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Three-dimensional exchange bias in $\{Co/Pd\}_N/FeMn$

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The exchange bias properties of Co/Pd multilayers biased by FeMn are reported. Co/Pd multilayers exhibit a strong interface anisotropy leading to a ground state where the magnetization is perpendicular to the film plane. In our experiments an FeMn layer was grown on top of the final Co layer of the multilayer and the sample cooled in saturating field both in the plane and perpendicular to the plane. Bias was observed in both directions, although it was stronger in the in-plane case, indicating that the spin structure at the interface deviates from the usual triple-q structure of FeMn.

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In general, magnetic anisotropies show rotational symmetry of an even order, as required by time reversal symmetry. An exception is the exchange anisotropy first discovered by Meiklejohn and Bean.[1] This is observed when a ferromagnetic (FM) and antiferromagnetic (AF) layer are brought into atomic proximity, so that there are exchange interactions between the interfacial planes of spins on either side of the interface. The most commonly reported manifestation of this exchange interaction is a shift in the hysteresis loop of the ferromagnetic layer, which can be interpreted as a unidirectional anisotropy. This is commonly characterized by an exchange field H_{ex}, the field through which the center of the hysteresis loop of the ferromagnet is shifted from zero. This loop shift leads to the term ‘‘exchange bias’’ and is of technological importance in the design of thin-film magnetic devices.[2,3] Moreover, while over four decades have elapsed since Meiklejohn and Bean’s discovery, a rigorous description of the phenomenon is still lacking.[4,5] In particular one problem has been the exact nature of the coupling at the interface between the materials.

In the simplest Meiklejohn-Bean picture the AF layer is terminated by a plane of spins that are all ferromagnetically aligned, as one might find in the correct (111) plane of a crystal of CoO, for instance. Leaving the issue of the magnitude of the bias field[4,5] to one side for the moment, it is straightforward to see how such a plane of spins might couple to an adjacent plane of FM spins and lead to a unidirectional anisotropy. On the other hand, exchange bias has been observed in a very wide variety of AF and FM systems, and for the purposes of this article we should note that many of these will not present such a simple spin structure at the interface to the FM layer—polycrystalline layers or materials such as FeMn that do not have uncompensated planes of spins in any direction still show good exchange bias properties. This issue was addressed in a widely cited paper by Koon,[6] who proposed a simple micromagnetic model where the spins in a compensated AF interface layer couple perpendicularly to the neighboring FM spins, prompting the use of the term spin-flop coupling to describe this state. A similar model was presented later by Kiwi *et al.*[7] Exchange bias then arises as the reversing FM layer winds a domain wall into the AF layer, along the same lines as originally suggested by Néel.[8,9] Mauri *et al.* showed how this winding idea could be used to reduce the bias field to experimentally observed values.[10] This orthogonal coupling was observed directly by neutron diffraction by Ijiri *et al.* in Fe_3O_4/CoO multilayers[11] and was invoked as the explanation for loss of bias in MnF_2/Fe at cooling fields exceeding the spin-flop field of the AF layer.[12]

However, as pointed out by Schulthess and Butler[13] and also Stiles and McMichael,[14] the Koon model is entirely two dimensional, all the spins described only by their in-plane angle. While the shape anisotropy of a thin film will tend to hold FM spins in the plane, as there is a magnetization to give rise to a demagnetizing field, it is not clear on what physical basis the AF spins should be so restricted. When this restriction is relaxed and fully three-dimensional (3D) spins are modeled, as in Refs. 13 and 14, it is found that spin-flop coupling cannot give rise to exchange bias in the manner proposed by Koon, as any in-plane AF wall that forms can unwind itself in 3D as the AF spins rotate out of the plane. Similar results were obtained by Camley *et al.*[15] Although there has been a wide theoretical debate around this point, it has not been directly addressed experimentally—in this article the results of experiments where the AF spins in an exchange-biased system have been set in both in-plane and out-of-plane directions are reported.

