Critical angle for irreversible switching of the exchange-bias direction in NiO-Cu-$Ni_{81}Fe_{19}$ films

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The stability of the reference magnetization in exchange-biased NiO (10–13 nm)/Cu (0.2–0.8 nm)/Permalloy (10 nm) layers was investigated by Kerr microscopic domain studies in an optical cryostat. The stability of the coupling was found to depend on temperature and on the direction of an applied magnetic field. We discovered different blocking temperatures $T_{B,hard} > T_{B,easy}$ for hard and easy axis magnetization reversals. Moving 180° domain walls are able to permanently switch the pinning direction by 180°. In rotational field experiments it could be proved that it is not the wall itself that acts on the antiferromagnet, but rather the torque of the ferromagnetic moment at the interface that probably remagnetizes the antiferromagnetic film by motion of a Bloch wall parallel to the film plane. We determined a critical angle α_C for permanent switching which depends on temperature.

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I. INTRODUCTION

Exchange coupling between ferromagnetic and antiferromagnetic films provides a fixed reference magnetization, which is essential for magnetoelectronic devices based on giant magnetoresistance and tunneling magnetoresistance effects. If the film system has been heated and cooled in the presence of an external magnetic field, the coupling acts as a bias field on the ferromagnetic film (exchange bias), leading to a hysteresis loop that is shifted along the field axis by the exchange bias field H_{eb}. Reviews on exchange bias can be found in Refs. 1–3.

For technological reasons, the temperature dependence of the exchange bias is of particular interest. The critical temperature for exchange coupling in an antiferromagnet is the Néel temperature. For applications, however, other critical temperatures play a more important role. These are, e.g., the minimum temperature needed to set the reference magnetization or the maximum annealing temperature that still preserves the reference. The so-called blocking temperature T_B is defined as the temperature at which H_{eb} becomes zero. This definition tries to give a parameter that is easy to measure and relevant for applications. The blocking temperature and the Néel temperature of the antiferromagnet are related to each other, but in general they are not identical. The clarification of this difference as well as an understanding of the blocking temperature is still a challenging subject of fundamental research.[4] The temperature behavior is commonly investigated by magnetometry or magnetoresistive current measurements. These methods are integral, meaning that information averaged over a certain area and hence over a random distribution of magnetic grains and orientations is obtained. In contrast, Kerr microscopy as a laterally resolving method allows a microscopic real-time observation of the magnetization process of the ferromagnetic layer and thus opens a different view on the coupling, antiferromagnetic (AFM) behavior and the blocking temperature.

In this paper the stability of the reference magnetization in NiO(10-nm)/Cu(0.2, 0.5, 0.8-nm)/Permalloy(10, 13-nm) trilayers (Fig. 1) is addressed by means of domain studies using Kerr microscopy. Low-temperature observation in an optical cryostat was employed, because the blocking temperature is reduced to about 200K for a 10-nm-thick NiO film (as compared to 520 K for bulk NiO). The Cu interface layer reduces, but does not interrupt the exchange coupling between Permalloy and NiO. Exchange biasing has been found to work even through nonmagnetic spacer layers[5], where the coupling strength decreases with increasing spacer layer thickness. An oscillating thickness dependence modulates the decrease of the coupling.[6,7] This allows one to vary the coupling strength independently of the stability of the anitferromagnetic layer. For our samples it might be possible that at the smallest thickness the spacer is not completely closed so that bridges can carry magnetization information between the ferromagnetic and antiferromagnetic films, thus leading to a residual coupling effect even if the Cu spacer should interrupt the coupling. However, the exchange interaction in the soft magnetic NiFe layer will average over the localized bridges leading to a reduced coupling strength in any case. In addition to the influence of the reduced coupling strength on the blocking temperatures as shown in this paper, the spacer layer leads to magnetization processes that are

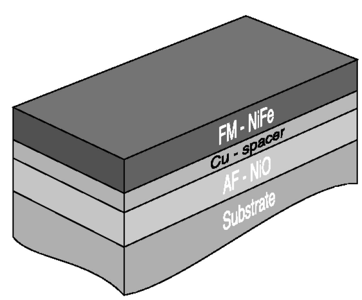

FIG. 1. Layer structure of the investigated systems. The films are prepared by magnetron sputtering.

