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Magnetic viscosity measurements reveal reversal asymmetry in exchange-biased bilayers

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We have used time-dependent magnetization measurements to probe the asymmetry in the magnetization-reversal mechanisms in exchange-biased MnF_2/Fe bilayers. Analysis of the magnetic viscosity on the magnetizing and demagnetizing sides of the exchange-shifted hysteresis loops reveals a striking asymmetry. This is due to different mechanisms for reversal on the two sides of the loop as elucidated previously by polarized-neutron reflectometry and anisotropic-magnetoresistance measurements. The viscosity data reveal asymmetric features that are intrinsically related to the nature of the exchange anisotropy.

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Asymmetrically shaped hysteresis loops have been observed in the field of exchange-induced anisotropy for over 40 years.[1,2] It is only very recently that experimental attention has been paid to understanding the exact nature of this magnetization-reversal asymmetry.[3–6] Work on the FeMn/NiFe system revealed an asymmetry in the domain-wall nucleation process on the magnetizing and demagnetizing branches of the hysteresis loops[3] and similar observations have been made in other systems.[5,6]

In the MnF_2 and FeF_2/Fe systems we observed a very stark asymmetry in certain cooling-field orientations where the reversal occurred by coherent rotation of the magnetization on the left-hand side of the negatively exchange-biased hysteresis loops, and by domain-wall nucleation and propagation on the right-hand side.[4] This simple observation was found to be due to the twinned nature of the antiferromagnet in the presence of perpendicular coupling[7] between the antiferromagnet (AF) and the ferromagnet (F). In essence, the tendency to perpendicular coupling for each individual twin leads to an effective ''45°'' coupling between the AF and the F. This unique anisotropy in the F layer leads to a situation where coherent rotation is favored on the left-hand side of the loop, while domain nucleation and propagation is favored on the right. References 4, 8, and 9 provide a more complete description of the situation. This reversal asymmetry, which was originally observed using polarized-neutron reflectivity,[4] can also be probed by using the anisotropic magnetoresistance (AMR) effect.[8,9] The sensitivity of the sample resistance to the angle between the magnetization direction and the current vector allows for observation of distinctly asymmetric resistance hysteresis loops when the magnetization reversal mechanisms are asymmetric.

Although this specific type of reversal asymmetry is restricted to the exact situation of twinned antiferromagnets it is clear that other systems[3,5,6] also display asymmetric-reversal phenomena. The FeMn/NiFe work[3] is a perfect example; an asymmetry exists but it is of a slightly different nature than that seen in MnF_2/Fe. At present the extent to which these asymmetrical effects are general to exchange-biased systems is unknown. In this paper we show that the *time dependence* of the magnetization can also reveal asymmetries in the magnetization reversal. These asymmetric features can be understood in terms of the same concepts outlined above. We suggest that the time-dependent measurements provide a powerful tool for studying the nature of the asymmetry in reversal mechanisms in exchange-biased systems. Such measurements can be used to clarify the generality of the reversal asymmetry in other systems.

Magnetization reversal is a thermally activated process. If a ferromagnetic material is saturated in a given direction then the field is reduced to a point close to the coercive field of the material, the magnetization will show a time dependence due to thermal excitation over the barrier to reversal. If this is characterized by a single energy barrier then the magnetic moment m (the sample magnetization is denoted by M) is given by an Arrhenius law,[10]

$$m(t,H) = A + B(H)\exp(-t/t_0), \tag{1}$$

where

$$\frac{1}{t_0} = f_0 \exp\left(\frac{E_A}{k_B T}\right). \tag{2}$$

Here, A and B are constants, H is the applied magnetic field, t represents the elapsed time, E_A is the activation energy, T is the measurement temperature, t_0 is the characteristic time scale, and the parameter f_0 is the ubiquitous ''attempt frequency.''