Such samples have just become available very recently and have been studied by only a very small number of groups. Kagerer *et al.* prepared Co/Pt multilayers that were biased with FeF_2,[39] although only the out-of-plane magnetic properties were investigated. Maat *et al.* also fabricated Co/Pt multilayers but biased them with CoO.[16] They investigated the biasing in various directions and found substantially more within the sample plane, which they related to the anisotropy of the single-q spin structure of the CoO. Further studies of this system investigated the symmetry of the magnetization reversal mechanisms.[17] More recently Garcia *et al.* investigated similar multilayers biased with FeMn.[18,19]

For this study, the samples were deposited by dc magnetron sputtering onto pieces cut from a (001)-oriented Si wafer in a custom vacuum system with a partial pressure of $H_2O < 10^{-8}$ Torr. Three different series of samples were prepared: in all three cases the first sequence of layers deposited was $\{Pd(20Å)/Co(9Å)\} \times N$, where N is the number of Co/Pd bilayers in the multilayer stack. The first series of samples were completed by another Pd (20 Å) layer, the second by a Cu (25 Å) layer, and the final series by a FeMn

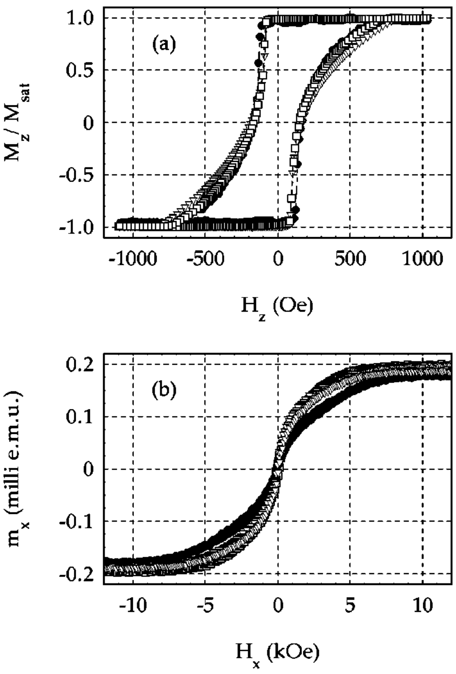

FIG. 1. Hysteresis loops for the three samples with $N=10$ in the as-grown state. (a) Polar MOKE loops and (b) in-plane VSM loops. In both cases open squares represent data for the {Pd/Co}×10/Pd sample, open triangles for the {Pd/Co}×10/Cu sample, and solid circles for the {Pd/Co}×10/FeMn/Ta sample.

(100 Å)/Ta (30 Å) bilayer. We define the *z* direction as perpendicular to the sample, which lies in the *x*-*y* plane. These layer thicknesses were confirmed using grazing-incidence x-ray reflectometry. X-ray diffraction indicated a preferential (111) texture in all the samples. Hysteresis loops were measured by two methods: out-of-plane loops were measured using the magneto-optic Kerr effect (MOKE) with the applied field and incident and exit beams parallel to the *z* direction, while in-plane loops were measured using a vibrating sample magnetometer (VSM), due to the strong mixing of the *z* and *xy* components of the magnetization in this MOKE geometry. Care was taken to ensure that the applied field was carefully calibrated so such quantities as the coercive field were measured to be indistinguishable in these two instruments. Field cooling was carried out in an external electromagnet containing a heated stage so that each group of five samples could be subjected to exactly the same thermomagnetic process simultaneously. The thermal cycle was the same in each field cooling case: a field large enough to entirely saturate the magnetization was applied, and the samples were heated to 200 ° C, maintained at that temperature for 10 min before heating power was removed, and they were allowed to completely cool to room temperature, taking typically 1 h to do so, before the field was switched off.

These multilayers have Co thicknesses below the critical thickness for perpendicular magnetization in the Co/Pd system[20] and will have an increasing tendency to break into a stripe or maze domain pattern as the number of layers increases in order to minimize magnetostatic energy.[21] That the layers have an easy axis along *z* is easily seen from Fig. 1, where data measured in the as-grown state are presented. The data displayed in panel (a) of this figure are from polar MOKE measurements, and the samples are seen to have a high out-of-plane remanence—the squareness of the loops is close to unity, and the coercive field is some 170 Oe. The samples are saturated in a field of only around 800 Oe, much smaller than the 17.6 kOe required to perpendicularly magnetize a Co slab (assuming the bulk magnetization value for Co and only shape anisotropy effects). Meanwhile the data presented in panel (b) of Fig. 1 are taken from in-plane VSM measurements. The remanence is almost zero in this case, while the saturation field is ∼7 kOe, indicative of a hard axis. Similar results were obtained for all the samples, with the coercivity in the *z* direction diminishing somewhat as *N* is reduced. None of these loops show any shift away from symmetry around $H=0$.

