Antiferromagnetic layer thickness dependence of the IrMn/Co exchange bias system

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A study of exchange bias in IrMn/Co systems is presented. Temperature and thickness dependence studies have revealed non-monotonic behavior in both exchange bias field and coercivity with both variables. In particular the exchange bias field shows a peak for low IrMn thicknesses that is suppressed at temperatures higher than about 200 K. Calculations using the domain state model of exchange biasing are able to describe all the features seen in the experimental data.

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I. INTRODUCTION

Although the discovery of exchange bias by Meiklejohn and Bean1 was over 40 years ago, the effect is still being investigated by the scientific community to understand the fundamental mechanisms controlling both the exchange bias ($H_{ex}$) field, and the coercivity ($H_c$) enhancement. It is well established that the exchange bias is the result of an interfacial interaction between a ferromagnet (F) and an antiferromagnet (AF), although the details of the microscopic mechanism are currently widely debated. The situation is complicated as one finds that there are a number of parameters which influence the exchange bias, such as the anisotropy, crystal structure, interface roughness, interfacial spin configuration and magnetic domains.

Experimental work to date has revealed that $H_{ex}$ is typically two orders of magnitude smaller than values predicted using the original model proposed by Meiklejohn and Bean1, where it was suggested that the biasing was a consequence of the competition between the Zeeman and the exchange coupling energy across an ideal, smooth, uncompensated interface. Experimentally however, it is known that biasing does exist at compensated interfaces and in non-crystalline materials.

Two main theoretical approaches have been pursued in an attempt to explain this discrepancy between the predicted and experimental values. Mauri et al. extended the idea of planar domain walls originating at a smooth AF interface, where the AF spins rotate in the plane, originally put forward by Néel. The exchange energy in this case is spread across the width of the domain wall. Subsequent models which have extended the Mauri concept have also shown limited success in fully explaining the exchange bias effect. In contrast, Malozemoff argued that an ideal interface was unrealistic, and roughness leading to magnetic defects gave rise to local random fields. The total energy of the AF, including the contribution from the random fields, is minimized by the formation of domains in the AF, which have domain walls perpendicular to the interface. Both theories have produced values for the $H_{ex}$ field that compare well with experiment under certain conditions, whereas the $H_c$ enhancement has been attributed to the formation of domains in the AF layer. However, no theory has so far been able to adequately explain both the $H_{ex}$ field, $H_c$ enhancement, and their temperature and AF thickness dependences in full.

Due to the nature of the spin structure of an AF, it is not a straightforward process to characterize such layers using conventional magnetization techniques. Neutron diffraction studies on FeO$_4$/NiO exchange biased superlattices have shown that the exchange biasing is related to the formation of both parallel and perpendicular domains, which are frozen into the AF layer on cooling through the blocking temperature ($T_B$) – this is the temperature above which $H_{ex}$ is zero. Similar measurements on FeMn/Co superlattices have shown that a complex random domain structure is found to be present when the magnetization of the Co layer is reversed and is dictated by the underlying random domain structure within the FeMn. The use of nonmagnetic impurities in the AF layer to form and influence domains has also highlighted the domain structure in the AF being the cause of biasing, and the mechanism which seems to control it.

Investigations of the AF layer thickness dependence in order to establish the thickness at which the biasing saturates are often made at room temperature, motivated by technological concerns, although some experimental data does exist where the work was carried out at low temperatures. Results so far have been interpreted in terms of parallel domain walls in the AF. In this paper we will present evidence that the dependence of $H_{ex}$ on the AF layer thickness and its variation with temperature in fact supports the idea of perpendicular domain walls. To do this we have used calculations from a more recent microscopic domain model, which is able to describe the experimental features we have found at all temperatures through the use of Monte-Carlo methods. The model is based on an irreversible AF interface magnetization which arises from volume domains originating from defects in the bulk of the AF. These volume domains...
then influence the spin structure at the interface.

