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Topical Review

A new paradigm for exchange bias in polycrystalline thin films

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ABSTRACT

In this paper we provide a review and overview of a series of works generated in our laboratory over the last 5 years. These works have described the development and evolution of a new paradigm for exchange bias in polycrystalline thin films with grain sizes in the range 5–15 nm. We have shown that the individual grains in the antiferromagnetic (AF) layer of exchange bias systems reverse over an energy barrier which is grain volume dependent. We show that the AF grains are not coupled to each other and behave independently. Understanding this process and using designed measurement protocols has enabled us to determine unambiguously the blocking temperature distribution of the AF grains, the anisotropy constant (K_{AF}) of the AF, understand the AF grain-setting process, and predict its magnetic viscosity. We can explain and predict the grain size and film thickness dependence of the exchange field H_{ex}. We have also studied interfacial effects and shown that there are processes at the interface, which can occur independently of the bulk of the AF grains. We have seen these effects via studies of trilayers and also via the field dependence of the setting process which does not affect the blocking. From separate experiments we have shown that the disordered interfacial spins exist as spin clusters analogous to a spin glass. These clusters can order spontaneously at low temperatures or can be ordered by the setting field. We believe it is the degree of order of the interfacial spins that gives rise to the coercivity in exchange bias systems. Based on this new understanding of the behaviour of the bulk of the grains in the antiferromagnet and the interfacial spins we believe that we have now a new paradigm for the phenomenon of exchange bias in sputtered polycrystalline thin films. We emphasize that the phenomenological model does not apply to core–shell particles, epitaxial single-crystal films and large grain polycrystalline films.

1. Theories and models of exchange bias

Since its discovery in 1956 [1] the phenomenon of exchange bias has been one of the most fascinating and complex effects that have ever been studied in the field of magnetism. After more than 50 years there is still no definitive theory that can account for the observed effects which include the well-known hysteresis loop shift (H_{ex}) and the increased coercivity (H_c) defined as half the loop width, when an AF material is placed in intimate contact with an *F* material and field cooled through its Néel temperature (T_N). One of the reasons why no clear and comprehensive theory has been developed lies in the fact that a wide range of samples has been studied. These include nanoparticles where obviously the AF/F interface is not flat [2], epitaxially grown films [3] where the interface is expected to be almost flat, and sputtered polycrystalline films [4] where the interface has significant roughness and can lead to both structural and magnetic disorder. Interestingly the largest exchange bias effects at room temperature are observed in polycrystalline granular films produced by sputtering and it is these materials exclusively that are used, or proposed, for applications in devices such as magnetic recording read heads and MRAM applications. It is such sputtered metallic polycrystalline films alone which this study addresses.

In a major review of exchange bias published in 1999 in Vol 200 of this journal entitled "Magnetism Beyond 2000" Berkowitz and Takano provided a comprehensive review of the known state of the art in this field [5]. Interestingly they state "*A model of the exchange bias mechanism must resolve the following discrepancies and questions:*

(1) *What structural and magnetic parameters are responsible for the drastic reduction of the interfacial exchange energy density from the ideal case?*
(2) *What are the origin and role of the interfacial uncompensated AF spins?*

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(3) *How is the magnitude of the exchange field dependent upon the AF grain structure?*
(4) *What determines the temperature dependence of the exchange field?*
(5) *What are the roles of interfacial exchange $J_{ex*}$and AF magneto-crystalline anisotropy K_{AF}in unidirectional anisotropy?"*

In this work we now attempt to answer these five questions based on a new conceptual paradigm of the exchange bias effect in polycrystalline materials.

1.1. Early theories of exchange bias

The first and simplest model proposed to explain the phenomena of exchange bias is that of Meiklejohn and Bean [1]. In their paper they studied single-domain spherical Co particles with and AF CoO shell. These particles had uniaxial anisotropy and their easy axis aligned parallel to the applied field. They assumed a perfectly uncompensated AF spin structure at the interface, which remains rigidly aligned along its easy axis due to a large anisotropy in the AF and a weaker exchange coupling of the AF to the F. This mechanism of exchange bias leads to values of the shift in the hysteresis loop, H_{ex}, two orders of magnitude larger than the observed values in small grain polycrystalline films although it predicts near correct values in other systems e.g. [6–8]. The original data of Meiklejohn and Bean is shown in Fig. 1.

Chronologically, the second model proposed to explain the exchange bias phenomenon was suggested by Néel [9]. Néel proposed an uncompensated AF spin structure at the interface.

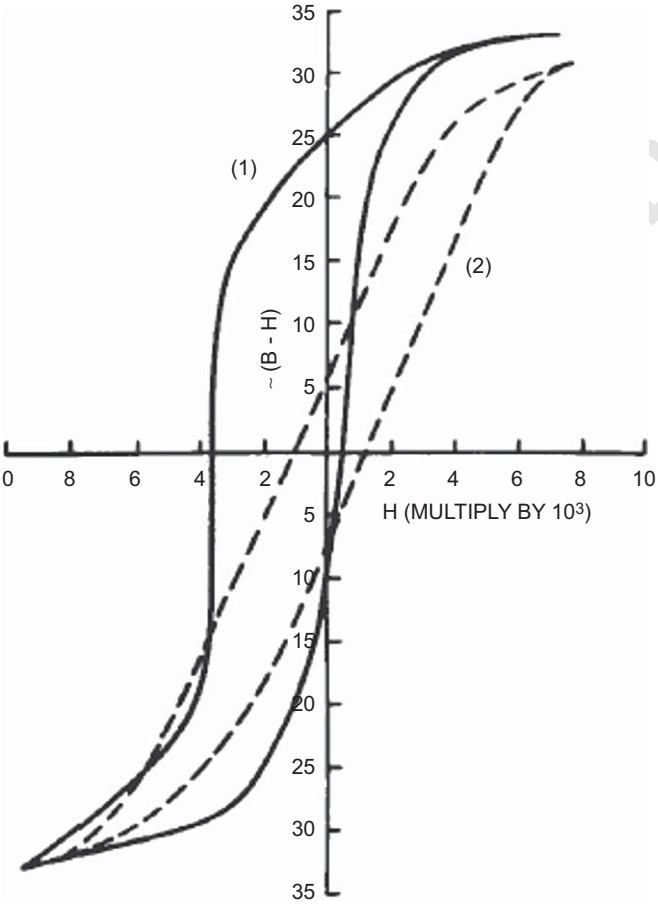

Fig. 1. Hysteresis loop of the Co particles embedded in their natural oxide measured at 77 K after field cooling in a 10 kOe magnetic field (solid line) [1].

However, he pointed out that this spin structure is subject to deformation and experiences irreversible changes during the reversal of the F layer. Consequently, both the exchange field, H_{ex}, and the coercivity, H_c, are affected by changes which occur in the AF during the reversal of the F. From his model, two contributions would be expected to H_c; an intrinsic F component and a term that would be proportional to the irreversible magnetisation changes which occur in the AF. Also, Néel considered that for realistic rough interfaces both AF sub-lattices would be present at the interface, leading to partial compensation of the AF moments. In the case of polycrystalline AFs the number of spins at the interface of each AF grain would be statistically distributed, leading to fluctuations in the moment of each AF grain. This model again fails to predict reasonable values for H_{ex}.

One of the most successful models of exchange bias is that of Fulcomer and Charap [10,11]. These workers undertook both experimental and theoretical studies of exchange bias in permalloy films where a treatment with acid vapour progressively oxidized the nickel in the alloy producing isolated AF grains on the surface of the films. They observed progressive changes in the exchange bias of such systems with both the grain size and the number of grains of AF material grown. Numerical modelling based upon a granular reversal model analogous to a Stoner–Wohlfarth system gave good agreement with experimental observations. In particular, Fulcomer and Charap predicted that the exchange field from the F acting on the AF would result in thermally activated changes in the orientation of the AF sub-lattices leading to variations in the observed value of H_{ex}. One important characteristic of this model is that a wide range of particle sizes and shapes was considered within the AF. This way the anisotropy and coupling energies were varied widely. The particle size distribution was assumed to be such that all areas are equally probable up to some maximum and that there were no larger particles. They found that it was important to consider a distribution of particle size although the exact form of that distribution was not critical. This model was able to predict the temperature dependence of H_{ex} and H_c over a wide range of temperatures even above T_N as reported by Grimsditch et al. [12]. This model has formed the base of other granular models based on thermal fluctuation effects. For instance, Nishioka et al. studied the temperature dependence of exchange bias in NiFe/FeMn [13] and Co/CrMnPt [14] using the temperature dependence of the coupling proposed by Fulcomer and Charap. More recently, Xi [15] proposed a thermal fluctuation model to study the dependence of the blocking temperature (T_B) in exchange bias bilayers also based on the work by Fulcomer and Charap. Xi found a monotonic increase in H_{ex} with AF layer thickness (t_{AF}) for large grain systems ($D \sim 40$ nm). He also found a linear decrease of T_B with measurement time and a linear increase with the Néel temperature of the AF grains. This model is significant because it extends the work of Falcomer and Charap into a more complete description of thermal effects. However, the calculations are for a single-grain volume and hence do not describe real systems.

In 1987, Mauri et al. [16] proposed the first domain model of exchange bias. They suggested that the formation of domain walls parallel to the F/AF interface results in a lower interfacial energy than that predicted by the model of Meiklejohn and Bean. This results in more reasonable values of H_{ex}. However, this model does not explain features such as the enhanced H_c of the F layer, or the reduction of the loop shift upon field cycling. One of the main assumptions of this model is that the AF spin sub-lattices and the F spins lie parallel to a perfectly flat interface. This model also relies on thick AF layers necessary to accommodate a domain wall parallel to the interface, although exchange bias has been reported for systems containing AF layers as thin as a few atoms.

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Malozemoff [17] introduced a random interface roughness between the F and AF layers and predicted values of H_{ex} of the same order of magnitude as those obtained by Mauri et al [16]. The rough nature of the interface gives rise to compensated and uncompensated areas at the interface that exert fields on the F interfacial spins. The magnetostatic energy of these fields and the anisotropy of the AF result in the formation of AF domains with domain walls perpendicular to the interface. The movement of such AF walls would explain the reduction of H_{ex} due to field cycling. However, this model is only applicable to single-crystal AFs. In the case of polycrystalline materials it is expected that more complex behaviour would result which would depend on the microstructure of the AF. Also, this model relies on surface roughness and does not explain the appearance of exchange bias on perfectly compensated interfaces.

Koon proposed a microscopic explanation of exchange bias for compensated F/AF interfaces [18]. In this model, the F magnetisation tends to orient perpendicular to the AF easy axis. Both compensated and uncompensated interfaces were predicted to lead to similar values of H_{ex}. These results do not concur with the theories of Mauri [16] and Malozemoff [17]. The main result of Koon's model was the prediction of spin–flop coupling at the interface between the F and AF layers. Exchange bias was due to the formation of a domain wall in the AF parallel to the interface when the F rotates away from the field cooling direction as the applied field is reversed. Furthermore, Koon also showed that the spins in the AF exhibit canting. This canting angle decays rapidly as a function of distance from the interface, becoming zero at 5–6 monolayers.

Schultness and Butler [19] solved the Landau–Lifshitz–Gilbert equation in order to study the exchange coupling at F/AF interfaces. They showed that for perfectly flat interfaces, spin–flop coupling does not lead to exchange bias although it leads to an increase in the F layer coercivity typical of exchange bias systems. The introduction of defects at the interface leads to values of the exchange field of the correct order of magnitude [20]. However, their model only applies to idealised situations in which both the AF and the F layers are single crystals and in a single-domain state.

After the turn of the millenium when Berkowitz and Takano posed their five questions [5] the use of large-scale atomistic models of magnetic systems became more common. This provided the opportunity for models of realistic spin systems to be used to predict the complex multifaceted behaviour of exchange-biased systems. Prior to this time all theories were either analytical or micromagnetic.

1.2. Recent theories of exchange bias

In 1999 Stiles and McMichael proposed a model to describe the behaviour of polycrystalline F–AF bilayers [21]. No intergranular exchange coupling in the AF was assumed. The AF grains were coupled to the F layer both by direct exchange to the net moments at the interfaces of the grains and by spin–flop coupling. In order to account for high field rotational hysteresis and the loop shift, some of the grains were assumed to make irreversible transitions. These transitions were included in the model in the form of a critical angle above which the AF becomes thermally unstable and undergoes a transition to another state. The temperature dependence of the loop shift was assumed to arise due to thermal instabilities in the state of the AF grains. At low temperatures, the AF grains remain in a stable configuration as the F layer magnetisation is rotated. At high temperatures, the AF grains become thermally unstable over long timescales due to thermal excitations over energy barriers. The model is valid for systems where the Curie temperature of the F layer is much greater than the ordering temperature of the AF [22]. Hence, the properties of the F layer were assumed to be temperature independent. The AF grains were classified as stable, partially stable and unstable as a function of temperature and field direction. The behaviour of each grain determines its contribution to the unidirectional anisotropy. The saturation of the unidirectional anisotropy at low temperatures is determined by the ratio of the average spin interfacial coupling energy to the zero temperature domain wall energy.