Now directing our attention to the differences between the samples, we can see that the samples capped with Pd and with Cu have very similar loops. Although Co has a surface anisotropy at both Co/Cu and Co/Pd interfaces that favors an out-of-plane magnetization, that in Cu has generally been reported to be considerably smaller.[22] The replacement of only one Co/Pd interface out of 20 with a Co/Cu one has not perturbed the system a great deal, however—the loops are almost indistinguishable. The loops for the FeMn/Ta-capped sample is also only slightly different in the as-grown state. The nucleation field for the formation of domains is slightly greater in the polar direction, while the saturation field is slightly higher in the in-plane loop. Both these effects can be understood by considering that the uppermost Co layer is now exchange coupled to the FeMn layer deposited on top of it—these spins will tend to stabilize the Co in its initial direction, making it slightly more difficult to pull over into the plane or break into domains. Already implicit in this idea is the notion of a perpendicular exchange bias, with the FeMn spins tending to couple to moments that are pointing *out of the film plane*.

The samples were then subjected to two field cool processes, in order to set the direction of the bias by bringing the samples down through the blocking temperature (∼150 ° C) with the magnetization of the Co/Pd multilayer saturated. The first was to cool the samples from 200 °C in a 4.5 kOe field directed along the negative *z* axis. After magnetometry a second field cooling process was carried out with a 8.4 kOe field directed along the negative *x* axis. Separate experiments indicated that the magnitude of the bias did not depend on the magnitude of the cooling field (provided it was sufficient to saturate the Co/Pd multilayer); nor did it depend on the order in which the two cooling procedures were performed.

In Fig. 2, the polar MOKE loops for the {Pd/Co}×6/FeMn/Ta sample are shown. Again the as-grown loop (open circles) is symmetric about zero field, with a coercivity of some 170 Oe. After the field cool in the $H_z=-4.5$ kOe field one can see that there is a clear positive shift to the loop of 94 Oe. The coercivity has risen a little to 193 Oe. This rise in coercivity is associated with structural changes that occur upon annealing, since the control samples also show small rises in coercivity: for {Pd/Co}×6/Pd from 133 Oe to 136 Oe and for {Pd/Co}×6/Cu from 126 Oe to 203 Oe. The

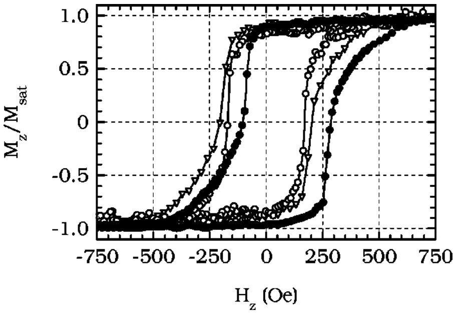

FIG. 2. Polar MOKE loops for the {Pd/Co}×6/FeMn/Ta sample, in the as-grown (open circles) and out-of-plane (solid circles) and in-plane field-annealed (open triangles) states. Perpendicular exchange bias is clearly observed after cooling the sample in an out-of-plane field.

coercivity does not rise substantially higher for repeated anneals to this temperature, indicating that any further changes in microstructure are quite small.

After again cooling in a $H_x = -8.4$ kOe field, it can be seen, upon examining the loop marked by open triangles, that the coercivity has again risen slightly to 204 Oe. However the loop is now centered again around zero field. If the in-plane hysteresis loop is measured with the field swept along the x axis, it is found that this loop is now offset in field by 134 Oe, whereas in the two previous cases—as-grown and post H_z anneal—no offset was observed. This in-plane offset can be seen in the inset of Fig. 3; as the bias field is very small compared to the saturation field of the samples in this hard direction it is difficult to discern in the full loops displayed in the main panel of this figure.

The consequences of these results for theory are significant. There are a large number of theories which are strictly two-dimensional xy models, like the theory of Koon.[6] The original paper by Mauri *et al.*[10] falls into this class, and there are several recent models that also have this characteristic.[7,23–28] Another approach is to attempt a 3D model with Heisenberg spins, but to explicitly introduce a term like a shape anisotropy to limit out-of-plane fluctuations of the AF spins,[29–32] although it is not clear on what physical basis this should be done. The present results indicate that these restrictions ought to be relaxed, in order that this three-dimensional nature of the exchange bias phenomenon can be fully expressed.