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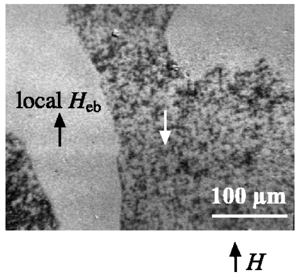

FIG. 2. Patchlike magnetization reversal within those areas, which have a bias direction antiparallel to the applied magnetic field in a directly coupled NiFe 10-nm/NiO 10-nm bilayer. Before the field was applied, an exchange bias pattern consisting of black and wide domains was achieved by zero field cooling as explained in the text.

dominated by the uniaxial anisotropy of the ferromagnetic layer. The resulting domain behavior is well defined, consisting either of wall motion or homogeneous rotation processes for easy and hard axis fields, respectively, whereas for directly coupled NiO/Permalloy films complex patch domains are observed during reversal (Fig. 2—also see Ref. 8). The well defined magnetization processes in the interspaced films allow a more explicit approach to the features of exchange bias than those in the directly coupled films.

II. EXPERIMENT

Extended NiFe/Cu/NiO films were dc/rf magnetron sputtered in the presence of a dc magnetic field of 24 kA/m at the sample to induce a preferred axis of magnetization. The layer thickness of $Ni_{81}Fe_{19}$ was 10 nm in all samples, whereas the thickness of the NiO layer was 10 and 13 nm and the thickness of the copper interface layer 0.2, 0.5, and 0.8 nm. The 0.5-nm Cu samples were produced in a different sputtering chamber under slightly different conditions.

The domain observation was performed in a digitally enhanced Kerr microscope applying the longitudinal Kerr effect.[9] To perform temperature-dependent observations, an optical cryostat and heating stage were mounted in the microscope, which allowed the *in situ* application of rotatable magnetic fields up to 300 kA/m. The accessible temperatures ranged from 10 to 850 K, covering the interesting temperature range for application related systems. To protect the samples from corrosion and water condensation during heat treatment and cooling, respectively, they were kept in vacuum during observation through a stress-free glass window. Long-distance objective lenses were used thereby limiting the spatial resolution to about 1 μm at best. The samples with 0.2 and 0.8 nm copper interlayer were further characterized in a superconducting quantum interference device magnetometer. The results of these experiments are reported in Refs. 7 and 10.

III. RESULTS

A. Domains above the blocking temperature T_B

Above the blocking temperature the magnetization process observed for all samples is that of an uncoupled soft

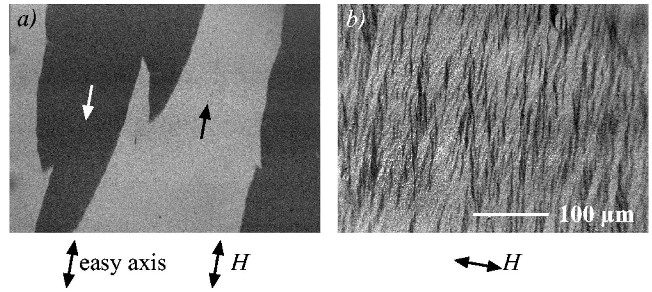

FIG. 3. Magnetization patterns at remanence in a 10-nm-thick Permalloy single film after applying fields along the easy axis (a) and hard axis (b).

magnetic thin film as illustrated in Fig. 3 for a 10-nm-thick single Permalloy film for comparison (for a review of such effects see Chap. 5.5.2 of Ref. 9). In easy axis fields, a domain wall motion often hindered by pinning sites is observed, leading to the formation of charge-reduced zigzag-shaped wall segments.[9] Wide domains can be generated by proper demagnetization along the easy axis. In hard axis fields, a much finer domain pattern is generated when releasing the field from saturation. This system of ''blocked'' domains evolves out of an incipient ripple structure, which reflects the irregular polycrystalline nature of the ferromagnetic layers[9].

