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Invited Paper

Thermal instability in exchange biased materials

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In the recent past it has become clear that one reason for the lack of understanding of the exchange bias phenomenon is the fact that the antiferromagnet is not stable against thermally activated changes. Therefore extreme care is required when measuring these effects. In this paper possible origins of thermal instabilities will be discussed and data shown from measurements undertaken using carefully defined measurement protocols which enable reproducible and therefore interpretable data to be obtained. One interesting feature of the study of these materials is that the actual state of the antiferromagnet cannot be measured directly and must be inferred from the behaviour of the ferromagnet after careful sequence of field and temperature cycling. We report on a study of the thermal stability of exchange bias systems, IrMn/CoFe and FeMn/NiFe, with different thicknesses and grain sizes. The influence of the grain size has been studied and shows that coercivity and exchange bias have different origins. Training effects studied over a range of temperatures from 4.2K to 400K indicate different contributions to thermal instabilities in the exchange biased system from the magnetic state of the antiferromagnet.

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1. Introduction

The exchange coupling at the interface between a ferromagnet, F, and an antiferromagnet, AF, induces a unidirectional anisotropy in the F which has been the subject of study since it was first discovered by Meiklejohn and Bean [1]. This exchange anisotropy gives rise to a shift in the hysteresis loop, induced after field-cooling from above the AF Néel temperature, T_N [2, 3]. The shift of the centre of the loop from the origin is the exchange field, H_{ex}. This shift derives from the amount of AF material that exists in a state such that its orientation remains unaltered when the F is reversed [4], and is accompanied by an enhancement of the coercivity, H_C, defined as the half-width of the loop [5]. These quantities are known to be sensitive to the effects of crystal texture, granularity, and the physical and magnetic microstructure of the AF/F interface [6]. The origin of the coercivity is not understood [e.g.7]. An anomalous magnetic viscosity in the F layer has been shown to be a consequence of thermally activated changes in the AF state during the time of measurement [8].

Much of the published literature is not specific with regard to the exact experimental conditions used to obtain the measurements. Also, it is often not appreciated that the state of the AF material can change by maintaining the system with the F layer saturated in the opposite direction to that in which it was originally set (reverse bias). Also, changes in its state of order may occur during a normal measurement of the properties of the ferromagnet. To obtain reproducible data for exchange biased systems it is necessary to reset the state of the antiferromagnet prior to the measurement of each loop. This is rarely done and much data published in the literature is therefore confusing and difficult to interpret. The amount of AF material that undergoes reversal depends on the temperature of measurement as reversal of AF domains or grains occurs over an energy barrier distribution. The study of the energy barrier distribution to reversal in the AF shows that the AF can be reset without the need to exceed the Néel temperature [7]. Also for each system, a temperature, T_{NA}, can be found at which thermal activation ceases. Evidence for this is provided by the fact that the value of H_{ex} decreases with increasing time spent with the F in negative saturation at temperatures above T_{NA}. However below T_{NA} the value of H_{ex} is stable even after extended periods at reverse bias and is reproducible over many field cycles following the measurement of an initial loop where invariably a training effect occurs. In this way reproducible data are obtained.

2. Experimental

Both, thermal activation and training effects are dependent on the AF grain size among other parameters. There have been several attempts to characterize the effect of the AF grain size on exchange bias [10]. In most of the studies available in the literature, the AF grain size is varied by changing either the seed layer or its thickness, i.e. [11]. Also, annealing at different temperatures has been used to promote grain growth [12]. In this work we have controlled the grain size of FeMn films using a novel sputtering technology known as HiTUS [13]. In this system, the plasma is generated by a RF antenna in a side arm remote from the sputtering chamber. A bias voltage is required underneath the target to sputter material although it is not necessary to sustain the plasma. This is due to the fact that above -100V bias, the target current saturates and the deposition rates can be varied maintaining high-density plasma. It has been shown that grain size of HiTUS sputtered films is controlled via the growth rate [14]. This is achieved by varying three parameters: bias voltage, RF power and/or process pressure. In this work, we varied the bias voltage between -200 and -1000V leading to changes in the grain size.