Based on this model, Stiles and McMichael found two different contributions to the enhanced coercivity: one was due to inhomogeneous reversal and the other to irreversible transitions in the AF layer [23]. Two different dependencies of H_c on the thickness of the F layer were predicted as a function of temperature based on the contribution from each term.

Stamps suggested the existence of two different mechanisms for exchange bias [24]. The first mechanism is due to the reversible formation of domain walls in the AF. A second contribution arises due to irreversible processes leading to asymmetric hysteresis loops. A key result of this model is the existence of higher-order coupling terms when more than one AF sublattice is present at the interface. The concept of a natural angle suggested by Camley et al. [25] was introduced to characterise exchange bias at mixed and geometrically rough interfaces. A mean field theory was used to explain the temperature dependence of H_{ex} and H_c. A thermal activation theory was used to describe the rate at which equilibrium was approached. These energy barriers were considered to be of two types: those involved with in-plane rotation of the magnetisation of the F layer and those related to out-of-plane rotation. There is then no need for anisotropy in the F layer for the appearance of the unidirectional anisotropy in contrast to the rigid models.

In 2002, Nowak et al. [26] proposed the domain state model for exchange bias based on their previous work [27]. The F layer was assumed to be coupled to a diluted AF. The dilutions were introduced in the system in the form of non-magnetic atoms. In this model exchange bias arises due to a domain state in the AF which develops during field cooling carrying an irreversible magnetisation. During the field cooling process, the AF is in contact with a saturated F layer and exposed to an external magnetic field. The dilutions introduced in the system favour the formation of this state since domain walls preferentially pass through the non-magnetic sites and, therefore, reduce the energy necessary to create a domain wall [26]. The domain state is a metastable state which develops and becomes frozen during cooling and depends upon the concentration of non-magnetic sites. This way no further assumptions about the size or structure of these domains are required. Monte Carlo simulations were performed on a model consisting of an F monolayer exchange coupled to a dilute AF film typically consisting of nine monolayers. Note that in this model K_{AF} is taken to be very large and hence the width of these domain walls is assumed to be zero. The model was used to predict several features of exchange bias. For instance, strong dependence of the exchange bias field on dilution, positive exchange bias, temperature dependence of exchange field, etc. The attempt to predict the peak in H_{ex} with t_{AF} was partially successful although the value of t_{AF} at which the peak occurred was only a few atomic layers. The model did not include discretised grains. Hence this model might be valid for single-crystal films but it is unlikely that it can be applied to polycrystalline films where the AF grains are exchange decoupled.

The same model was applied to study the temperature dependence of H_{ex} and H_c using a mean field approach [28]. A significant enhancement of the coercivity was found reaching a maximum around the Néel temperature of the AF layer. The domains formed after field cooling using the mean field approach

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have the same structure as those obtained with Monte Carlo simulations. However, there are differences in the magnitude of the loop shift obtained using both methods. The enhancement of the coercivity was attributed to the coupling of the F layer to the part of the AF interface layer magnetisation that follows the external field during a hysteresis loop measurement. Since the AF layer carries an induced magnetisation above T_N, the enhancement of H_c persists well above the transition temperature. On the other hand, H_{ex} originates in the frozen part of the magnetisation of the AF interface layer. The main conclusion of the work is that the exchange bias and the coercivity are not related.

In 2006, Saha and Victora [29] proposed a large-scale (2.5×10^5 elements) micromagnetic model for polycrystalline exchange bias systems with uncoupled AF grains. Each F grain was coupled to a neighbouring AF grain that rotated uniformly under the effect of thermal fluctuations. The evolution of the F layer magnetisation was determined by solving the Landau–Lifshitz–Gilbert equation for each degree of freedom. The coupling between the F and AF arises due to the surface roughness of the AF grains. The dependence of the exchange bias and coercivity as a function of temperature and AF layer thickness were studied. One of the most important conclusions of this model is that systems with AF symmetry higher than uniaxial exhibit an asymmetric magnetisation reversal when compared to a sample with uniaxial anisotropy. The model also predicts an increase in H_{ex} at reasonable values of t_{AF} but does not predict the broad peak commonly observed.

Recently, Choo et al. [30] proposed a granular model of the magnetic properties of exchange biased F–AF bilayers. The F layer consists of strongly exchange coupled grains. The magnetisation reversal processes involve non-uniform magnetisation states. The F layer was modelled using a standard micromagnetic approach. On the other hand, the AF layer was formed of highly anisotropic exchange decoupled grains. The F and AF layers are coupled by magnetostatic and exchange interactions. The magnetic state of each AF grain is controlled by thermally activated processes. Hence, a kinetic Monte Carlo approach was used to model the properties of the AF. As in the model of Fulcomer and Charap [10,11], the energy barrier to reversal for each AF grain is given by the product of its volume and anisotropy constant, taking into account the orientation of the grain with respect to the local magnetic field. The probability for each grain to undergo magnetisation reversal is given by the Néel–Arrhenius law. The magnetic state of the AF layer is represented by an order parameter (P). This order parameter is indicative of the magnitude and sign of the exchange field acting on the F layer. For their calculations, a value of 4×10^6 ergs/cm^3 was assumed for the anisotropy of the AF layer. The distribution of grain sizes within the sample was taken to be lognormal. It was shown that thermal instabilities in the AF layer lead to a peak in the coercivity around the median blocking temperature.

In a more recent work, the same group studied the temperature dependence of exchange bias in bilayer systems [31]. This version of their model takes into account the temperature dependence of the anisotropy of the AF. It was predicted that the measured value of the median blocking temperature depended on the strength of the intrinsic interlayer coupling due to the reduction of energy barriers. This dependence was found to be rather weak for normal values of the coupling strength.

Despite the extensive and often complex attempts to model exchange bias systems there is still no theory that is able to predict the value of exchange bias in any real system. Critically the theories and models provide no guide to those trying to develop new materials and systems for applications. For example, what is the optimum grain size for a given application? None of the models predict the film thickness and grain size dependence of H_{ex}. Similarly what is the role of the anisotropy or interface structure? Significantly none of the models come close to answering the five questions posed by Berkowitz and Takano [5] in a comprehensive way.

One reason for the failure of the models is the wide variety of samples that have been studied. The second reason is that most experimental data have been taken at arbitrary temperatures without regard to possible thermal instabilities in the AF layer. Similarly the degree of order in the AF at the beginning of the measurement is generally unknown and rarely checked.

We now describe a series of experiments undertaken taking into account these factors. We have studied sputtered films mainly of IrMn/CoFe, which is the system used exclusively in applications.

2. Experimental studies of polycrystalline exchange bias systems

There is a plethora of data in the literature relating to exchange bias systems. As indicated in the introduction there are a number of categories of exchange bias systems including nanoparticles, epitaxial films and polycrystalline films. The work described in this paper is restricted to metallic polycrystalline films with small grains between 5 and 15 nm and hence the brief review of existing experimental work applies only to these materials. Furthermore we concentrate on the key issues of grain size, film thickness and thermal effects.

When comparing previous measurements with our more recent studies a significant difficulty occurs because until recently almost all authors did not take care to ensure that the AF layer was fully set. To our knowledge, in no other published work has an experimental measurement been undertaken to check that the AF layer was fully set before the measurement began. Furthermore whilst some authors have clearly been aware of the thermal activation process affecting the order in the AF, no systematic methodology has previously been applied to quantify and more importantly control, thermal processes in the AF during measurements. Nonetheless there are a number of important works which have provided signposts to the resolution of these issues.

For example, following on from the initial work of Fulcomer and Charap [10,11], van der Heijden et al [32] discussed in some detail the time dependence of H_{ex} at different temperatures. Interestingly the published data appears linear in ln t although the authors did not plot the data in this form. In a separate work the same authors showed that holding the ferromagnet in a direction opposite to that in which it was originally set can reverse the shift in the hysteresis loop including a reversal in the sign of H_{ex}[33]. Unfortunately the original state of order in the samples was not verified but the concept of reversal of the AF in the presence of a reversed exchange field under the influence of thermal energy is clearly defined.

One of the critical variables for applications of exchange bias is the variation of the value of H_{ex} with the thickness of the AF layer (t_{AF}). It is widely accepted that this variation has $H_{ex} \propto 1/t_{AF}^{\lambda}$. There are reports that the value of λ ranges from 0.3 [34] to 1 [35]. However, as noted in these works this inverse relationship between H_{ex} and t_{AF} only occurs for large thicknesses. At low thicknesses an increase in H_{ex} with t_{AF} is observed. This is assumed to be due to the existence of a critical thickness at which domains can form in the AF although there is no experimental evidence for the presence of such entities. The domain state model [26] is able to predict a peak at extremely low t_{AF} but the remainder of the variation is completely different to that which is observed experimentally.

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The situation regarding the grain size dependence of H_{ex} in polycrystalline films is even more confusing. For example Takano et al. [36] reported a decrease in H_{ex} with increasing AF grain size (D) in NiFe/CoO bilayers following $H_{ex} \propto 1/D$. Uyama et al. [37] observed a decrease in H_{ex} with increasing grain size above 50 nm but also observed the well-known broad peak in the variation of H_{ex} with t_{AF}. Again for large grain sizes an approximate $1/t_{AF}$ variation was observed. The peak in the variation of H_{ex} with t_{AF} was again attributed to the onset of AF domain formation in agreement with a prediction of the domain state model although the experiment preceded the theory. Thus there is not clearly an established description of the effect of grain size and film thickness on the value of H_{ex}. Also other than speculation as to the onset of AF domain formation, there is no clear mechanism to account for these variations, particularly the $H_{ex} \propto 1/t_{AF}$ form observed by many groups, but not predicted by the domain state model [26].

Whilst there is no agreement from different measurements of exchange bias there have been a significant number of papers which are in broad agreement that the AF grain size increases the thermal stability in exchange bias systems [38]. There is also evidence that the AF grain anisotropy also has a direct impact on the value of the mean blocking temperature. Hence this is perhaps an indication that the phenomena of thermal stability and the actual value of H_{ex} achieved are not directly connected to each other. It will be shown subsequently that this is the case.

It should be noted that the foregoing brief review of experimental data is not intended to be comprehensive and is specifically intended to be focused on polycrystalline films generally produced by sputtering and generally consisting of metallic materials with small grains. Also the literature which has been surveyed has been selected so as to focus on the topics which are discussed in the next section of the review.

2.1. The York Protocol

There are a number of challenges presented in trying to study the exchange bias phenomenon. The first of these is that for materials having technological applications it is difficult to heat the sample to the Néel temperature of the AF layer. For all current applications the alloy $IrMn_3$ is the material of choice. This material has T_N equal to 690 K and this temperature would result in diffusion in multilayer films. However it has been found that field cooling from temperatures as low as 475 K can result in the setting of IrMn layers. This setting process is achieved by the thermal activation of the orientation of the AF lattice within each grain as first described by Fulcomer and Charap, albeit for the disorientation of the AF lattice. A further challenge comes about because of the thermal instability of AF grains themselves. As shown by Fulcomer and Charap the state of order in the AF can change during a measurement, making parameters such as H_{ex} and H_c highly non-reproducible. To overcome these challenges careful management of the thermal and magnetic history of the AF is essential to ensure that a uniform state at the beginning of a measurement can be reproduced in a subsequent measurement to allow for comparability.

The second challenge when studying exchange bias is that the AF layer itself gives no signal in a conventional magnetic measurement. All that can be achieved is that careful control of the thermal and magnetic history allows one to infer changes in the state of the AF grains, which then give rise to changes in the properties of the adjacent F layer. In one sense this is not dissimilar to the study of black holes in astronomy which cannot themselves be seen but their effects on neighbouring objects can be discerned.

We have designed a set of magnetic field and temperature protocols that enables reproducibility of measurement to be achieved to high resolution. These protocols allow us to determine and control the state of order in the AF both prior to, and during each measurement. They also allow for the degree of order in the AF to be varied in a controlled and systematic manner.