In our NiFe/Cu/NiO films we typically generated domains like those in Fig. 3(a) by heating the sample above the blocking temperature. To obtain some domain walls within the field of view, we first created a blocked state in a hard axis field, which was then resolved and widened in an easy axis field. An example of 180° domains being magnetized along the in-plane uniaxial anisotropy axis of the NiFe film and created by this method in an exchange-coupled trilayer is shown in Fig. 4(a).

B. Domain observation below the blocking temperature T_B

A domain state that is created by the described method above the blocking temperature, can be ''frozen in'', i.e.,

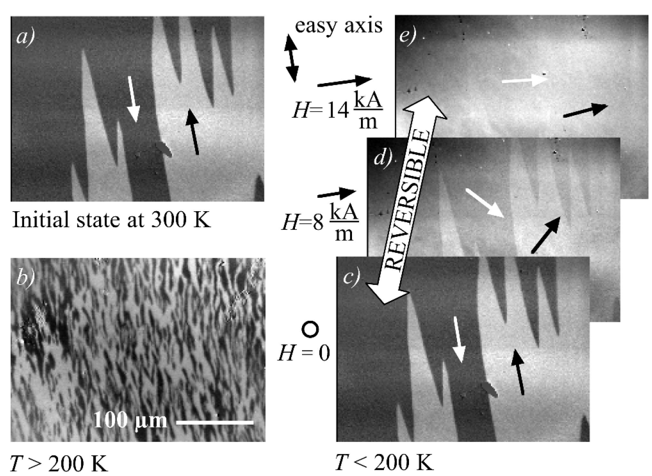

FIG. 4. Hard-axis magnetization processes in a NiFe10-nm/Cu 0.2-nm/NiO 10-nm trilayer. The frozen-in domain state (a) is magnetized irreversibly above 200 K (b) and reversibly by rotation processes below 200K [(c)–(e)].

TABLE I. Critical blocking temperatures for hard and easy axis magnetization. Note that the samples with 0.5 nm copper have been deposited in a different chamber. They are not necessarily comparable to the others. The permalloy thickness was 10 nm for all samples.

Layer thickness Cu/NiO	0.2 nm/ 10 nm	0.8 nm/ 10 nm	0.5 nm/ 10 nm	0.5 nm/ 13 nm
$T_{B,hard}$	200 K	236 K	300 K	>400 K
$T_{B,easy}$	135 K	165 K	220 K	350 K

stabilized by cooling the sample below the blocking temperature. This pinning effect is caused by the creation of a local, unidirectional exchange anisotropy that follows the domain magnetization directions.

If an increasing hard axis field is applied to such a stabilized domain state [Fig. 4(c)–4(e)], the Kerr contrast continuously diminishes, indicating a homogeneous rotation process. On lowering the field, the magnetization reversibly returns into the initial state. No ripple or blocking is observed, in contrast to the uncoupled state [Fig. 4(b)] that occurs above 200 K. We found it impossible to erase the frozen-in pattern in hard axis fields up to 300 kA/m at any temperature below the decoupling temperature. The specific temperatures for hard axis fields, as derived from Kerr microscopic observations, are given in Table I (upper row) for different samples.

In easy axis fields the behavior becomes more complicated. If a field along the preferred axis is applied to the frozen-in domains (Fig. 5), the 180° domain walls start to move in a similar way as expected for an uncoupled film, but at higher fields of about 50 kA/m depending on spacer layer thickness and temperature. The wall displacement is demonstrated in Figs. 5(b) and 5(d) by difference images: Here the initial, frozen-in domain state was taken as a background image, which was then subtracted from an image in an applied field. Those areas that have been wiped out by the moving domain walls show up in a black contrast.