Two sets of samples were measured. The first contained IrMn(t_{AF})CoFe(10nm), with t_{AF} = 3 and 5 nm. The preparation of these samples has been described previously [15]. Two other sets of samples having the same structure Si/Cu/$Ni_{80}Fe_{20}$ (10nm)/ $Fe_{50}Mn_{50}$ (t_{AF} nm)/Ta, t_{AF} = 7, 10nm, and labeled t-7 and t-10 were grown using high target utilization sputtering (HiTUS). This system has been shown to achieve reproducible control of grain size and size distribution via control of the sputtering rate [14, 16]. Measurements were made using a vibrating sample magnetometer, VSM, with a noise base of 5×10^{-6}emu. Fields were stable to better than 0.5Oe/h. Fields were swept at 50 Oe/min. Fluctuations in temperature were less than ± 0.2 K.

In order to obtain reproducible data it is required that the state of the AF prior to any measurement is known. This is achieved by resetting the AF by means of thermal activation. Accordingly, it is necessary to apply a positive saturating field to the F (point 1 in Fig. 1) and heating at a temperature high enough to overcome those energy barriers to reversal that during previous measurements were susceptible of thermal activation. This implies that it is not necessary to heat above T_N to reset the AF. We established that heating at 373K for 90 minutes while having the F saturated in positive bias ensures that the state in the AF is reproducible for both IrMn samples. This procedure resulted in superposition of hysteresis loops measured at the same temperature. It was established by experiment that this procedure would result in the maximum shift in the hysteresis loop once the sample was cooled to a thermal activation free temperature, T_{NA}. Figure 1 shows different resetting conditions for IrMn(3nm)/CoFe(10nm) when the measurements are done at T_{NA}. In the case shown, 383K for 22.5 minutes reorders the AF when the sample is measured at 77K. Application of the resetting conditions produces the same original loop as previously measured and removes the shifts produced in each branches of the hysteresis loop due to training effects, discussed in section 5.

3. Intrinsic behaviour of an exchange biased system

It is necessary to establish a temperature at which the hysteresis loop can be measured without thermal activation occurring during the time of measurement. This was undertaken by resetting the AF and immediately cooling the sample to a lower temperature where holding the F in negative saturation for different times does not result in further changes in the AF and therefore the superposition of subsequently measured hysteresis loops is achieved. This occurs at a temperature where the amount of thermal activation occurs while the F is kept in negative bias is negligible. For the IrMn(t_{AF})/ CoFe(10nm), the values of T_{NA} were 100K and 77K for t_{AF} = 5 and 3 nm respectively, while for NiFe(10nm)FeMn(10nm) T_{NA} it is 60K. Figure 2 shows the near superposition of hysteresis loops obtained after exposure to reverse-bias fields for 1 and 15min. At present we are unable to account for the small change observed in H_{c2}. However we are sure that the AF is thermally stable at least for short times as the value of H_{c1} is unaltered. Measuring from the reverse bias condition after reordering in forward bias allows for the removal of athermal training effects on H_{c1}. H_{c2} may still be subject of athermal training effects as discussed in section 5.

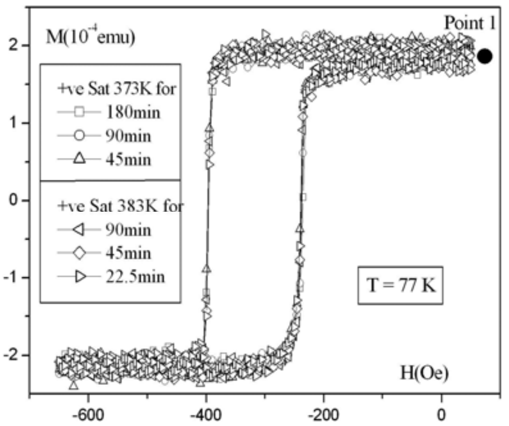

Fig. 1. Resetting conditions for IrMn(3nm)/CoFe(10 nm).