The basis of the York Protocol for the measurement of blocking temperatures is shown schematically in Fig. 2a. The precise sequence of the measurement is shown in Fig. 2b. At the time of starting a measurement the state of order in the AF is generally unknown. Often polycrystalline thin films are sputtered in the presence of a magnetic field. This field induces an easy axis in the F layer but the magnetisation of the F layer during growth and the inevitable heating can lead to the establishment of some degree of order in the AF layer which is generally deposited first. However, from measurements undertaken in York, but not reported here, we have found that this initial state of order in the AF is highly non-reproducible.

Thus the first requirement is to set the AF in a reproducible manner. This is achieved by heating the sample to increasing temperatures with a magnetic field sufficient to saturate the F layer applied in the known direction of the F layer easy axis, to the maximum temperature (T_{set}) which does not result in interfacial diffusion. The onset of significant diffusion can be observed by heating for different periods of time and observing generally adverse changes in the resulting exchange bias. For reasons which will become clear, we have found that a setting time (t_{set}) of 90 min is adequate for all samples we have examined. A time of 90 min has been used because, as shown in Section 2.6, the variation of H_{ex} with the time of setting (t_{set}) is linear in $\ln t_{set}$. After a time of 90 min the change of H_{ex} with t_{set} is very small ($< 1\%$) allowing for reproducibility in the value of H_{ex}. This setting process is undertaken with a sample mounted in a conventional vibrating sample magnetometer, which in our case is a Veeco ADE Model 10 system.

For highly detailed measurements the reproducibility of the setting process should be checked by repeating the procedure outlined above. It is now necessary to ensure that no thermal activation occurs during the time of measurement of the systems. This is achieved by cooling down with the setting field still applied, to a temperature at which no thermal activation occurs. We designate this temperature T_{NA}. T_{NA} is determined by having first cooled to a trial T_{NA} and reversing the field so that the F layer saturates in the opposite direction to that used for setting. The sample is then held in that state of magnetisation for a short period of say 1 min and a hysteresis loop measured. The process is repeated but this time the sample is kept with the F layer reversed for half an hour. If the hysteresis loop does not reproduce then thermal activation will have taken place during the measurement and a lower value for T_{NA} must be sought by trial and error. It should be noted here that care must be taken at each stage to measure two hysteresis loops so that the well-known first loop training effect can be removed from the measurement. This training effect occurs on the first hysteresis loop and is believed to be due to some form of spin–flop coupling which is removed by a single hysteresis loop cycle [39] The slow training effect that occurs on the second and subsequent loops has been shown to be due to thermal activation of the AF grains and is removed by measuring at T_{NA}[40].

Once the sample is correctly set and T_{NA} established, reproducible measurements can be obtained. Interestingly it is now possible to undertake controlled thermal activation. This is achieved by first setting the sample and establishing T_{NA}. The magnetisation of the F layer is reversed to negative saturation. The sample is then heated for 30 min in appropriate steps to

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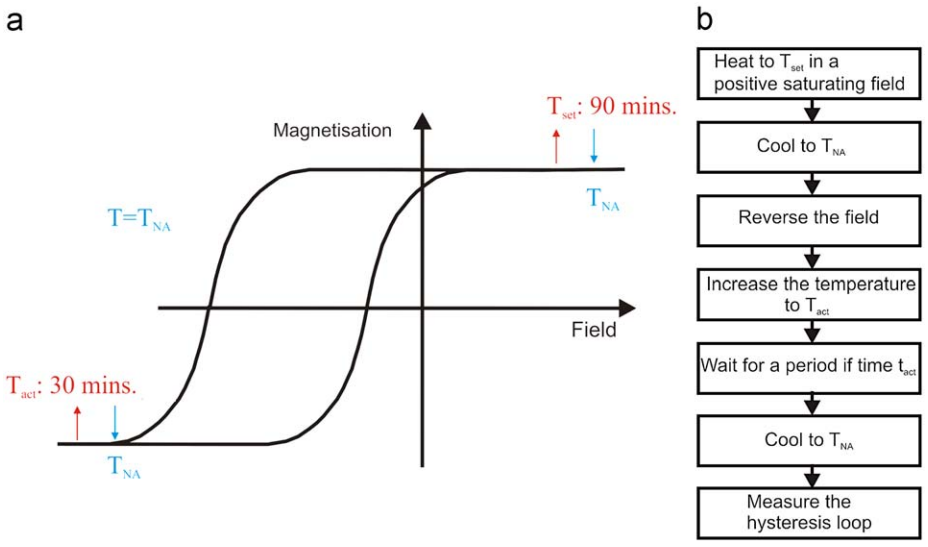

Fig. 2. (colour online) (a) Schematic diagram and (b) measurements steps of the York Protocol.

activation temperatures T_{act}, but subsequently cooled back to T_{NA} prior to the measurement of the complete hysteresis loop starting from negative saturation. Starting from this point removes the first loop training effect and measuring at T_{NA} ensures that slow thermal training does not occur [40].

A thermal activation time (t_{act}) of 30 min was chosen again because of the $\ln(t_{act})$ dependence of the resulting order in the AF layer. It also implies that on resetting the sample between each measurement the setting time of 90 min would completely reverse any activated grains to their original 'set' state. These times also negate any thermal activation that may occur during the temperature rise and fall.

Although the above measurement protocol is somewhat tedious and difficult to observe accurately, we have generated application software on our magnetometer, which enables measurements to be made in a relatively automated manner. Using the York Protocol highly reproducible data can be obtained which has elucidated very detailed measurements of effects both in the bulk of the AF and at the interface.

2.2. Measurement of the blocking temperature

The blocking temperature T_B is conventionally defined as the temperature at which the exchange field goes to zero. The conventional method to determine T_B^{con} is based on the measurement of hysteresis loops with increasing temperature until the loop shift becomes zero. Above T_B^{con}, the exchange bias remains zero. According to Fulcomer and Charap [10], T_B^{con} will correspond to the blocking temperature of the AF grain with the largest anisotropy energy. In polycrystalline systems each grain has its own blocking temperature and therefore the AF is characterised by a distribution of blocking temperatures. When the conventional measurement procedure is used, the AF is subject to thermal activation during the time of measurement at a logarithmic rate [41]. This leads to changes in the state of order in the AF from measurement to measurement as well as lack of reproducibility in the data.

Fig. 3 shows a comparison of the measurement of $H_{ex}(T)$ via the two methods. As expected the standard technique (H_{ex}^{con}) gives a lower maximum value of T_B^{con} and a wider distribution. This is a consequence of the magnetic viscosity in the AF and values of T_B^{con} obtained will depend on the timescale of the measurement. This was predicted by Xi [15] and first reported by van der Heijden et al [32]. The measurement of $H_{ex}(T)$ via the York Protocol ($H_{ex}^{YP}(T)$) gives highly reproducible data which are not subject to timescale effects since all the data are measured at a temperature T_{NA} where the system is free of thermal activation. Note that due to limitations in our new cryostat H_{ex}^{YP} can only be measured down to 100 K.

Thermal activation of the AF means that the hysteresis loop of an exchange biased system can be shifted in the opposite direction to that in which it was originally set in a controlled manner [42]. The York Protocol has been used to measure the *mean* blocking temperatures $<T_B^{YP}>$ of all the samples studied in this work. Examples of the hysteresis loops obtained following this measurement procedure are shown in Fig. 4. Heating with the F layer reversed changes the order in the AF from the original state to the reverse orientation as shown in the schematic diagram in Fig. 5. The amount of AF material that undergoes reversal will be a function of the temperature and the exchange field from the F layer.

The value of H_{ex} is then proportional to the difference in the fractions of the AF grains oriented in opposite directions

$$H_{ex}(T_{act}) \propto \int_{T_{act}}^{\infty} f(T_B)dT_B - \int_{0}^{T_{act}} f(T_B)dT_B \quad (1)$$

Eq. (1) is valid if the coupling between the F and AF grains is independent of the AF grain size. The quality of the fit between our theory and the data, discussed in Section 2.5, shows that this is the case. However, this may not be valid for very thick AF layers where columnar growth is not present.

The York Protocol leads to a different meaning of $<T_B^{YP}>$. In our measurement $<T_B^{YP}>$ occurs at the point where equal fractions of the volume of the AF grains are oriented in opposite senses and hence is a measurement of the median blocking temperature in the system which we denote as $<T_B>$. The conventional measurement actually determines the maximum value of T_B and depends critically on the form of the distribution at high values. All measurements described in the work were obtained via the York Protocol.

2.3. Measurement of K_{AF}

Polycrystalline metallic AFs consist of an assembly of grains distributed in size, typically 10 nm in diameter. Each grain is

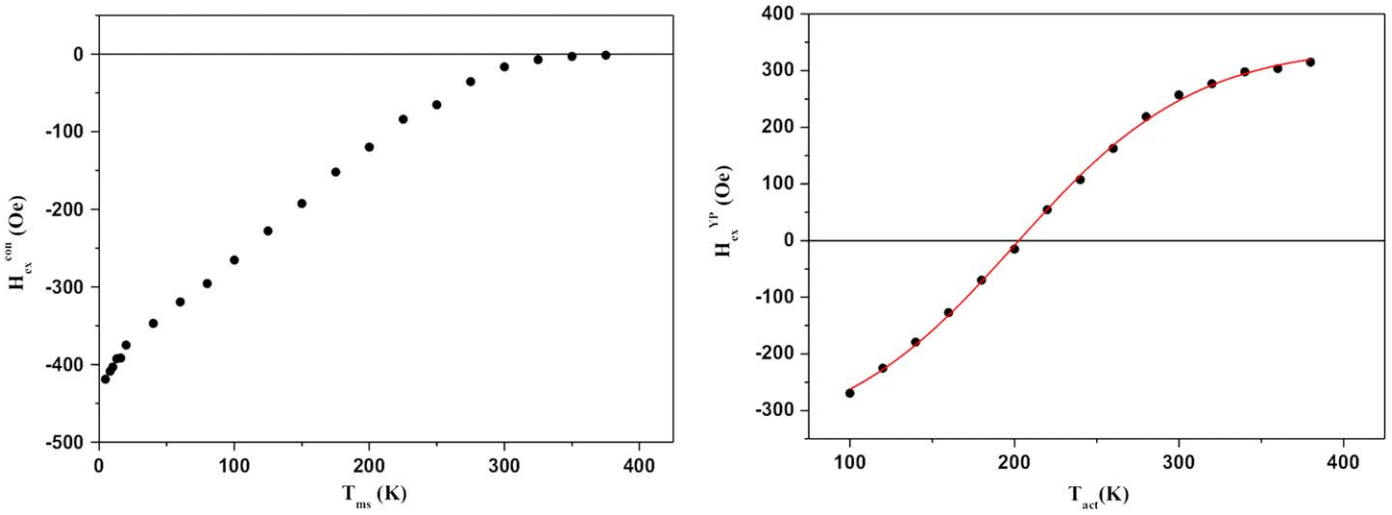

Fig. 3. (colour online) Measurement of the blocking temperature for a CoFe/IrMn bilayer following the conventional measurement procedure and the York Protocols.

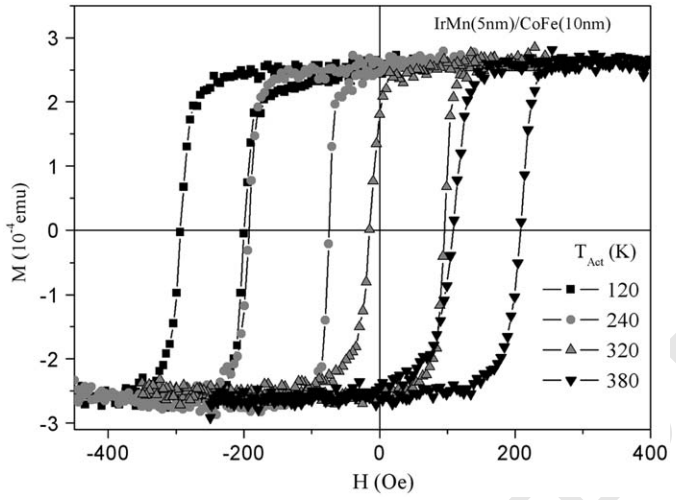

Fig. 4. Examples of hysteresis loops obtained following the York Protocols for a CoFe/IrMn exchange bias system.