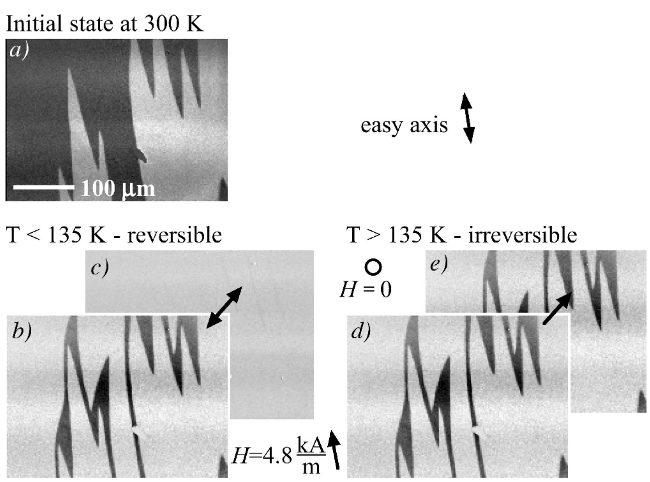

FIG. 5. Easy-axis magnetization processes in the same sample as in Fig. 4. Starting from the frozen-in state (a), the magnetization process is characterized by wall displacement as shown by difference images as explained in the text. The wall displacement is reversible [(b) and (c)] or irreversible [(d) and (e)] for low and high temperatures, respectively.

The wall motion can be reversible or irreversible, depending on the temperature range. Switching off the field at temperatures below about 135 K, the walls of the weakly disturbed domain state [Fig. 5(b)] reversibly move back to their initial position, leaving an almost contrast-free difference image [Fig. 5(c)]. At temperatures above 135 K, but below the blocking temperature, the disturbed domain state remains unchanged after switching off the field [Figs. 5(d) and 5(e)]. So obviously the blocking temperature seems to be lower for easy axis than for hard axis fields. Therefore we divide the blocking temperature into $T_{B,hard}$ and $T_{B,easy}$ with $T_{B,hard} > T_{B,easy}$. The easy axis reversibility at $T < T_{B,easy}$ is even true if the field is released from saturation. Specific temperatures as they were determined from domain experiments like in Figs. 4 and 5 are collected in Table I. The blocking temperatures are obviously not only determined by the thickness of the antiferromagnetic layer as known from the literature.[4] We also found different blocking temperatures for different spacer thicknesses (Table I). Note that the samples with 0.5-nm Cu spacer are not necessarily comparable to the others, since they have been deposited in another chamber.

Interesting is the blocking-range between $T_{B,hard}$ and $T_{B,easy}$, in which 180° domain walls are irreversibly shifted in easy axis fields as observed for all temperatures above $T_{B,easy}$ [Figs. 5(d) and 5(e)]. It is amazing that, within the blocking range, the moved domain state is stable against any subsequent hard axis field treatment. A new domain state, which was created in an easy axis field by the shift of 180° domain walls, now seems to be the pinned state. This indicates that the antiferromagnetic film was obviously affected by the wall motion in the ferromagnetic film.

C. Rotating field experiments

The effect of a moving domain wall on the antiferromagnet can possibly be explained by two mechanisms: (i) The stray field of the domain wall (which is a 180° Néel wall) could locally destroy the frozen-in antiferromagnetic order, which is then rebuilt by the new domain that is left behind after the wall passed by. Such wall stray field effects were observed by Parkin and co-workers[11,12] in a trilayer system, in which hard and soft ferromagnetic layers were interspaced by a nonmagnetic layer. It was found that the hard layer can be demagnetized in magnetic fields much smaller than its coercive field, when these fields are used to repeatedly switch the magnetization of the adjacent soft magnetic layer. The demagnetization is caused by the fringing fields of Néel walls in the soft layer, which easily exceed several hundred kA/m. (ii) The magnetization angle in the ferromagnetic film, which is rotated by 180° when the wall moves along, could also affect the spins in the antiferromagnetic film by action of the exchange torque at the interface.

To decide on these two possible interpretations, a rotating field experiment was carried out on the sample with 10-nm NiO and 0.5-nm Cu spacers. The magnetization in the ferromagnetic film should be forced to rotate without any domain wall displacement. For this, the sample was first saturated in