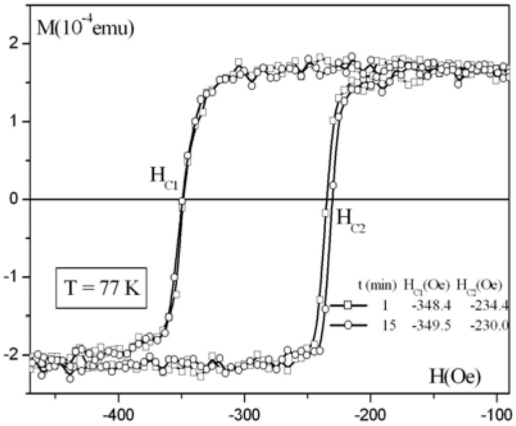

Fig. 2. Effect of reverse bias for 1 and 15 min, for IrMn(3nm)/CoFe(10 nm).

4. Energy barriers and blocking temperature

The distribution of energy barriers to reversal can be studied by reversing the order of the AF grains or domains at different temperatures of activation, T_{Act}, for a fixed amount of time with the F saturated in reverse bias. The measurement of the new state is undertaken at T_{NA}. A resetting stage was applied to obtain the same magnetic state prior to each measurement with the conditions as specified previously, 373 K, 90 min and 2 kOe applied to the F. Subsequently, the temperature was lowered to 77 K, i.e. the thermal-activation-free temperature. At this temperature the applied field is reversed to saturate the F in the negative direction. The temperature is then raised to T_{Act}, temperature at which thermal activation takes place for a fixed amount of time, in our experiment t_{Act}=30 minutes. It is expected that the change in the order of the AF will vary as *ln*(t) [17]. Hence after 30 minutes further changes would be negligible [7]. The temperature is then lowered to 77K and the hysteresis loop is measured. Thermal activation in negative saturation progressively changes the order in the AF from the original state to reverse orientation. The amount of AF material that experiences a change in its state will be a function of the time, temperature and the exchange field exerted by the F layer as shown in Fig. 3.

From Fig. 3 it can be seen that a distribution of energy barriers to reversal exists in both IrMn/CoFe samples. The results obtained are plotted in Fig. 4 showing the variation of the exchange bias and coercivity with T_{Act}. All measurements were made at T_{NA} for each sample. It is important to notice that the measurements of these hysteresis loops, were made starting from negative bias. This way the variation of H_{ex} can be interpreted in terms of thermal activation during the time spent at negative saturation. This procedure avoids any contribution to the H_{ex} due to spin reorientation phenomena occurred in the AF during the first reversal of the F [18] (see section 5).

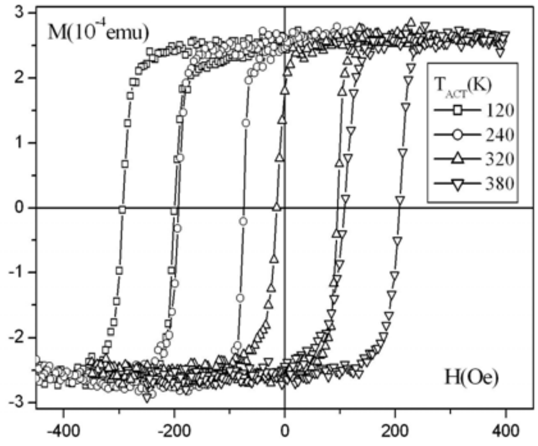

Fig. 3. Effect of thermal activation on IrMn(5nm)/CoFe(10 nm).