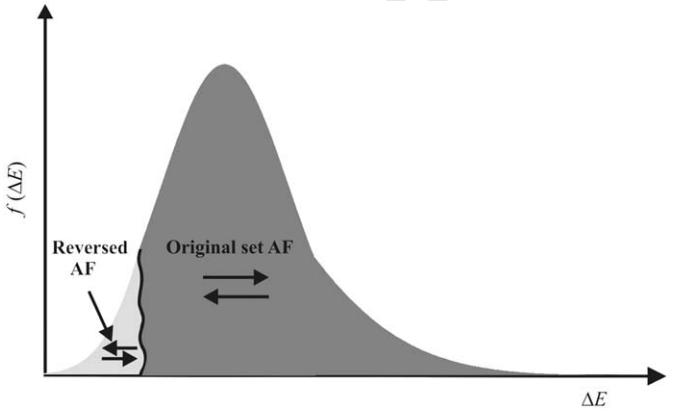

Fig. 5. Schematic of the energy barriers to reversal, showing the fractions of the energy barriers oriented parallel or antiparallel to the original set direction.

believed to contain a single AF domain due to their size. Single-domain F or AF particles are subject to thermal activation leading to a magnetic transition over an energy barrier $\Delta E = KV$[10,11], where V is the volume of the AF grain and K its anisotropy constant. The critical volume below which this behaviour is observed depends on the measurement time and the temperature. The relaxation time is given by the Néel–Arrhenius law as used by Fulcomer and Charap [10].

$$\tau^{-1} = f_0 \exp\left[-\frac{K_{AF}V(1-H^*/H_K^*)^2}{k_B T}\right] \quad (2)$$

where f_0 is taken to be $10^9\ s^{-1}$, V is the grain volume, k_B is Boltzmann's constant, K_{AF} is the anisotropy of the AF grain and T is the temperature. H^* is the exchange field from the F layer which lowers the energy barrier to reversal (ΔE) of the AF grains promoting thermally activated transitions. H_K^* is a pseudo-anisotropy field similar to the anisotropy field (H_K) in ferromagnets. The energy barrier is taken to be $K_{AF}V$ as the limiting case. The value of the pseudo-anisotropy field (H_K^*) and the exchange field (H^*) are unknown. However we assume that H^*/H_K^* is small. We have shown that this assumption is correct for most F/AF systems used in applications [43]. It is important to note that K_{AF} is temperature dependent since its origin is magnetocrystalline. Its temperature dependence is of the form $K_{AF}(T) = K_{AF}(0)(1 - T/T_N)^n$. We have used a value for n of unity based on $K_{AF} \propto [m_{AF}(T)/m_{AF}(0)]^3$ and the approximation $m_{AF}(T) \propto (T - T_N)^{1/3}$ where m_{AF} is the moment of one of the AF sub-lattices [22].

There is no experimental evidence of intergranular exchange coupling between AF grains in metallic polycrystalline AF layers. In ferromagnetic polycrystalline films, such as Co alloy recording media, intergranular exchange coupling occurs via an RKKY type mechanism. This requires a significant moment on each grain to polarise the conduction electrons and the coupling can be reduced or even eliminated by a few atomic layers of antiferromagnetic CoCr alloy at the grain boundary [44]. Hence, AF/AF coupling via an RKKY mechanism is known not to occur. Also, from the contrast observed in bright and dark field TEM images it is clear that our samples are not crystallographically ordered and therefore direct exchange coupling between the AF grains would not occur. Hence, we believe that an independent single-domain AF grain model similar to that of Fulcomer and Charap applies. Also, the size of the AF grains used in all sputtered films having technological applications lies in the range 5–20 nm. At these sizes it is hard to conceive that an AF domain wall could exist within a grain. Indeed, there are no reports of AF domain effects in polycrystalline films.

There have been a number of attempts to measure the value of K_{AF} in polycrystalline AF films. Most notably Mauri et al. [45] determined K_{AF} for FeMn from the value of H_{ex} at room temperature via

$$H_{ex} = \frac{t_{cr}}{t_F}\frac{K_{AF}}{M} \tag{3}$$

where t_{cr} is the thickness of the AF layer above which there is a sharp onset of H_{ex}, t_F is the thickness of the F layer and M its saturation magnetisation. This gave $K_{FeMn}(295)=1.35\times10^5$ ergs/cm^3. There are clearly a number of problems with this methodology. Firstly, unless the measurements are made under thermal activation-free conditions it is unclear what fraction of the AF is stable and contributing to K_{AF}. Similarly, as shown in Section 2.2, there can be an uncertainty as to the fraction of the AF that is 'set' giving a similar discrepancy. This is particularly so for IrMn due to its high value of T_N.

The more serious problem with this technique is the effect of the interface coupling. In all cases this factor is unknown and can only be controlled to a limited degree. However, the York Protocol removes the issue of thermally disordered grains and allow for the set fraction to be assessed. In the H_{ex} versus temperature curves, such as that shown in Fig. 3, the point at which $H_{ex}=0$, is unique since the fraction of the AF oriented in opposite senses is equal. Including the interfacial coupling parameter $C*$, at $<T_B>$, H_{ex} is of the form

$$H_{ex}(<T_B>)\propto C^*\left[\int_0^{<T_B>} f(T_B)dT_B - \int_{<T_B>}^{\infty} f(T_B)dT_B\right]=0 \tag{4}$$

At $H_{ex}=0$ the effect of C^* is negated allowing K_{AF} to be determined at that temperature. This effect may not completely cancel if there is a grain size dependence of the interfacial spin order.

We have undertaken a detailed study of a series of samples with structure Si/Cu(10 nm)/CoFe(5 nm)/IrMn(t_{AF})/Ta(10 nm) (t_{AF}=3, 4, 6, 8, 10 and 12 nm) prepared using a HiTUS sputtering system [46]. The distribution of grain sizes within the AF has been measured from bright field TEM images measuring over 600 grains. The lateral grain sizes and hence the grain volumes followed a lognormal distribution [47]. The median grain volume is given by $V_m=\pi D_m^2 t_{AF}/4$ where t_{AF} is the thickness of the AF layer. From Eq. (2)

$$K_{AF}(<T_B>)=\frac{\ln(tf_0)}{V_m}k_B<T_B> \tag{5}$$

The value of the relaxation time used to calculate K_{AF} was t=1800 s since this was the time for the thermal activation process. Fig. 6 shows the variation of the median blocking temperature $<T_B>$ with the thickness of the AF layer. $<T_B>$ increases with t_{AF} due to an increase in the volume of the AF grains and, therefore, their thermal stability increases as predicted by Xi [15]. We used T_{set}=225 °C but T_{NA} changed from sample to sample depending on the grain volume (i.e. film thickness). Hence T_{NA} was varied for different samples although a single low temperature could have been used as long as it did no cause ordering of interfacial spins as discussed in Section 3.3.

In order to calculate the value of K_{AF} at room temperature, the bulk value of T_N has been used for the temperature dependence of K_{AF}, i.e. T_N=690 K [48], although the value for thin films may be lower. Fig. 7 shows the variation of K_{AF} with the thickness of the AF layer. For samples with $t_{AF}\geq4$ nm $K_{IrMn}(295\text{ K})=(5.5\pm0.5)\times10^6$ ergs/cm^3. For t_{AF}=3 nm, K_{IrMn} decreases slightly to $(4.7\pm0.5)\times10^6$ erg/cm^3. This is most likely due to a lack of crystallisation of the grains.

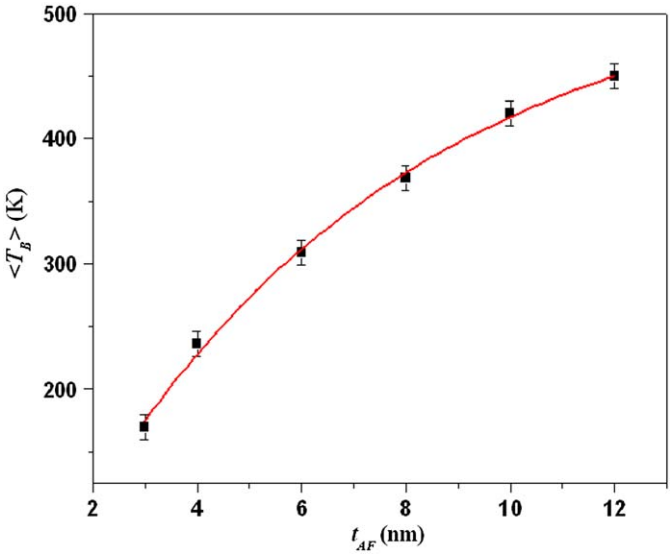

Fig. 6. (colour online) Variation of the median blocking temperature $<T_B>$ with the thickness of the AF layer. The line is a guide to the eye.

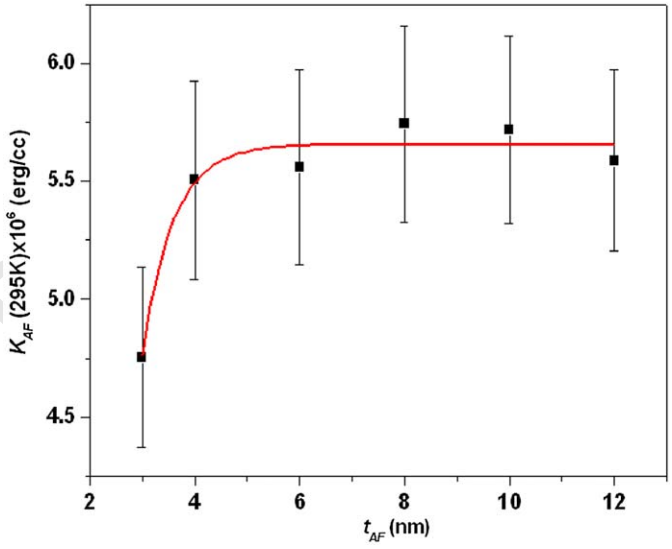

Fig. 7. (colour online) Variation of K_{AF} with t_{AF} at room temperature. The line is a guide to the eye.

The value of K_{AF} we obtain for IrMn is significantly higher than those reported by others [49]. This is to be expected since the sample is completely set before measurement, the effects of disordered grains are removed and the effect of interface coupling negated. The error tolerances are remarkably small due to the large number of grains measured (>600) to obtain D_m and hence V_m.

The sources of systematic inaccuracy lie in the fact that Eq. (2) strictly applies to a system with aligned easy axes. Also the value of f_0 for an AF is unknown. In subsequent work we have shown that improved texture of the IrMn can increase the measured value of K to $(2.0\pm0.5)\times10^7$ ergs/cm^3[50]. Hence all measurements of K_{AF} determined in such systems must be regarded as effective values.

2.4. Setting limitations

IrMn is widely used in most technological applications due its high corrosion resistance, high magnetocrystalline anisotropy and

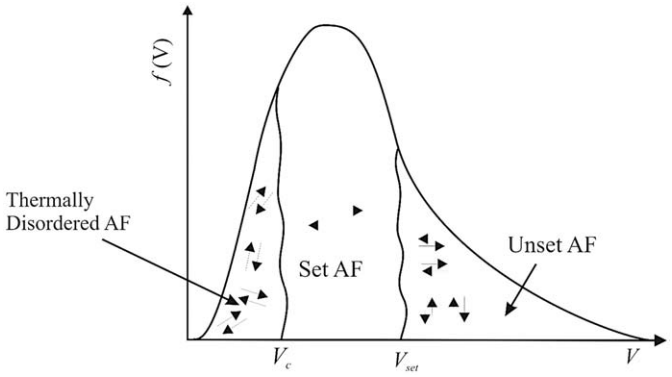

Fig. 8. Schematic of the grain size distribution in the AF layer after setting at a temperature T_{set} in a magnetic field and cooling to a temperature where a fraction of the AF is thermally unstable.

the large loop shifts achievable. Since T_N for this system is much greater than room temperature, it is not possible to field-cool the system from above T_N since this would damage the structure of the sample. Hence, the AF has to be set below T_N, where the setting process is via thermal activation. Fig. 8 shows a schematic of the energy barrier distribution to reversal within an AF layer at $T > T_{NA}$ and after resetting at a temperature T_{set} for a period of time t_{set}. In this figure the conditions applied during the setting stage were not sufficient to set the whole AF distribution. As a consequence, a fraction of the AF grains with $V > V_{set}$ remain unaligned with the F layer. At temperatures above T_{NA} another fraction of the AF grains with $V < V_c$ are not thermally stable at the temperature of measurement T_{ms}. This is analogous to the concept of superparamagnetic particles in the model of Fulcomer and Charap [10]. Hence, only the grains in the window given by V_c and V_{set} will contribute to H_{ex}. Assuming a uniform value of K_{AF} at the temperature of measurement and based on this concept of the two critical volumes we can write

$$H_{ex}(T_{ms}) \propto \int_{V_c(T_{ms})}^{V_{set}(T_{ms})} f(V)dV \tag{6}$$

This simple calculation, whose basis is shown in Fig. 8, can be used to predict the AF grain size and thickness dependence of H_{ex} to high accuracy. Of course, certain aspects of the fabrication process may result in partial setting of grains with $V > V_{set}$. However, again the quality of the fit to the data in Fig. 8 indicated that this is a minor effect.