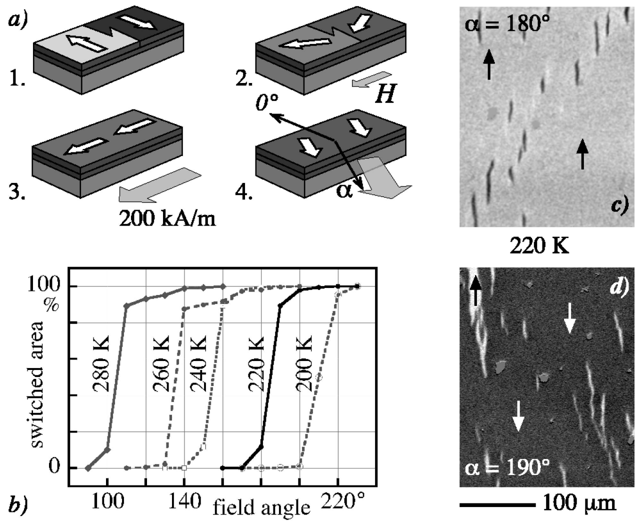

FIG. 6. (a) Schematics, explaining the four steps of our rotating field experiment. Rotating the saturation field beyond $\alpha = 180°$ leads to an irreversible switching as shown by domain images in (c) and (d). The fraction of the switched area, as derived from Kerr images, is plotted in (b) as a function of the field angle for various temperatures.

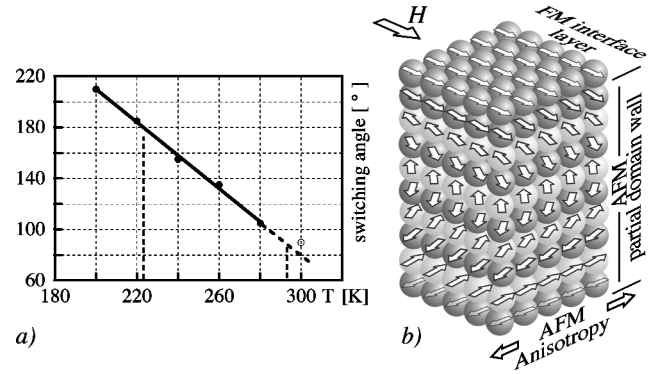

FIG. 7. (a) Switching angles as a function of temperature, derived from the rotating field experiment. (b) Schematic visualization of a horizontal Bloch wall entering the antiferromagnetic film according to Ref. 13.

hard axis direction [steps 1–3 in the sketch of Fig. 6(a), $\alpha = 90°$], where only magnetization rotation is observed as shown before. For complete saturation of the ferromagnetic layer, a field strength of 20 kA/m was sufficient, which is much lower than the expected spin flop field of at least several 1000 kA/m. A further rotation of the saturation field by an arbitrary angle drags the magnetization along without producing domain walls (step 4). To switch the field off after the rotation process, it has first to be turned back in the hard axis (steps 4 to 1). Otherwise domain walls would sweep through during field reduction. The initial domain state should remain unchanged, if the presence of domain walls were responsible for the irreversible changes reported before.

A typical observation of this experiment is shown by the two domain images in Figs. 6(c) and 6(d), where only one domain of the frozen-in 180° pattern is presented. The domain magnetization was initially stabilized at 0° along the preferred axis, which results in a white domain. After saturation at 90° and subsequent field rotation, no change in magnetization of the observed domain was obtained after switching-off the field for angles up to 170° at 220 K. At a rotation angle of 180° [Fig. 6(c)], small reversed domains have been irreversibly nucleated, and for 190° [Fig. 6(d)] the magnetization had almost completely switched by 180°, resulting in a black domain at remanence. For rotation angles beyond 190°, the images appear homogeneously dark, for angles lower than 180° they are homogeneously bright. The areal fraction of reversed domains is plotted in Fig. 6(b) as a function of temperature (switched area 0% means no reversed domains, and 100% represents the fully reversed state after complete switching). With increasing temperature, the average switching angle decreases.

IV. DISCUSSION

The rotational field experiment shows that it is not the domain wall itself, which causes the irreversible change of the initial domain magnetization observed in Fig. 5 (no domain walls were present in the rotating experiment). There is rather a critical angle α_C of the ferromagnetic magnetization relative to the frozen-in direction, which is responsible for the irreversible switching if it is exceeded. Assuming an easy axis in the antiferromagnet parallel to that of the ferromagnet, the rotating ferromagnet seems to switch the sublattice magnetization of the antiferromagnet by 180°, presumably by exerting a torque on the antiferromagnet spins across the interface. This results in the observed reversion of the pinning direction by 180°.