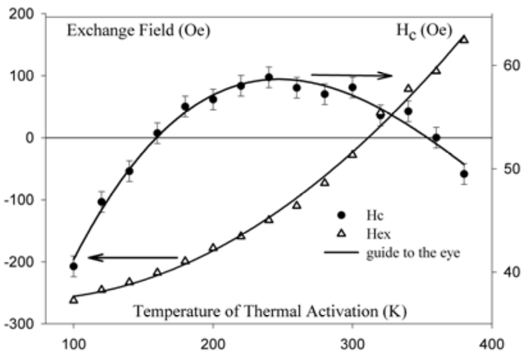

Fig. 4. Variation of H_{ex} and H_C for IrMn(5nm)/CoFe(10 nm).

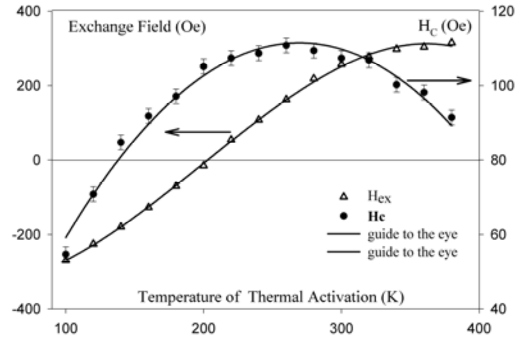

Fig. 5. Variation of H_{ex} and H_C for IrMn(3nm)/CoFe(10 nm).

The temperature at which H_{ex} equals zero, is the mean blocking temperature, T_B. This procedure ensures that almost the whole of the energy barrier distribution is measured. Accordingly, T_B is the temperature at which half of the distribution of energy barriers to reversal in the AF is in the original state, whilst the other half has reversed its state due to thermal activation. Figs. 6 and 7 show the energy barrier distributions, *f*(ΔE), obtained by differentiation of $H_{ex}(T_{Act})$. From this figure it is clear that a distribution of energy barriers exists within IrMn with significant barriers extending to low temperatures.

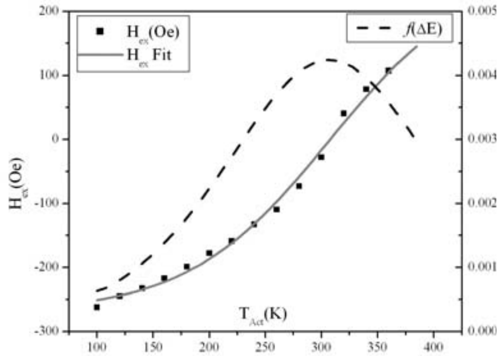

Fig. 6. Distribution of energy barriers to reversal in IrMn(5nm)/CoFe(10 nm).

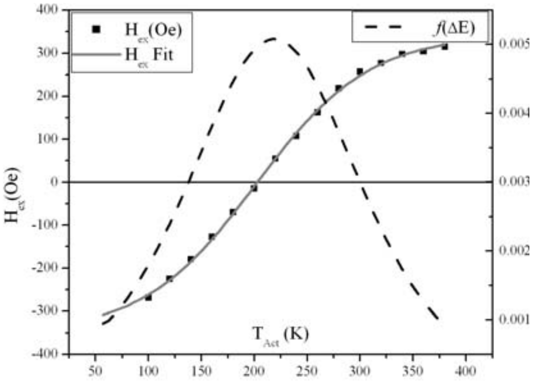

Fig. 7. Distribution of energy barriers to reversal in IrMn(3nm)/CoFe(10 nm).

Figs. 6 and 7 show that for the samples measured the volume of the AF that is pinning the F at room temperature is 53% of the total for IrMn(5 nm) while for IrMn(3nm) it is only 16%. These percentage values are estimated as the percentage of the area under the curve of the normalised distribution of energy barriers, $f(\Delta E)$, between the temperature of interest, 300 K, and infinity. The area under the curve below the temperature of interest, T_{Act}, corresponds to superparamagnetic spins with lower blocking temperature than T_{Act}.