II. EXPERIMENTAL TECHNIQUES

The Co/Ir$_{25}$Mn$_{75}$ system was studied experimentally within a spin-valve structure. This allowed the free Co layer within the spin valve to be used as a control layer to which the biased Co layer could be directly compared, whilst having only a minimal effect on the properties of the pinned layer – orange-peel coupling fields were \( \sim \) a few Oe. It also allowed magneto-transport measurements to be performed, the resistance measurements were done using a standard four point probe dc technique. Typical (300 K) magnetoresistances of our spin-valves were \( \sim 7 \) per cent, whilst typical (300 K) sheet resistances were \( \sim 10 \, \Omega/\square \). The specimens were deposited by magnetron sputtering at an argon working pressure of 2.5 mTorr. Each set of specimens consisted of 15 samples which were grown during the same vacuum cycle. The base pressure prior to the deposition was of the order of \( 2 \times 10^{-8} \) Torr. The spin-valve structures Ta(75 Å)/Co(40 Å)/Cu(23 Å)/Co(26 Å)/IrMn(\( t_{AF} \))/Ta(25 Å) were deposited onto silicon (100) substrates in an in-plane forming field of magnitude 200 Oe at ambient temperature. The IrMn layers were deposited from a Mn target with chips of Ir attached to its surface, energy dispersive x-ray absorption spectroscopy yields a composition in the deposited film of \( \sim \) Ir$_{25}$Mn$_{75}$. Deposition rates were determined by measuring the thickness of calibration films by low angle x-ray reflectometry. Measurements of individual layer thicknesses within spin-valves by this technique is not straightforward

\[ t_{AF} = 13 \, \text{Å} \quad (a) \]

\[ t_{AF} = 26 \, \text{Å} \quad (b) \]

\[ t_{AF} = 39 \, \text{Å} \quad (c) \]

In panels (a) and (d) the equivalence IrMn orders, and tests have shown that a pinned layer set at room temperature shows the same biasing properties when heated above the blocking point and then field cooled in -0.4 T. (Throughout this paper \( H_{ex} \) is a positive quantity, always opposite in sign to the cooling or setting field). No dependence of \( H_{ex} \) on the magnitude of the cooling field was found provided that this field was large enough to completely saturate the Co layer. Temperature dependence measurements were then done as a series of hysteresis loops at increasing temperatures. Example loops from both these measurement techniques are shown in Fig. 1. In panels (a) and (d) the equivalence of \( H_{c} \) in the free and pinned layers can be seen as the entire sample switches together. Meanwhile in the lower four panels, (b), (c), (e), and (f), the switching of the free and pinned layers can be clearly separated, with the free layer identified by its slightly smaller magnetic moment and much softer magnetic properties. This allows us to determine \( H_{ex} \) and \( H_{c} \) for this layer alone by measuring the fields at which it switches: as is conventional, \( H_{ex} \) is the average of these two fields, whilst \( H_{c} \) is half the difference between them.

\[ H_{ex} = \frac{H_{c1} + H_{c2}}{2} \]

\[ H_{c1} \quad \text{and} \quad H_{c2} \]

\[ H_{ex} \quad \text{and} \quad H_{c} \]

\[ H_{ex} \quad \text{and} \quad H_{c} \]

\[ H_{ex} \quad \text{and} \quad H_{c} \]
III. EXPERIMENTAL RESULTS AND DISCUSSION

In Fig. 2, the temperature dependence of the $H_{ex}$ and

$H_c$ of some representative samples are shown for various

IrMn layer thicknesses. The $H_{ex}$ of all the samples falls

with temperature and vanishes at $T_B$. The $H_c$ of the

pinned Co layers shows a peak at $T_B$ and a sharp up-

turn as very low temperatures are approached. There is

no feature in the $H_{ex}$ plot that accompanies this diver-

gence below ~ 20 K. The free layer coercivity increases

approximately linearly from its room temperature value

of 30 Oe to 50 Oe at 2K. It is important to note that the

pinned layer coercivity at high temperatures matches

that of the free layer very closely, indicating comparable

intrinsic magnetic properties. With this in mind, it can

be seen that the trends observed in the biased Co layers

are therefore not due to the magnetization or anisotropy

constant of the Co itself varying significantly with tem-

perature. Hence the changes must be occurring as a re-

sult of what is occurring in the AF layer.