2.5. Grain volume dependence

We have studied the dependence of H_{ex} on the AF grain volume [51]. This has been done via two separate experiments in which we changed the AF thickness and the lateral grain size. Samples with composition Si/Cu(10 nm)/CoFe(2.5 nm)/IrMn(t_{AF}=3, 4, 6, 8, 10 and 12 nm)/Ta(10 nm) were used. Samples with four different lateral grain *sizes* were prepared for each AF thickness. This control of grain size is easily achieved with our HiTUS sputtering system in which the growth rate, which is controlled via the DC bias, controls the grain size to high resolution. The columnar growth, which has been observed via cross-sectional TEM, ensures that samples of different thickness have almost identical lateral size distributions. Varying the bias voltage allows us to prepare samples of constant thickness and different lateral size. The configuration of the system ensures that the grain size on TEM grids is the same as that in the films [46]. The grain size distribution for each sample was measured from bright field TEM images using a JEOL 2011 electron microscope. Over 600 grains were measured for each sample. All the

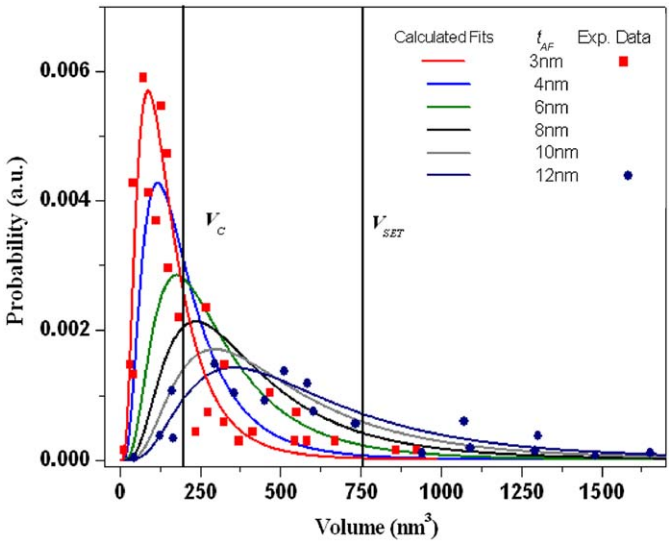

Fig. 9. (colour online) Grain volume distributions for the samples with different AF thickness.

distributions followed a lognormal function. Fig. 9 shows the volume distributions for samples with different AF thickness. Even though the distributions are represented by solid lines they were calculated from the actual experimental data some of which is shown. Note the significant asymmetry of the distribution of grain volumes which is greater than that in D due to the D^3 factor.

Having calculated the value of K_{AF} for our samples, we can now proceed to determine the value of the two critical volumes V_c and V_{set}. Since V_c determines the volume below which an AF grain is thermally unstable and the hysteresis loops were measured at room temperature, we can write

$$V_c(T) = \frac{\ln(100 f_0) k_B 293}{K_{AF}(293)} \tag{7}$$

where the factor 100 arises from the relaxation time τ being equal to the measurement time. Similarly, the second critical volume relating to the limit of the set grains will be given by

$$V_{set}(T) = \frac{\ln(5400 f_0) k_B \times 498}{K_{AF}(498)} \tag{8}$$

where the factor 5400 s is due to τ being equal to the time used to reset the AF, i.e. 90 min. Using the value obtained in the previous section for K_{AF}, V_c=200 nm^3 and V_{set}=757 nm^3. These values correspond to lateral grain sizes of 4.9 and 9.8 nm for a film thickness of 10 nm. The two vertical lines in Fig. 9 represent the two critical limits.

Fig. 10 shows the variation of H_{ex} with the AF lateral grain size for three different AF thicknesses measured at room temperature ($\sim$293 K). Again, prior to measurement the samples were reset at 498 K in a positive field sufficient to saturate the F layer, for 90 min. For low AF thickness (4–6 nm), the exchange field increases with increasing AF grain size. However, for thicker samples, a decrease of H_{ex} with t_{AF} is also observed. For low values of t_{AF}, a large fraction of the AF grains are thermally unstable and, therefore, do not contribute to the loop displacement. By increasing the AF thickness, the number of unstable grains is reduced leading to an increase in H_{ex}. For thicker AF layers, $\sim$12 nm, the AF is mostly thermally stable. However, the measured value of H_{ex} is now limited by the fraction of AF grains that cannot be set. Hence, a decrease of H_{ex} with the AF grain size is observed. These data and the quality of the fits mean that we can now explain the apparently irreconcilable data of Takano et al. [36] and Uyama et al. [37].

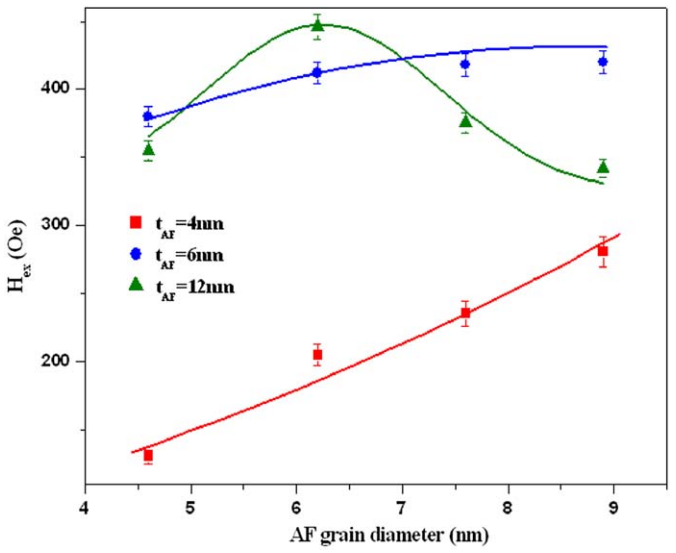

Fig. 10. (colour online) Variation of H_{ex} with the AF grain size for three different AF thicknesses. The solid lines are calculated from Eq. (6) (using Eqs. (7) and (8)) using the measured value of $f(V)$ with K_{AF} determined from Eq. (5).

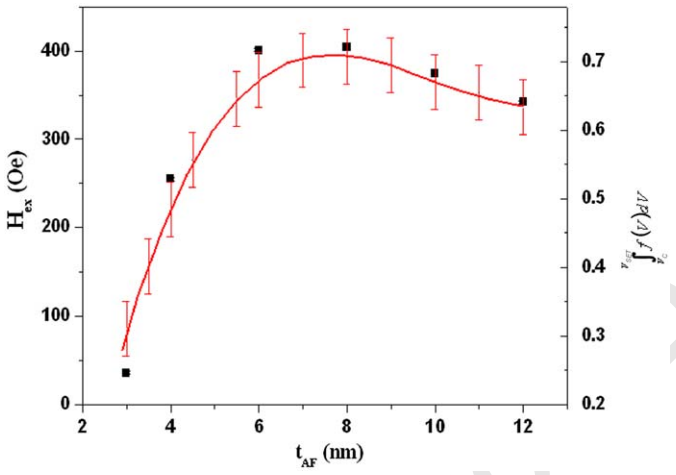

Fig. 11. (colour online) Variation of the exchange field with the thickness of the AF layer. This samples had D_m=7.6 nm and $\sigma_{\ln D}$=0.33. The solid line was calculated from Eq. (6) (using Eqs. (7) and (8)) with K_{AF} measured from Eq. (5) and using the measured values of $f(V)$.

Fig. 11 shows the variation of H_{ex} with the thickness of the AF layer. For this experiment the lateral grain size was D_m=7.6 nm and $\sigma_{\ln D}$=0.33. The measurement procedure was the same as that for the samples with different lateral grain size. $H_{ex}H_{ex}$ increases sharply with increasing AF thickness reaching a maximum at ~8 nm. For thicker samples, a decrease in H_{ex} is observed following approximately a $1/t_{AF}$ variation [34,35]. The slow decrease in H_{ex} for the thicker samples is due to the grain volume distribution being highly skewed for the larger grains. The physical origin of the $1/t_{AF}$ variation can be seen in Fig. 9 where the AF grain volume distribution approximates to 1/V at high values. This is equivalent to $1/t_{AF}$ for constant grain sizes. If the distribution were symmetric, the sharp increase in H_{ex} would be followed by an equally sharp decrease in the exchange bias, which is not observed experimentally as can be seen from Fig. 11. Note that all the lines in Fig. 10 and Fig. 11 correspond to calculations using Eq. (6), i.e. the integral between the two critical volumes V_c and V_{set} of the grain volume distribution. The agreement between this simple theory and the experimental data is remarkable. This suggests that a simple granular model including an accurate measurement of the volume distribution, can account for the grain volume dependence of the exchange bias.

The fit between the experimental values and the lines calculated from Eq. (6) is excellent. Of course the values of H_{ex} have been scaled along the ordinate using $C*$ as a fitting parameter. However, to fit to the form of the data, within error, to the grain volume measurement confirms the validity of our simple theory. Further confirmation of our theory is provided by the calculated lines in Fig. 10. No other theory has ever provided a fit to data for the grain size distribution, particularly considering that the calculated values are based on actual measured grain sizes. Note that the error bars on the theoretical line are derived from the error in the measurement of the grain sizes and the value of K_{AF}. None of the theories based on AF domains can predict the form of the data. This is particularly so when the thickness of the AF layer is 12 nm.

2.6. Magnetic viscosity in the AF

Due to the two critical volumes introduced in Section 2.5, a narrow AF grain size distribution is needed to ensure the setting and thermal stability of the entire distribution. However, samples produced by sputtering present a wide distribution of grain sizes. This distribution of grain volumes leads to a wide distribution of energy barriers $f(\Delta E)$. In ferromagnetic systems, this wide distribution of energy barriers leads to a $\ln(t)$ law in the time dependence of the magnetisation [52]. In the case of AF films, the wide distribution of ΔE gives rise to an $\ln(t_{set})$ law for the degree of order in the AF. The order parameter (P) then controls H_{ex} which follows an $\ln(t)$ law. Therefore, the setting process will have an associated time dependence coefficient $S=(dP/d\ln t_{set})$ that can be written as [53]

$$S(T) \propto 2P_S k_B T f[V_P(T)] \tag{9a}$$

$$V_P(T) = \frac{\ln(t_{set} f_0) k_B T}{K(0)(1 - T/T_N)} \tag{9b}$$

where P_s is the saturation value of the AF order and $f(V_P)$ is the critical value of the energy barrier distribution at the setting temperature that determines V_p. This critical value of the energy barrier will be determined by the grain volume distribution, the temperature dependence of K_{AF} and the setting time.

In order to validate this idea, a sample with composition Si/Ru(5 nm)/IrMn(10 nm)/CoFe(3 nm)/Ta(10 nm) was prepared [54]. The grain size distribution within the AF layer was measured from bright field TEM images. Over 600 grains were measured and the grain size distribution was found to be lognormal with $D_m=(6.0\pm0.4)$ nm and $\sigma=(0.33\pm0.03)$, where D_m is the median grain size and σ is the standard deviation of $\ln(D)$.

Prior to measuring the time dependence of H_{ex}, the original state of the AF has to be known. This is done by heating the F/AF system to a temperature T_{set} in the presence of a negative field $H_{set}(=-1$ kOe) large enough to saturate the F layer. The bilayer was held in this configuration for 5400 s. If T_{set} is large enough, all the AF grains will reverse. In our case, T_{set}=498 K was sufficient to reverse the whole AF distribution. This was confirmed from the full reversal in the value of H_{ex}. This particular system was chosen for this experiment so that the whole AF could be set at moderate temperatures. Further increases in T_{set} or waiting times resulted in very small changes ($<1\%$) in H_{ex} over a period of 6 h.

Once the state of order in the AF was set, the system was heated to the aligning temperatures T_{Al}, at which the AF is reset in the opposite sense to that in which it was originally set. This was

done for different periods of time t_{Al}. This was done in the presence of a positive field large enough to saturate the F layer. By increasing the *aligning* temperature we increase the fraction of the AF that undergoes reversal. This procedure was repeated for a wide range of temperatures ($293\ K \le T_{Al} \le 498\ K$). After the aligning process, the system was cooled to room temperature where the AF is free of thermal activation and hysteresis loops were measured starting with the F layer in negative saturation. This way, training effects were removed.