From Figs. 4 and 5 it is clear that T_B obtained from this procedure does not correlate with the peak in the value of H_C [10]. This is in contrast with the procedure for determination of the blocking temperature generally used. Such technique involves the measurement of H_{ex} as a function of temperature via sequential measurements of hysteresis loops which does not involve each loop being measured with the AF in a defined state and allows for sequential time dependent, thermally activated reversal when the F is cycled. Therefore, T_B is not defined in terms of the energy barrier distribution in the AF and such measurements do not represent $f(T_B)$.

5. Training effects

The dependence of training on temperature for IrMn(5 nm)/CoFe(10 nm) has been studied and the results obtained are presented in Table 1. Three hysteresis loops were measured consecutively, labelled n = 1, 2 and 3. The exchange field stabilised for n = 3. These data have been obtained after resetting the AF prior to every measurement using the conditions specified in section 4. The sample was then quenched to the temperature of measurement, and subsequently hysteresis loops were measured. Previously it was determined that the thermal activation free temperature for this sample was 100 K. Above this temperature the reduction of H_{ex} with field cycling includes thermal activation effects. Below this temperature, thermal activation of the AF grains or domains is not able to overcome the energy barriers to reversal. In this range of temperatures the anisotropy energy of the AF dominates over thermal activation.

Table 1. Training effects at a range of temperatures for IrMn(5nm)/CoFe(10 nm).

T(K)	$H_{c1}(n)$ (Oe)			$H_{c2}(n)$ (Oe)			ΔH_{c1}	ΔH_{c2}	$H_{ex}(n)$ (Oe)		
n	1	2	3	1	2	3	1 to 2	1 to 3	1	2	3
16	-417	-394	-393	-338	-338	-338	23	0	-378	-366	-365
20	-410	-388	-387	-336	-335	-336	22	0	373	-361	-361
40	-394	-374	-373	-321	-322	-322	20	-1	-358	-348	-348
80	-372	-352	-349	-301	-302	-302	20	-1	-337	-327	-326
100	-336	-318	-316	-283	-283	-282	18	1	-310	-300	-299
175	-290	-270	-267	-231	-231	-229	20	2	-260	-250	-248
225	-249	-227	-224	-184	-183	-183	22	1	-216	-205	-204
275	-211	-186	-182	-133	-133	-132	25	1	-172	-159	-157
325	-152	-129	-125	-70	-68	-67	23	3	-111	-98	-96
375	-99	-85	-82	-13	-12	-12	14	1	-56	48	-47

Our attention has been focused on the behaviour of the first, H_{c1}, and second, H_{c2}, coercive points of each hysteresis loop and we have observed two different sources of training. There is source of training that is temperature dependent and affects both H_{c1} and H_{c2}. This is observed above T_{NA} when the times spent by the F at positive and negative saturation are not the same. This leads, generally, to a shift towards positive field values for H_{ex}, H_{c1} and H_{c2} due to thermal activation of the AF grains. This shift increases with the number of cycles if the time spent at negative saturation is larger than at positive saturation. A contribution of thermal activation to the shift of H_{c1} and H_{c2} towards negative values would be expected with field cycling when the amount of time spent at positive saturation is larger than that spent at negative saturation. The data shown in Table 1 were obtained from hysteresis loops in which the amount of time spent of both positive and negative saturation by the F was kept as equal as possible, except for the shift that H_{c1} experiences after the first reversal of the F due to spin reorientation on the second and third loop. This type of training is a consequence of the biaxial anisotropy of the $Ir_{30}Mn_{70}$ [18]. This leads to a relaxation of the AF magnetic state to a second energy minimum. This is also reflected in the asymmetry of the loop which is more square for the first reversal of the F than for subsequent loops. This effect appears to be largely temperature independent. However, recent studies on the same system IrMn/CoFe [19], following the same procedure, showed that there is a progressive reduction in the degree of squareness of the first loop, implying that there is a distribution of energy barriers to spin reorientation. Given that our measurement procedure held the F at positive and negative saturation for almost equal periods of time and that the effects of spin reorientation persist to 4.5K for both IrMn(5nm) and IrMn(3nm), it appears that the spin reorientation is not entirely a thermally activated process and appears to have thermal and athermal components.