Naïvely, one might expect $H_{ex}$ to fall off with tem-

perature in proportion to the AF order parameter of the

IrMn, going to zero at the Néel temperature ($T_N$). From

Fig. 2(b) this is not obviously the case, the $H_{ex}$ is found
to decrease approximately linearly to zero at the $T_B$ for
the sample containing the 14 Å thick IrMn layer.

Similar peaks in $H_c$ at $T_B$ observed in oxidized NiFe

layers have been explained using a thermal fluctuation

model by Fulcomer and Charap. They assumed that the

small oxide particles only coupled to the magnetic

NiFe film and were independent of each other. In this

form the varying sizes of particles would lead to super-

paramagnetism. In the system studied here, it would be

very unlikely that the grains would behave independently

of each other because of the dense, continuous nature of
the film. A more recent mean field theory by Wee et al.,
which considers an epitaxial system, has shown similar
results for uncompensated interfaces based on a parallel

domain wall. They predict a blocking temperature that

arises due to thermal dissipation of a domain wall, where

the $T_B$ is the temperature at which the domain wall is

no longer sustainable. Stiles and McMichael give a de-

scription of peaks in $H_c$ which combines aspects of both

theories described above using a mesoscopic model

assuming effective magnetic moments for the grains of
the AF. In general what is important is that in this

range of temperatures, a large fraction of the hysteretic

losses occur in the AF layer, giving rise to an enhanced

coercivity.

That an interaction between the two layers still exists

above $T_B$ can be seen from the significantly enhanced $H_c$
as shown in Fig. 2(a). The true nature of this interaction

is open to debate. In models such as those mentioned

above, as well as our own, above $T_B$ the entire spin struc-
ture of the AF layer reverses with the ferromagnet as the
energy barrier holding it in place can be overcome by the

torque exerted by the F spins once it has dropped below

a certain height. One might believe that the temperature

at which $H_c$ drops to the intrinsic value seen in the free

layer sets a lower limit on $T_N$. However, one cannot

exclude that an AF even in its paramagnetic phase has

some influence on the reversal of the F. The AF expe-

riences an interfacial exchange field originating from the

FM and a paramagnet in a field has a magnetization. Of

course, it must follow the F during reversal, which means

that it is reversible, and will yield zero $H_{ex}$ but possibly

an enhanced $H_c$.

Experimentally, there are two temperatures which are

important. These are $T_B$, which is the point at which $H_{ex}$
tends to zero, and the temperature at which the $H_c$ of

the biased layer falls to that of the free layer which we assume
to be just below or equal to $T_N$, subject to the caveats

given above. The size and width of the coercive peak is

found to decrease, as does the difference between $T_B$ and

$T_N$, as the IrMn layer is made thicker. This effect is simi-
lar to that reported recently in the MnF$_2$/Fe system by

Leighton et al., where the peak in $H_c$ was observed to

be suppressed for very high AF layer thicknesses. This
was explained within the Stiles-McMichael model as being determined by within which layer (AF or F) the hysteretic losses predominantly occur. This would seem to imply that some energy term which is volume dependent is becoming more important in determining $T_B$. We find that $T_B$ and $T_N$ converge at 550 K, the $T_N$ of bulk IrMn, for very thick IrMn layers. Until recently, it was thought that the reduction in $T_B$ due to a lowering of the AF layer thickness was a consequence of a reduction in the ordering temperature of AF ($T_N$). This was assumed to be a result of finite-size scaling effects. However, it has been shown in neutron diffraction studies that $T_B$ does not follow $T_N$, and moreover superparamagnetic effects due to grains cannot fully explain the behavior.