Fig. 12 shows the time dependence of H_{ex} over the range of temperatures studied as a function of $\ln t_{Al}$. H_{ex} varies linearly with t_{Al} for all values of T_{Al} as indicated by the quality of the linear fits. The gradient $dH_{ex}/d(\ln t_{Al})$ exhibits a peak with T_{Al} at ~413 K. Above 413 K, the gradient decreases. It is important to note that the values of H_{ex} obtained after resetting at 293 and 498 K are the same but of opposite sign. This is a further confirmation that the entire AF energy barrier distribution was fully reversed.

The time dependence coefficient $S(T)$ can now be measured from the slope of the lines in Fig. 12. Fig. 13 shows the variation of $S(T)$ with T_{Al}. Note that the error bars in Fig. 13 are based only on the linearity of the lines in Fig. 12 and do not take into account errors in the grain size distribution nor $K_{AF}(T)$. The coefficient $S(T)$ increases with increasing values of T_{Al} reaching a peak at the measured value of $< T_B >$ to ± 5 K. Fig. 13 also shows the calculated values of $S(T)$ based on Eqs. (9a) and (9b) (solid line). For the calculation a time constant t_{Al}=120 s was used since that was the shortest period of time spent at T_{Al} for each set of data points. The absolute values of the term $f(V_P)$ have been scaled by a constant factor to account for the proportionality sign in Eq. (9a). The fit between the theoretical prediction and the experimental data is again remarkable. Note that the calculated curve deviates slightly from the experimental data points at the edges of the distribution. That is due to the fact that when measuring the grain size distribution most of the grains have a grain size ~D_m. Therefore, fewer grains with diameters at either end of the distribution can be measured leading to a higher uncertainty. The fact that the distribution is symmetric comes from the temperature dependence of K_{AF}.

Again our simple model has been found to describe the observed behaviour accurately. This is the third independent measurement where our theory has been found to fit the data. Again no other model can fit this result.

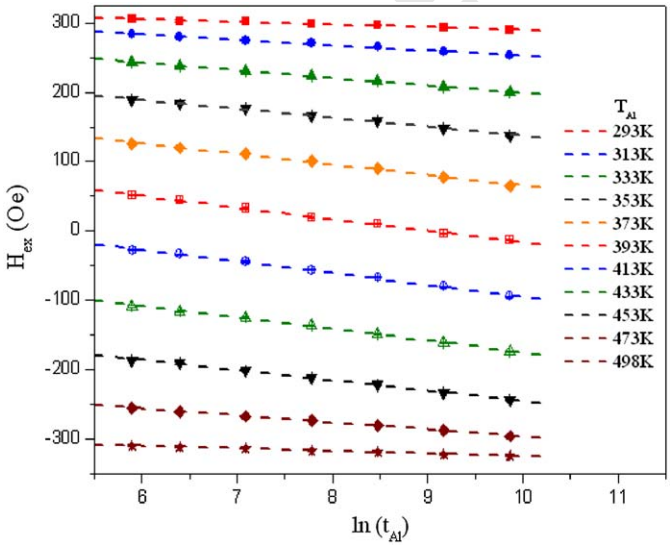

Fig. 12. (colour online) Time dependence of the exchange field with $\ln(t_{set})$.

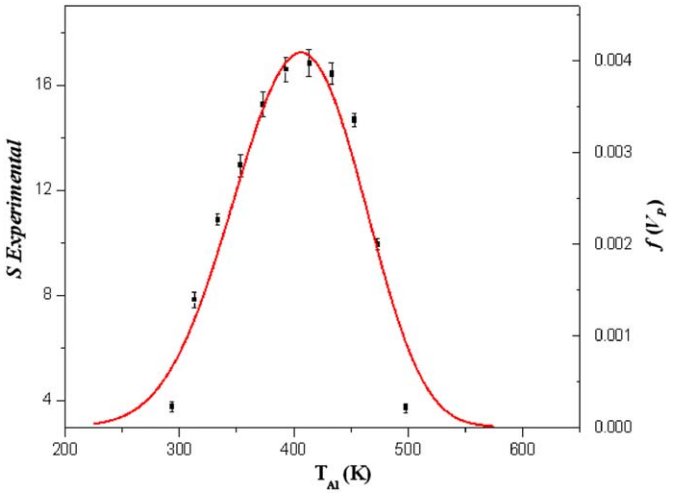

Fig. 13. (colour online) Experimental and calculated time dependence coefficients for a IrMn(10 nm)/CoFe(3 nm) exchange couple.

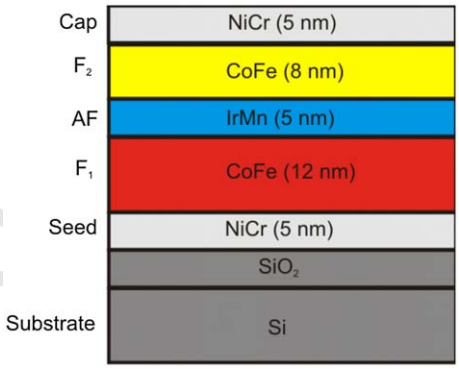

Fig. 14. (colour online) Structure of the trilayer sample studied.

3. Interfacial effects

For many years there has been significant controversy over the role of interfaces in exchange bias [e.g. 55,56]. This controversy concerns whether the interface itself gives rise to the shift in the hysteresis loop. This is obviously the case in core–shell nanoparticles where the thickness of the antiferromagnet is comparable to the typical thickness of an interface itself. There also exists the difficulty of understanding the role of the interface in the coupling between the F and AF layers. Obviously there will be a difference in the coupling between a compensated and uncompensated interface. Also the degree of spin order at the interface is difficult to ascertain. However, we have been able to obtain some information about the behaviour of spins at the interface and their influence on exchange bias via a series of unusual experiments. These experiments relate to studies of trilayer systems, field setting effects and an observation of spontaneous ordering of interface spins at low temperatures.

3.1. Trilayer studies

We have reported on studies of trilayer stysems where a single AF layer is coupled above and below to F layers of different thickness [57]. A schematic of the system studied is shown in Fig. 14. Due to the F layer thickness dependence of exchange bias, such a system gives rise to two hysteresis loops shifted by differing amounts [58]. The initial hysteresis loop for the system is

shown in Fig. 15. Fortuitously because of the thicknesses of the F layers chosen, this system gives rise to a plateau region between the two loops. Note also from Fig. 14 that the thickness of the AF layer is only 5 nm. From the data shown in Fig. 15 it is clear that it is now possible to undertake the thermal activation measurements described in Section 2.1 with either both F layers oriented in the opposite direction to that in which the AF was originally set, or to have one F layer oriented in the opposite direction and the other remaining in the set direction. This is simply achieved by undertaking thermal activation in a reverse field with a value coinciding with the plateau between the two loops.

Fig. 16 shows measurements of thermal activation with both loops oriented in the reverse direction. As expected the thermal activation process re-orients the AF system progressively as the temperature of activation is increased. This results in both hysteresis loops translating from negative field into positive field in a similar manner to that which occurs for a single-layer system when the thermal activation is undertaken with just the single F layer reversed. Fig. 17 shows the effect of thermally activating the antiferromagnet using just the exchange field from the thicker ferromagnetic layer. Thermal activation at moderate temperatures ($<155\ ^{\circ}C$) shifts only the loop for the thicker ferromagnetic layer. In fact detailed studies have shown that the loop for the thinner layer with the larger exchange bias remains unaltered for activation temperatures of 130 °C or lower, to the resolution of our measurement capability, which is about 1 Oe.

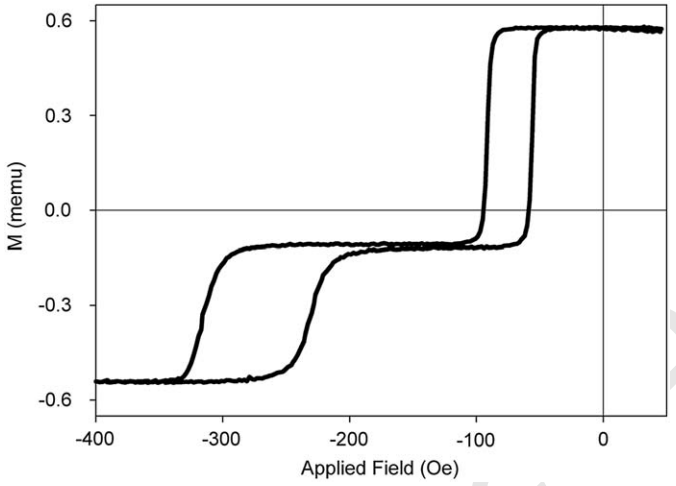

Fig. 15. Initial hysteresis loop for a CoFe(2 nm)/IrMn(5 nm)/CoFe(12 nm) trilayer.

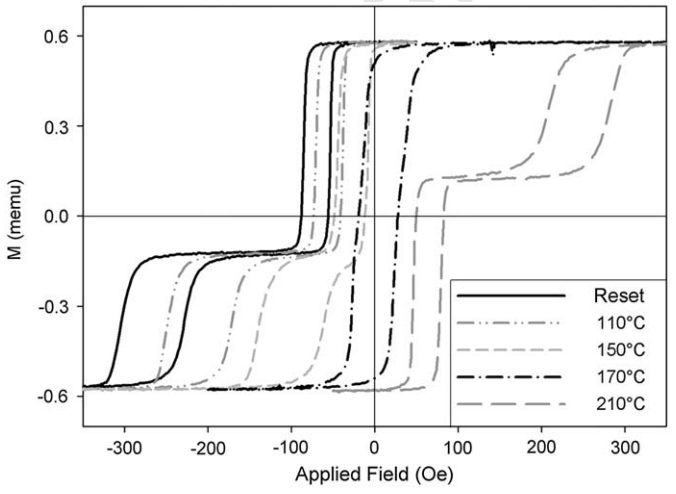

Fig. 16. Thermal activation for a trilayer system with both F layers at negative saturation for a variery of temperatures.

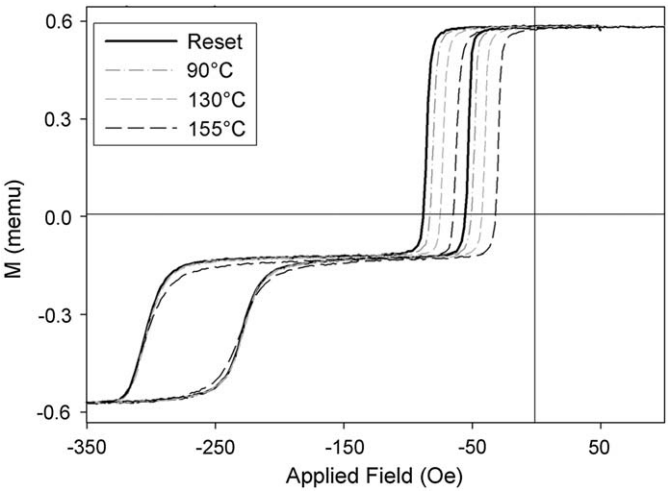

Fig. 17. Thermal activation measurements for the trilayer system at −150 Oe (i.e.only one F layer reversed) for a variety of temperatures.

Given the thickness of the AF layer (5 nm) there is only one possible explanation for this observation in that the material whose structure was changed is that found at the interface between the thicker F layer and the AF layer. Obviously both the interface with the thinner layer and the bulk of the material must be entirely unaffected by the thermal activation process when only the thicker F layer is reversed.

These results imply that there is some degree of order in the interface spins that is established when the AF is originally set. The degree of order in the interface spins then determines the strength of the coupling between the AF and F layers. Surprisingly the data in Fig. 17 shows that an exchange field from an adjacent F layer together with sufficient thermal energy can reorder the interface spins without affecting the bulk of the AF.

Interestingly the fact that the loop for the thicker layer shifts further with increasing temperatures indicates that the order in the interface spins does not have a single-order parameter. This implies that there is a distribution of activation energies for the reordering of these spins. This distribution is clearly different to that of the distribution of energy barriers to reversal in the bulk of the AF grains. Further evidence for this view is presented in Section 3.2 below. Given that we believe the degree of order in the interface determines the coupling between the AF and F layers, it will give rise to a distribution of coupling energies.