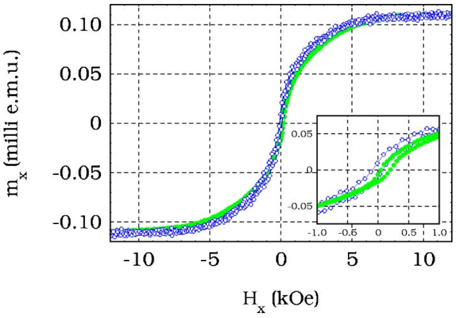

FIG. 3. In-plane VSM loops, measuring moment m, for the {Pd/Co}×6/FeMn/Ta sample, in the as-grown (open circles) and in-plane field-annealed (solid circles) states. The data for the measurement after the perpendicular field cool have been omitted for clarity; they too showed no exchange bias. The inset shows a magnified view of the data around the origin. Exchange bias along the x axis is evident after the in-plane field cooling procedure.

Since truly Heisenberg AF spins cannot give rise to exchange bias through the spin-flop mechanism, one therefore anticipates that the spins in the FeMn that give rise to the bias are collinear with the bias direction. This notion is borne out by recent experimental results. Takano *et al.* measured a small thermoremanent moment on a CoO layer that corresponded to the direction of the bias when a Co layer was placed in contact with it,[40] although this experiment does not exclude the possibility that the small moment was the result of a canted spin-flop state. X-ray dichroism experiments can lead to direct determinations of spin alignments, and there have been a small number of recent attempts employing this technique. Antel *et al.* studied an FeMn/Co bilayer; they found that there was an uncompensated Fe moment that was parallel to the induced bias.[33] Hase *et al.* studied the related IrMn/Co system and measured a weak Mn moment that responded to perturbations in the Co direction caused by an applied field that indicated that it was coupled to the Co in an antiparallel fashion.[34] Similar results have been reported by Sànchez-Hanke and Kao for NiO.[35] All these results are from entirely two-dimensional experiments.

In addition and similarly to Maat *et al.*,[16] it can be seen that there is substantially less bias in the z direction than in the x-y plane. In their investigation of CoO-biased multilayers they described a possible explanation for this as being related to the single-q spin structure of CoO (Ref. 36)–upon field cooling the antiferromagnetic spin structure falls into the nearest easy axis to the ferromagnetic moment direction. In (111)-textured CoO samples these (117) easy axes will not be found to have the same projections in the sample plane as normal to it, and Maat *et al.* suggest that it is this difference in the projection of the AF spin structure that leads to the difference in bias field.

The spin structure of the γ-FeMn layers that has been used in this study is considerably more complex, previously reported to have a triple-q structure in bulk form.[37] Since these samples are also weakly (111) textured, it ought to be possible to follow a similar analysis to that found in Ref. 16. However, due to the tetrahedral symmetry of the triple-q structure, it is not possible to find a projection that is not completely compensated, whatever the direction of the cooling field. Naively, therefore, one might expect that the magnitude of the bias field would be the same in all directions, which is at odds with the experimental findings reported here. The form of the interfacial spin structure may differ, due to the presence of structural defects, uncompensated spins, or the exchange field of the Co. On the other hand, it is worth noting that very recent calculations suggest that the true ground state of γ-FeMn is not exactly the triple-q spin structure,[38] but is a variation on it with small random fluctuations in the direction and magnitude of the local moments reflecting the random nature of the substitutional alloy. The exact nature of these fluctuations at the interface will naturally play a key role in determining any exchange bias.

The idea of the bias arising due to uncompensated AF spins or some other interfacial defects can help to explain the difference in bias field for the z and x-y directions. Some extrinsic effect is required as the difference cannot be ascribed to the isotropic bulk spin structure; the most likely place to find these defects, based on previous work, is at the interface. These uncompensated spins at the interface will be strongly coupled to the FM layer, and it is therefore quite physically reasonable to expect to see some demagnetizing effects that will tend to keep them in the sample plane, reducing the bias field in the out-of-plane direction. Two interrelated conclusions can be drawn: the three-dimensional nature of the bias experimentally confirms that AF spins are not confined to the sample plane and that the use of FeMn does not result in equal bias fields in all directions in space, as might be expected from the high-symmetry triple-q spin structure. Both these point towards the importance of the exact form of the interfacial spin structure, emphasizing the importance of a proper treatment of defects and disorder in untraveling the underlying causes of exchange bias.

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