In Fig. 7 the critical angle, extracted from the diagram in Fig. 6, is plotted as a function of temperature. The decrease of this angle is linear with temperature. Angles lower than 90° cannot be measured due to the constraints of the method, which starts with hard axis fields ($\alpha = 90°$) to avoid a displacement of domain walls. The temperature which corresponds to the critical angle of 180° is identical to the blocking temperature (220 K) that was found in an easy-axis-experiment as in Fig. 5 for this sample (see Table I). There the motion of 180° walls was found to be responsible for the irreversibility. Since this domain wall motion over a particular area is nothing else than a rotation of the ferromagnet magnetization by 180°, the blocking temperature $T_{B,easy}$ can be explained by the critical angle of 180°, which is able to act on the antiferromagnetic layer at that temperature. The same argument works for $T_{B,hard}$, where a 90° rotation is able to destabilize the antiferromagnet.

The existence of such a critical angle α_C was theoretically predicted by Stiles and McMichael.[13] According to their model, the critical angle is the result of the formation of a partial domain wall in the antiferromagnet. This means that the AFM spins at the ferromagnetic (FM)-AFM interface follow the FM rotation, whereas the sublattice magnetization far away from the interface stays along its anisotropy axis, leading to a spring like adaptation of the antiferromagnetic magnetization to a given ferromagnetic orientation [as schematically presented in Fig. 7(b)]. Single antiferromagnetic grains will finally switch, if the angle between the interface layer of the antiferromagnet and the intrinsic uniaxial anisotropy exceeds a critical angle. Depending on the ratio of in-plane to out-of-plane anisotropy in the antiferromagnet, out-of-plane spin excitations are considered to take place during pinning reversal or not. For the modelled out-of-plane reversal, the critical angle is limited to 180°.[13] The temperature dependence of this mechanism is reported in Ref. 14. Critical angles which exceed 180°, as observed in our systems, are not explained by this mechanism. We therefore favor the explanation that Bloch wall-like partial domain walls, in which the spin rotation is restricted to the film plane, remain pinned to the ferromagnet and propagate into the antiferromagnet with increasing wall angle. If the width of the partial domain wall becomes comparable to the layer thickness the wall annihilates by leaving the antiferromagnetic film along its thickness direction.

The decrease of the critical angle with increasing temperature, as shown in Fig. 7, could be explained by the assumption that the wall width of the partial domain wall increases not only with increasing wall angle but also with increasing temperature. The required critical wall width for switching is then reached at lower angles for higher temperatures.

Obviously the critical wall width becomes lower when the thickness of the AFM layer decreases. Therefore, the critical temperatures $T_{B,hard}$ and $T_{B,easy}$ are decreasing for decreasing AFM-layer thickness (see Table I), referring to the previously made assumption that the wall width of the partial domain wall increases with increasing temperature.

In a recent publication by Gogol *et al.*,[15] the idea of forming partial domain walls in the AFM layer parallel to the interface was supported by the interpretation of Lorentz-microscopical investigations of exchange-biased CoFe/IrMn bilayers. They took the different domain patterns observed for thick and thin AFM-layers as a hint for this idea. Following their arguments, the thin AFM-layer should not be thick enough to support the formation of a partial domain wall, and is therefore unable to provide a stable exchange bias. This is widely consistent with our experiments. Note that the existence of a partial domain wall in the antiferromagnet was first predicted by Mauri *et al.*[16]

V. CONCLUSIONS

By way of Kerr microscopy observations of coupled NiFe/Cu/NiO trilayer systems with varying Cu and NiO-layer thicknesses, we found that blocking phenomena occur over a wide temperature range. The decoupling takes place at lower temperatures for easy axis magnetization processes than for hard axis reversal. In the temperature range between $T_{B,easy}$ and $T_{B,hard}$, a reversal of the pinning direction was observed as the result of the displacement of 180° domain walls. The mechanism behind this pinning reversal is the 180° switching of antiferromagnetic grains if the magnetization exceeds a critical angle that is temperature dependent in general. The stability of the exchange bias depends therefore on the NiO-layer thickness as well as on the thickness of the nonmagnetic spacer layer, which reduces the coupling strength.

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