular field is too low, the interfacial magnetization of the AF grains remains unaffected. If, on the other hand, the external field becomes too high, the interfacial AF magnetization vectors rotate beyond their initial field cooling position, and the asymmetry again disappears. Since the model is able to consistently explain the evolution of the training effect and the asymmetry of the hysteresis loop, including the peculiar influence of a perpendicular magnetic field, we are confident to have identified the microscopic origin of the asymmetry and training effect in CoO/Co bilayers.

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