The IrMn thickness and temperature dependence of both the exchange bias field and coercivity are shown in Fig. 3. At room temperature the onset of exchange biasing appears at a critical AF layer thickness of 21 Å and continues to increase up to a layer thickness of 40 Å, where it saturates at a value of 270 Oe. These findings are similar to results obtained previously that were explained by the idea of a partial domain wall in the antiferromagnet parallel to the interface. On this basis one would expect a wall thickness in the IrMn of only some 40 Å. In contrast, as the temperature is decreased down to 2K, which removes almost all thermally activated processes, one finds that the critical thickness for the onset of biasing is less than 14 Å, our thinnest sample. At about this thickness $H_{ex}$ rapidly increases to 950 Oe where it peaks at 20 Å, before falling to a constant value of ~620 Oe above 40 Å. At this stage one might consider the possibility that the variations in the value of $H_{ex}$, with their steep temperature dependence, were related to the discontinuity of layers thinner than $t_{AF} \approx$ 40 Å. Under the same growth conditions used to prepare the IrMn layers in our spin-valves, we deposited IrMn on a piece of thermally oxidised Si whilst making an in-situ four-point resistance measurement. This film was subsequently subjected to x-ray reflectometry to determine its total thickness and hence the rate of deposition. We found that the film becomes conducting (viz. the percolation transition takes place) at an equivalent thickness of ~11 Å. Whilst complete continuity will require a few Å more, the superior wetting of metal-on-metal growth in our spin-valve structures will mean that a few Å less are needed. These corrections will roughly cancel, leaving us with a $t_{AF}$ for continuity well below the thickness where variations in $H_{ex}$ set in at any temperature.

The exchange field at low temperatures exhibits very similar features to the predictions of the random field model proposed by Malozemoff. The low thickness onset of ~10 Å for exchange biasing questions the existence of parallel domains – can the wall be so thin? – whilst the overall shape of $H_{ex}(t_{AF})$ favors the idea of an in-plane domain structure due to interface roughness due to volume defects. A similar peaked form to the $H_{ex}(t_{AF})$ curve was observed at room temperature in a very similar materials system (IrMn/CoFe), but in that instance the peak was ascribed to the variation of (111) texture with IrMn thickness. These samples differ from our own in that the IrMn was deposited prior to the CoFe that is to be pinned, whilst it was also buffered with a magnetic material, permalloy, so some differences in microstructure are to be expected. Since we are able to reversibly suppress the peak in our data simply by varying the temperature, it seems that its presence is related to variations in the magnetic, rather than the physical microstructure of our spin-valves. The complex behavior within this set of data cannot be fully explained using existing models.

Turning our attention to the coercivity of these layers, it is interesting to note that the $H_c$ begins to increase with the IrMn layer thickness before any sign of biasing at all temperatures, as seen in Fig. 3(b). As this occurs even at 2K, we believe that this is inconsistent with the idea of the $H_c$ peak arising from superparamagnetic effects, as the thermal fluctuations will be too small. The AF interfacial and bulk spin structures are
reversible until the AF anisotropy energy barrier is sufficiently large to stabilize them against torques from the rotating F spins to which they are coupled. This leads to additional energy dissipation in the AF layer, manifesting itself as additional coercivity. The peak in coercivity correspondingly shifts to smaller thicknesses consistent with the exchange bias onset. It is clearly demonstrated that the peaks found in the enhanced coercivity are an intrinsic part of the exchange bias. Comparing this discussion of the peak in $H_{\text{ex}}$ with $t_{\text{AF}}$ with that given above for the peak in $H_{\text{ex}}$ with $T$ it is clear that it is possible to draw parallels between the manner in which biasing appears as function of $t_{\text{AF}}$ and the way it vanishes at $T_B$ — there is a conceptual similarity between rising temperature and falling AF layer thickness.