The origin of this distribution is not clear but the presence of a distribution implies that spin reorientation is a purely rotational process rather than a domain wall effect. Such distributions can arise from a spread of grain volumes or from variations in the anisotropy due to crystal defects or orientation variations in the direction of the cubic edges in the AF relative to the field direction.

6. Grain-size effects

Two sets of five FeMn/NiFe samples were grown with different thicknesses $t_{AF} = 7$ and 10 nm, and labeled t-7 and t-10. For each set the grain size was varied in the AF layers only, i.e., the grain size was not varied in the F, seed, or cap layers. The grain size of the lognormal distribution was characterized by the median diameter D_m, which is given by,

$$D_m = e^{\mu}, \qquad \mu = \langle \ln D \rangle . \qquad (1)$$

Where D is the measured grain diameter. In the range measured, D_m showed a monotonic increase with sputtering rate [20]. Hence, five pairs of samples were obtained with the same D_m and different t_{AF}. Hysteresis loops were measured at room temperature on as-prepared samples allowing thermal activation of lower energy barriers when the F is in reverse bias and the possibility of some AF grains going into a quasi-superparamagnetic state.

Fig. 8 shows the monotonic increase of H_{ex} with increasing D_m. The close correlation between H_{ex} and D_m indicates that the increase in H_{ex} is due to increasing grain size in the AF. This indicates that for FeMn the energy barriers to reversal are grain size dependent although we cannot determine if this is an area or volume effect. The increase in H_{ex} with increasing grain size contradicts the idea that smaller grains produce larger H_{ex} due to a larger number of uncompensated spins in the AF/F interface [21].

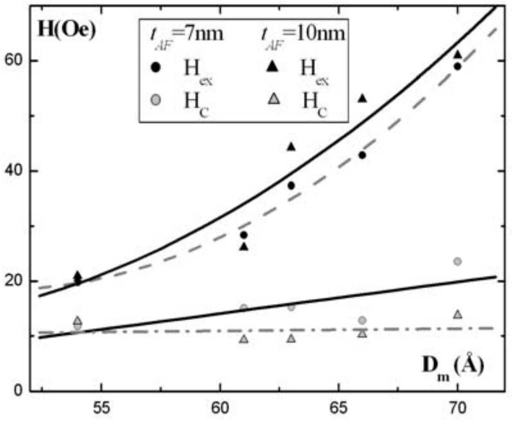

Fig. 8. H_{ex} and H_C as a function of D_m.

We find that at room temperature, thermal activation over grain size dependent energy barriers dominates over grain-boundary effects leading to a decrease in H_{ex}. The effect of the grain size on H_C is much smaller than that on H_{ex}, pointing to the different origins of these effects. We believe that H_{ex} is determined by the degree of order in the AF, whereas H_C arises due to interfacial effects. At room temperature a significant fraction of the AF does not have a stable orientation even though T_N has not been exceeded. Hence it is in a quasi-superparamagnetic state.

Our results indicate that the exchange bias is grain-size dependent but more complex spin structure effects at the interface cannot be excluded. Also, the origins of exchange bias and spin reorientation show that each effects occurs over a distribution of energy barriers. Therefore, these results show that exchange bias is a complex phenomenon involving a combination of thermal effects in the AF and complex spin correlations in the AF.

7. Conclusions

We have studied in detail the phenomenon of training and its dependence on temperature [9]. This phenomenon consists of a shift of the hysteresis loop back towards zero field when a sample that was not previously exposed to a reversed field is exposed to field cycling. This results in a reduction of the value of H_{ex}. By measuring training both above and below T_{NA} we are able to distinguish thermal from athermal effects in this complex phenomenon.

Acknowledgements

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