3.2. Field dependence of interfacial order

It has been known in the magnetic recording industry for many years that a large field is required to set the AF layer in a spin valve or a spin tunnel junction head. This leads to something of a conundrum since the F layers are typically a CoFe alloy which saturates readily in a few hundred Oe and is certainly fully aligned in a field of 1 kOe. The usual understanding is that it is the exchange field from the F layer that aligns the AF grains. Hence the application of a field greater than that required to saturate the F layer would be of no benefit. In recent years almost all companies manufacturing spin valve or spin tunnel junction read heads have begun to use superconducting magnets, offering fields of 2 T or greater, for the setting process. The exact motivation for

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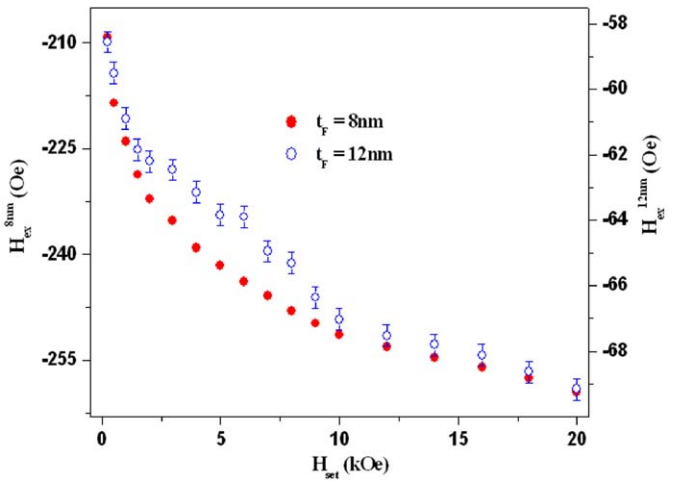

Fig. 18. (colour online) Variation of H_{ex} with H_{set} for the two CoFe layers in the trilayer system. Note the different axes for the two layers.

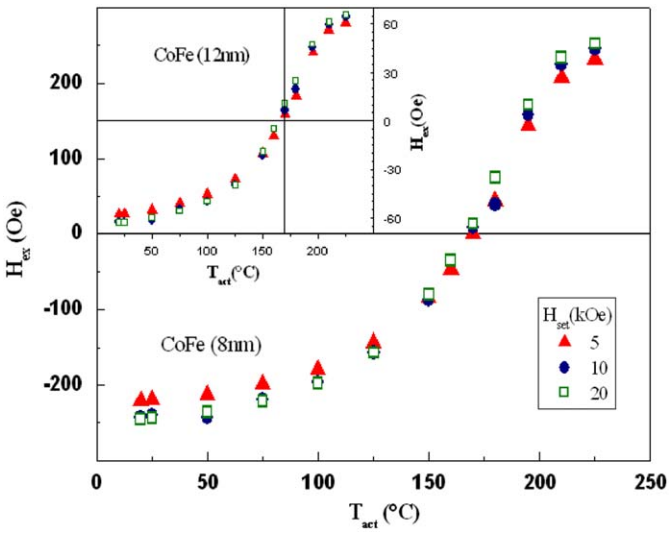

Fig. 19. (colour online) Thermal activation at negative bias with different setting fields for both CoFe layers.

doing this has never been published but it is known to lead to a significant improvement in H_{ex}[59].

In order to try and establish the origin of this improvement we have undertaken studies of the value of the exchange bias achieved as a function of the setting field (H_{set}). The field limitation in our vibrating sample magnetometer meant that measurements could only be made from low fields up to 2 T. Fig. 18 shows the variation of the exchange bias in the same trilayer system used for the experiments described in Section 3.1. Hence we have two separate CoFe layers that can be considered. Note the two separate axes for the two layers. For this system an increase in the exchange bias of approximately 20% is observed for both layers when the setting field (H_{set}) is increased from 1 to 20 kOe. Interestingly the exact form of the variation for the two layers is not identical. This is a further indication that the applied field is changing the interfaces and that the bulk is unaffected. Also when considering Fig. 16 in conjunction with Fig. 18 it is clear that both F layers are fully saturated by 350 Oe and hence the exchange field from the F layer is maximised. Subsequent changes in the exchange bias beyond this point are therefore not due to changes in the bulk AF and have to be due to changes in the interface spin order.

This suggestion has been confirmed by undertaking measurements of the distribution of blocking temperatures in the same manner as that described in Section 2.2 having set the AF layers in different fields. Fig. 19 shows the variation of blocking temperatures for the trilayer sample after setting in fields of 1, 10 and 20 kOe. The distribution of blocking temperature is identical in each case to within the resolution of the measurement which is ± 2 K. Hence there has been no alteration in the order of the bulk of the AF and this confirms that the setting field effect is entirely due to interfacial spins and the effect of their order on the coupling across the F/AF interface.

This result appears to contradict the findings of Fitzsimmons et al. [60], who found a variation in the net AF moment in an FeF_2 single crystal with cooling field. We cannot account for this discrepancy other than to remark that the exact superposition of the $H_{ex}(T)$ curves for different cooling fields indicates that any change in the bulk of the AF grains is tiny and is unlikely to account for the increase in H_{ex} which is of the order of 25%.

In addition to the data presented here we have shown that field alignment of the interfacial spin order and the consequent coupling across the interface occurs in many different systems [61]. However, whilst the magnitude of the effect changes the value of H_{ex} by typically 20–30%, there is significant variation in the scale of the effect. There appears to be some correlation with the texture of the AF and some influence of the composition of the F layer at the interface. At this time insufficient data are available for us to be able to define exactly the origin of the effect. One factor that is now clear is that we are able to write down a formula which links the exchange bias not only as being proportional to the degree of order and stability in the AF layer, but also to the behaviour of the interfacial coupling.

Under ideal conditions there would be an intrinsic value of H_{ex} denoted H_{ex}^{i}, but this value of the exchange bias is moderated by the fraction of the bulk of the AF that contributes to H_{ex} and also by the strength of the interfacial coupling C^*. We have now shown that C^* is a thermally activated factor which also depends upon the setting field used and hence should be written as $C^*(H_{set}, T_{set})$. Hence the value of H_{ex} is

$$H_{ex}(H_{set}, T_{set}) = H_{ex}^{i} C^*(H_{set}, T_{set}) \int_{V_c(T_{meas})}^{V_{set}(T_{set})} f(V)dV \quad (10)$$

Note that in Eq. (10) we replace the proportionality factor in Eq. 4. A further consequence of the data shown in Fig. 19 is that this data again confirms that the interface spins behave largely independently of the AF grains and can be viewed almost as a separate layer in the system.

3.3. Temperature dependence of interfacial spin order

The nature of the spin structure at the interface remains to some extent an open question. Clearly from the field dependence of the exchange bias it is obvious that the spins at the interface are not independent. If they were then fields of the order 2 T would have little influence. These spins experience the exchange field from the F and the AF layers and hence some degree of order is to be expected. Spin–spin interactions will also occur. Hence it might be expected that these spins would exist in clusters or behave cooperatively in a manner similar to, but not necessarily identical to, a spin glass.

In an attempt to compare the blocking temperatures derived from the York Protocol and those from a standard measurement of the blocking temperature distribution at elevated temperatures, we have undertaken a study of both IrMn/CoFe and FeMn/NiFe [62]. These data are shown in Figs. 20 and 21, respectively. As can be seen, both measurements of the blocking temperature

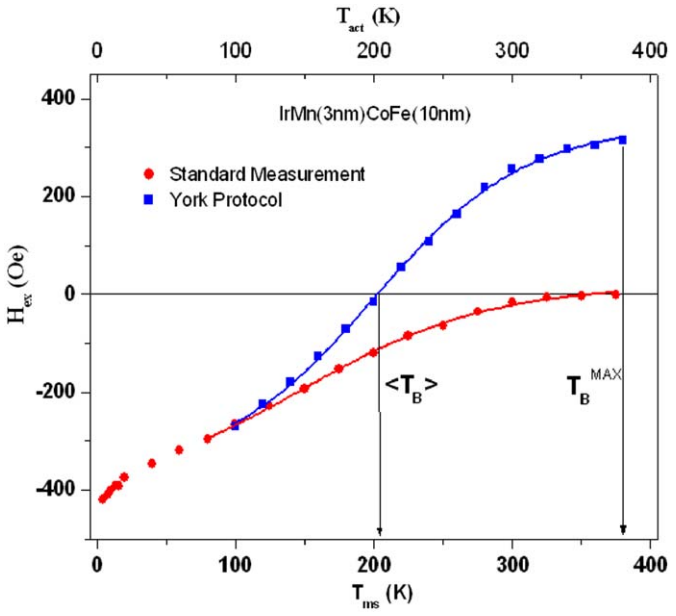

Fig. 20. (colour online) Exchange bias field measured following the York Protocol and measured via standard procedure for IrMn(3 nm)/CoFe. Solid lines are a guide to the eye.

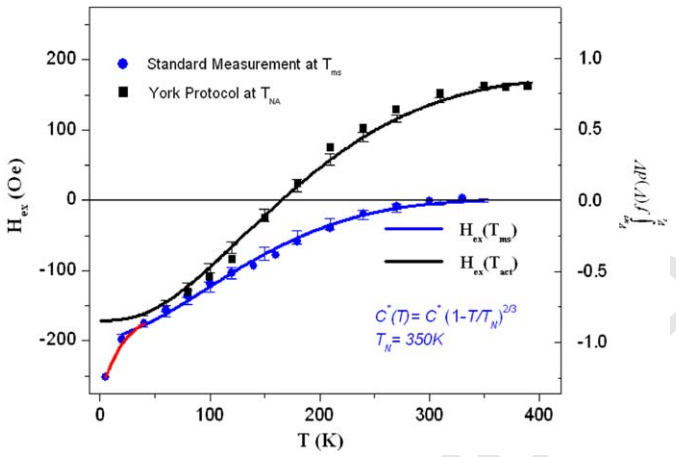

Fig. 21. (colour online) Exchange bias field measured following the York Protocol and measured via standard procedure for FeMn(10 nm)/NiFe. Solid lines were calculated from Eq. (6) using Eqs. (7) and (8).

distribution provide similar results although obviously the measurements made at elevated temperatures produce a lower value of the maximum blocking temperature. However, at low temperatures it is seen that the conventional measurement suddenly produces a remarkable increase in the measured value of H_{ex} well below the value of the minimum blocking temperature T_B^{min}. For the York Protocol measurements we establish the temperature T_{NA} so that the AF is completely stable and hence the only origin of this further increase in H_{ex} must lie in the interfaces. We have interpreted this effect as being due to an increase in the ordering of the spins at the interface which then increases the degree of coupling across the interface between the F and AF layers. Fig. 21 shows that this effect is more pronounced in FeMn almost doubling the value of H_{ex}. Hence the parameter C^* discussed above will also have a marked temperature dependence due to spontaneous ordering of the disordered spins at the interface. Similar effects were observed by Takano et al. [36].

From the form of the low-temperature data in Figs. 20 and 21 the spontaneous spin freezing at low temperatures does not occur at a single temperature. Hence it is not a phase transition similar to a Curie temperature in a ferromagnet but is more similar to a blocking temperature distribution in a spin glass. This is not surprising since due to interface roughness, the exchange field experienced by each cluster will vary. Due to the distribution of grain size it might be expected that the cluster size will also vary. This in turn will lead to a variation in cluster–cluster exchange and dipole–dipole interactions.

3.4. Field dependence of interfacial spin ordering

Given the experimental data described above it is now possible for us to speculate on the nature of the interfacial spin ordering. In a separate study of films with structure Ta(5 nm)/Seed/IrMn(t_{AF})/CoFe(2 nm)/Ru(5 nm) we have again examined the variation of H_{ex} with H_{set} the setting field, where the seed is NiFe, Cr or Ru. Since the value of H_{ex} increases with H_{set} this implies that the order of the interface spins is ferromagnetic in character. This suggestion is supported by the spontaneous ordering that occurs at low temperatures. We postulate that this ordering should follow some type of Langevin function ($L(x)$), assuming that the coupling between the F and AF layers depends linearly on the degree or order of the interfacial spins. Hence we would expect a variation of the form

$$H^{ex}(H_{set}, T_{set}) \propto H_{ex}^{i} L(x) \quad \text{with } x = \frac{N\mu_B(H_{set}+H^*)}{k_B T_{set}} \tag{11}$$

where H^* is now the exchange field which the spins experience due to both the F and AF layers and spin–spin interactions, μ_B is the Bohr magneton and N is the number of spins in each cluster. Unfortunately this form of Langevin function contains two unknowns, H^* and N. It is now impossible to obtain a unique fit to the data. However it is possible to put boundaries on the number of spins that must be ordering to give rise to the increase in H_{ex}. For example if the extreme value $N=1$ is taken, i.e. assuming that the spins act singly, then the resulting value for the exchange field would have to be of the order of 2×10^5 kOe which is physically unrealistic. Typical exchange fields for ferromagnetic materials derived from molecular field theory are of the order of 10^4 kOe. Given that this is an interface between an F and an AF layer the value for bulk iron quoted above would also seem to be inappropriate. However, if a value of half of the exchange field for iron is used then we are able to fit our experimental data allowing the parameter N to float. Using the value $H^*=0.5 \times 10^4$ kOe we obtain a good fit to a range of data giving a value of N in the range 10–50. This result is entirely consistent with our observation of the spontaneous freezing of the interface spins which occurs at temperatures consistent with spin cluster behaviour. This value for N would also be indicative of spin clusters that would be associated with each grain. However, it is clear from the form of variation of H_{ex} with H_{set} that the ordering of the spins within the cluster is ferromagnetic in character. This must be the case otherwise the coupling would be expected to decrease with an increase in the setting field. Of course it is perfectly possible that cluster–cluster interactions can also occur. To resolve this possibility large scale atomistic computer simulations of the interface will be required.