IV. MODELLING

Naturally it is desirable to make the qualitative arguments above more rigorous. The most striking feature of the experimental data is the appearance of the peak in $H_{\text{ex}}$ at low $t_{\text{AF}}$ and its suppression on warming the sample. In the past, only the model of Malozemoff$^{13}$ predicted such a peak, but since it is a zero temperature model it cannot account for the changes as the temperature is raised. We have therefore made use of the so-called domain state model$^{19,25,26}$, which also takes into account disorder in the sample, but makes use of Monte Carlo techniques to treat finite temperature. The model consists of one monolayer of a ferromagnet (F) and $t$ monolayers of antiferromagnet (AF) (see Ref. 26 for a sketch of the model). To include a certain amount of structural disorder the AF is diluted or, in other words, a fraction $p$ of randomly chosen sites of the AF are left without a spin. The F is exchange coupled to the topmost layer of the AF. Previously the model has been used to simulate experimental systems where the AF is deliberately diluted by growth using Mg impurities or or oxygen deficiency in CoO$^{19}$. However, at the heart of the model is the idea that a number of exchange bonds in bonds in the AF are broken at random, leading to places where domain walls can form at a reduced energy cost. In the experimental systems where the AF is deliberately diluted by growth using Mg impurities or or oxygen deficiency in CoO$^{19}$, during a field cycle the interface magnetization of the AF displays a hysteresis, following the F due to the interface coupling. Additionally, the whole curve is shifted vertically due to the fact that after field cooling the AF is in a domain state with a partly frozen surplus magnetization. This shift of the entire hysteresis loop of the AF proves the existence of an irreversible magnetization stored in the AF domains. While the irreversible domain state magnetization of the AF acts as an additional effective field on the F, resulting in exchange bias, the reversible part...
magnetization and, hence, the bias field. The corresponding domain wall energy increases with the interface extending through the whole AF layer. Above this thickness the domain state magnetization in the AF is reversible, giving rise to additional coercivity without bias. As the temperature \( T \) of the system plays a crucial role for the stability of the AF interface magnetization. As we increase \( T \), the interface becomes more and more unstable because of enhanced thermal fluctuations. Consequently, more monolayers of AF are required to stabilize the interface, and hence the peak is shifted towards higher values of \( t_{AF} \) at higher temperatures. However, beyond a certain temperature the interface becomes too unstable and therefore the exchange bias disappears. Thus the blocking temperature, defined as the temperature at which exchange bias disappears, can be interpreted as the temperature above which the interface monolayer of the AF loses its stability during hysteresis. There are examples in the literature of materials systems where the peak is not observed at any temperature, and we are able to adjust our model to account for this simply by varying the value of \( p \).

The behavior of the coercive field is qualitatively different (see Fig. 4(b)). It follows the reversible part of the interface magnetization of the AF because it is influenced by the uniaxial (rather than unidirectional) anisotropy of the AF which, via its interface magnetization, is transferred to the F. This effect depends on the amount of interface magnetization only, and not on its stability which – as explained above – is important for the exchange bias field. Consequently, the coercive field is greater for thinner layers even for lower thicknesses where the exchange bias field is shrinking. For higher temperatures, however, the AF might already be paramagnetic for the lowest thickness we simulated leading neither to exchange bias nor to an enhanced coercivity. When compared to the experimental data of Fig 3 (b), we can see that again the theory reproduces the experimentally observed phenomena: the coercivity falls with rising temperature, and a peak is observed in \( H_c \) at the onset thickness of exchange bias. Only the very sharp upturn in \( H_c \) at low temperatures seen in the experimental data is not fully reproduced within this model, and here further refinements are necessary.

V. IN CONCLUSION

To conclude, we have shown that the AF thickness dependence of the exchange bias, when studied at various temperatures, yields a very rich phenomenology. At low temperatures \( H_{ex} \) sets in at very low thicknesses and then passes through a sharp peak before decaying to a constant value that is maintained out to the largest thicknesses we have measured. As the temperature is raised, the onset thicknesses rises, the peak moves to higher thicknesses and is gradually suppressed, and the high thickness value falls. Any theory that hopes to describe exchange bias should be able to reproduce all the observed features. In addition to the difficulties associated with fitting a planar domain wall into an AF layer only

![Graph](image-url)
14 Å thick, such models are unable to succeed in predicting a peak in $H_{ex}$. Comparison of the experiments and calculations presented in this paper strongly favors the idea of an in-plane domain structure being an essential ingredient for a realistic description of biasing. Coercivity enhancement at the onset or disappearance of $H_{ex}$ (due to either thickness or temperature) is intrinsic to a peak in 14 Å thick, such models are unable to succeed in prediction these effects together.

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