Hence the conclusion of this study of the bulk and interfaces and the mechanisms by which they behave quasi-independently, is that a structure similar to that shown schematically in Fig. 22 must exist. The control of the parameters within this structure is the challenge for materials physicists wishing to design structures for specific devices. Clearly if factors are available which control the behaviour of the interface spin clusters then it should be

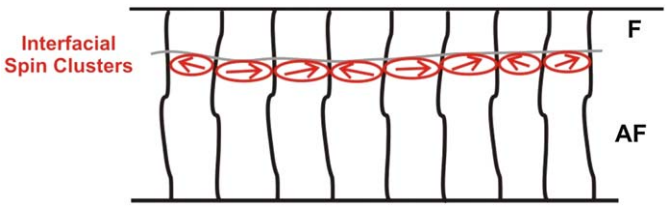

Fig. 22. (colour online) Spin clusters at the F/AF interface.

possible to design AFs specifically for alignment under given setting conditions.

3.5. Origin of the coercivity

One of the outstanding issues associated with exchange bias in polycrystalline metallic thin films is the origin of the coercivity of the F layer. The coercivity itself derives from the F layer where the grains are strongly exchanged coupled together and the reversal mechanism is via domain wall motion. This is in contrast to the AF layer where the individual AF grains are not exchange coupled as shown by the excellent fit between the measured grain size distribution and the various parameters of the blocking process in Section 2. The presence of the interface spin clusters as described above merely adds a further degree of complexity to the possible origins of the coercivity.

What is clear is that the measured coercivity of the F layer does not appear to correlate in any systematic way with the degree or order in the AF. There are changes in the coercivity at low temperature where all the AF grains are thermally stable but these do not appear to correlate with the orientation direction of the AF grains. However, it must be that the additional anisotropy giving rise to the enhanced coercivity has its origins within the bulk of the AF layer. This increase in anisotropy must in some way be transmitted via the interfacial spin clusters to the ferromagnet to give rise to the coercivity. Hence the enhanced coercivity is almost certainly a combination of the effects in both the bulk and interface spin clusters within the material.

Fig. 23 shows the variation of both coercive fields with the setting field for the 8 nm F layer of the trilayer system discussed previously in Section 3.1, where H_{c1} and H_{c2} refer to the coercive fields on the demagnetising and magnetising branches of the loop, respectively, after the removal of training effects. Clearly having verified that the setting field is affecting only the interface spins, the observation of a variation in the coercive fields of a similar form to that observed for the exchange bias indicates that the origin of the coercive *fields* is dominated by interface cluster ordering. Hence the value of H_{ex} is controlled by the order in the AF mediated by the interfacial spin order. However, this order determines H_c providing only an indirect connection between the two parameters.

The mechanism by which the interfacial spin clusters control the coercivity is as yet unclear. The variation of H_{ex} with the setting field is clearly of Langevin form and would therefore imply that these clusters do not have any intrinsic anisotropy due to magnetocrystalline effects. However, weak shape anisotropy could not be precluded. Also these interfacial spin clusters must acquire some level of anisotropy from the F or AF layers from which they derive. They may also have an effective anisotropy simply via the spin–spin interactions within or between the clusters. This behaviour would be analogous to that which is known to exist in spin glasses. The origin of the coercivity would then lie in the ability of the clusters both to rotate with the F layer when a field is applied and thereafter to see a lowering of energy by remaining in that orientation. This would give rise to a resistance for the cluster, and hence the F layer, to reverse when

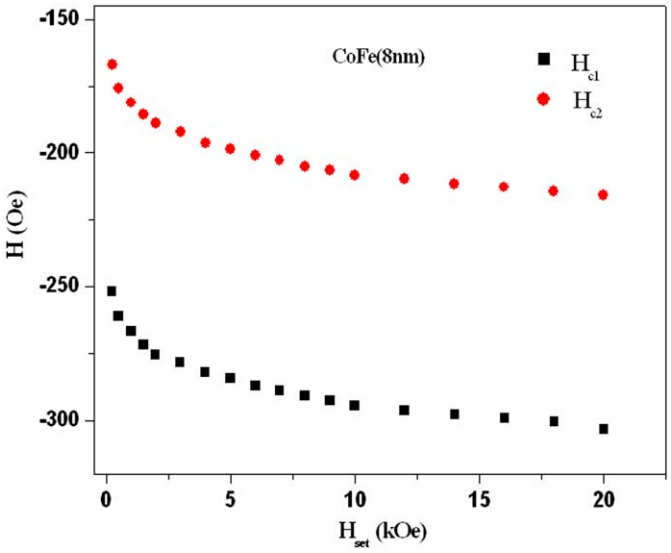

Fig. 23. (colour online) Variation of the coercive fields for the thinner CoFe layer with setting field in the trilayer system described in text.

the field is reversed. It is the depth of the potential well into which the clusters sink which then controls the value of the coercivity. The exact origin of this potential well is at present unknown. It could reside simply within an increase in order within the spin cluster or even a change in spin cluster shape. Of course cluster–cluster interactions could also produce low-energy configurations as is the case in spin glasses, which again could give rise to a coercivity. Further extensive studies of these interfacial spin effects are ongoing at this time.

4. The questions of Berkowitz and Takano

One objective of this review is to attempt to answer the five questions posed by Berkowitz and Takano in 1999 [5].

(1) "What structural and magnetic parameters are responsible for the drastic reduction of the interfacial exchange energy density from the ideal case?"
Two factors reduce the value of H_{ex} from the ideal case. The first is the lack of complete order in the AF due to setting limitations and thermal disorder. The second is the level of disorder in the spin clusters at the interface.

(2) "What are the origin and role of the interfacial uncompensated AF spins?"
We cannot be definitive about the origin of interfacial uncompensated spins, but we believe that the majority come from the AF grains. As we have shown they exist in spin clusters and serve to transmit the anisotropy from the AF to the F layer. They exhibit complex variations in order with field and temperature and are able to alter their state of order without affecting the bulk of the AF.

(3) "How is the magnitude of the exchange field dependent upon the AF grain structure?"
The magnitude of H_{ex} depends on the AF grain structure insofar as it affects the value of K_{AF}. This implies that the degree of crystallinity [63], the phase [64] and the texture [50] can all impact K_{AF}. The second factor is the grain volume and critically its distribution which give rise to $f(\Delta E)$ where $\Delta E = K_{AF}V$.

(4) "What determines the temperature dependence of the exchange field?"Again two factors control the temperature

dependence of $H_{ex.}$ The first is the variation of K_{AF} with temperature. This in turn determines the value of V_c and V_{set} and therefore the value of the integral which describes the fraction of the AF that contributes to H_{ex}.

The second factor controlling $H_{ex}(T)$ is the effect of temperature on the degree of order in the interfacial spins. From the data in Figs. 20 and 21 we believe that this degree of order is relatively constant until the spin glass like freezing commences below 77 K. Similarly the order is unaffected until temperatures well above $<T_B>$ [65] indicating the lack of connection to the bulk of the AF grains.

(5) "What are the roles of interfacial exchange J_{ex} and AF magnetocrystalline anisotropy K_{AF} in unidirectional anisotropy?"

It is perhaps an error to think of the interfacial exchange in terms of a single parameter J_{ex}. Our work shows that the behaviour of the interfacial spins is much more complex due to the presence of clusters in which spins behave cooperatively and which also allow for cluster–cluster effects.

The issue of the AF magnetocrystalline anisotropy has been addressed in the answer to question 4 above. The key parameter here is that K_{AF} is magnetocrystalline and is therefore temperature dependent.

We believe that our new paradigm can explain most known features of exchange bias in polycrystalline systems. It must again be emphasised here that our paradigm applies only to sputtered, granular systems with a grain size distribution with diameters ca. 10 nm. It will not apply to epitaxial single-crystal systems, large grain sputtered systems and core–shell nanoparticles. However, the York Protocol may enable improved data for these systems to be obtained.

We believe that above we answer almost all the questions of Berkowitz and Takano. The sole outstanding part of question 2 is the origin of the interface spins which our studies cannot elucidate. There are outstanding issues still to be resolved concerning factors such as the coercivity and the first loop training effect. These topics are under investigation at the time of writing.

5. Further studies and technological applications

In addition to work reviewed in this article which is focussed on the key questions asked by Berkowitz and Takano [5], we have undertaken a number of additional studies using the York Protocols. These works have mainly been in collaboration with industrial laboratories who now routinely use our model of exchange bias in polycrystalline films.

For example we have shown the correlation between AF crystalline texture and $<T_B>$ [50]. Here the texture enhances K_{AF} to values of the order of 2×10^7 ergs/cm^3 giving rise to systems with $<T_B>$ =477 K for a grain size of only 4 nm. Hence the use of appropriate seed layers has become critical to achieve the correct anisotropy in technological applications. Our work in this area has allowed thinner AF layers to be used in read heads with values now below t_{AF}=7 nm giving good thermal stability. Similarly our insight into the setting process and particularly its dependence on time and temperature has enabled systems to be developed where, with optimisation of the AF grain size distribution, almost the entirety of the AF grains contribute to H_{ex} albeit for a system with a very thin (2 nm) F layer, we have reported a world record for H_{ex} in polycrystalline systems of 3.6 kOe at room temperature in collaboration with an industrial laboratory [61].

Our understanding of the nature of exchange bias in polycrystalline films also allows us to speculate on the possibility of the development of an optimised system. The specification for such a system would include a high value of H_{ex} which would require a contribution from all the AF grains being set. It would also require an interface with limited disordered spin clusters or clusters that are somehow rigidly aligned. There would then be a need for a high degree of thermal stability to give a high value of $<T_B>$. This would depend on the anisotropy and the grain volumes. Such a system could not be set by thermal activation.

Hence the ideal system would be one with a relatively low T_N so that the AF grains could be fully set. A well-textured system with high K_{AF} and large grains ca. 12 nm, would then give a value of $<T_B>$ close to T_N. Modification of the interfaces, for example by doping [66], may well reduce the need for high field setting. However, for a well-designed system the requirement for high fields is easily achieved.

6. Conclusions

In this work we have summarised an extensive series of experiments undertaken in York over the last few years. From these experiments we have shown conclusively that the value of the exchange bias in a metallic polycrystalline exchange bias system is controlled by the proportion of the AF layer that is thermally stable and set. A remarkable agreement between the integral across measured grain volume distributions and the variation of the exchange bias with both film thickness and grain size has been achieved. No other existing model of exchange bias systems is capable of achieving a correlation between theory and experiment of this quality over such a wide range of observations. The consequence of this remarkable agreement is that the AF grains are essentially independent of one another and reverse their orientation via a reversal mechanism analogous to that observed in Stoner–Wohlfarth systems. This has been confirmed by the fit to the film thickness, fit to the grain size and the fit to the logarithmic setting rate of the exchange bias.

We have also shown that there are interfacial effects that occur in exchange bias systems that are completely independent of the degree of order in the AF layer. These interface effects have been shown unambiguously by studies of trilayers where only one layer is affected by an engineered change in the interface. Studies of the setting field dependence of exchange bias have shown that this effect is due to interface spins and that the degree of order in the AF is completely unaffected by the setting field. The fact that such effects follow a near Langevin function leads to the conclusion that the interface spins exist in clusters containing between 10 and 50 spins. It is the behaviour of these spin clusters which is analogous to that of a spin glass that transmits the anisotropy from the AF layer to the F layer. Furthermore it is the complex behaviour of these clusters allowing for inter and intra cluster interactions as well as interactions via the exchange coupling to the AF and F layers that gives rise to the coercivity of the F layer in exchange bias systems.

This new paradigm for exchange bias in polycrystalline films has already and will continue to allow for the design of antiferromagnets for specific applications and also the design of ferromagnets which are tailored to specific setting conditions. Such advances are critical for future developments in read head technology and MRAM devices.

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