In real systems it is more likely that there are many barriers with a corresponding distribution of barrier heights. For a wide range of barrier heights the expected time dependence of the moment is the ''classic'' logarithmic form where[10]

$$m(t,H) = C(H) + S(H)\ln(t) \tag{3}$$

with $S(H)$ being termed the ''magnetic viscosity'' and $C(H)$ a constant at a given measuring field. $S(H)$ is expected to reach a peak value near the coercive field H_C where the rate of change of the moment with time reaches a maximum. Note that the intermediate case with a small distribution of barrier heights is more complicated to quantify[10,11] although it has been claimed that an exact analytical solution to such a problem will involve a series expansion in higher powers of $\ln(t/t_0^*)$, where t_0^* is another characteristic time scale. Physically this corresponds to a situation where the moment against the logarithm of time is ''S-shaped'' close to the coercive fields.[11]

In this paper we show that the magnetization-reversal asymmetry is apparent in the form of the time dependence of the moment and more clearly in the field dependence of the parameter $S(H)$.

MgO (substrate)/ZnF_2 (250 Å, buffer layer)/MnF_2 (600 Å, AF)/Fe (120 Å, F)/Al (30 Å, cap) layers were deposited by high-vacuum electron-beam evaporation in a system with a base pressure in the 10^{-8} Torr range. Details were given in a previous publication.[12] High-angle x-ray diffraction with the scattering vector both in and out of the sample plane, grazing incidence reflectivity, and reflection high-energy electron diffraction were employed to characterize the structure. Briefly, the flourides are twinned epitaxial [110] layers with the rutile crystal structure while the Fe overlayers are polycrystalline. To within experimental uncertainty, the Néel temperature of the layers is identical to the bulk temperature of 67.3 K. Time-dependent measurements were made on several samples but the data in this paper are restricted to a single representative sample. The AF/*F* interfacial roughness as determined from the reflectivity profiles was found to be ~7 Å for this sample.

The time-dependent magnetic-moment measurements were made in a commercial Quantum Design superconducting quantum interference device magnetometer from $10<t<10^5$ s and at temperatures in the range $4.5<T<300$ K. Several precautions need to be made when attempting such measurements in magnetometers of this type. The basic complication is that the sample moment is measured by moving the sample vertically through a set of counterwound gradiometer coils. The moment is then deduced from the exact shape of the spatial dependence of the signal voltage. This causes two problems: (i) the measurement takes a finite time and (ii) the sample may experience deviations from the nominal magnetic field due to magnetic field inhomogeneity. To counter these problems the spatial range of the sample sweep is limited to ±1 cm, which results in a measurement time of 1.5 s. We restrict our range of interest to $10<t<10^5$ s where this represents a negligible error. The field inhomogeneity over such distances is of the order of 0.001%, which is considered to be of little consequence. Direct measurements of the superconducting quantum interference device signal voltage as a function of time without moving the sample are consistent with the data shown in this paper, albeit with increased noise levels. During the collection of a time-dependence data set the temperature was stabilized to ±20 mK at low temperatures and to ±50 mK at 300 K. Magnetic-viscosity experiments on exchange-biased polycrystalline spin valves[11] have pointed out that changes in the AF spin structure during the measurement period cause variations of the effective field generated by the exchange-induced anisotropy, leading to the possibility of difficulties with interpretation. Epitaxial samples such as the ones used in this study display no training effects or differences between irreversible and reversible measurements.[13] We can therefore be confident that no such effects influence our measurements.

Figure 1 shows the magnetization hysteresis loop of a typical sample at 300 K and after cooling in a field $H_{FC}=+2$ kOe to 10 K. The exchange bias (H_E) and coercivity are

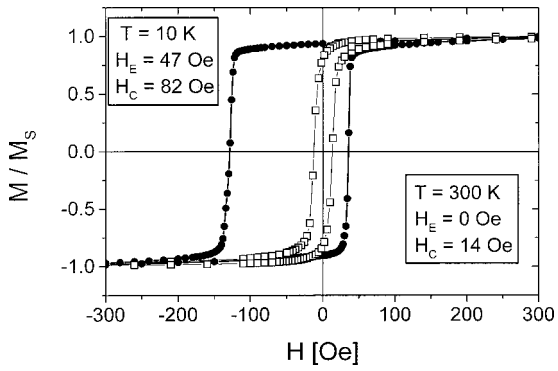

FIG. 1. Magnetization hysteresis loops taken at 300 K (open circles) and after cooling in a field $H_{FC}=2$ kOe to 10 K (solid circles). The data are normalized to the saturation magnetization M_S. The values of H_E and H_C are displayed for the two curves.

−47 Oe and 82 Oe, respectively, which is consistent with previous work on samples with similar interfacial roughness and thickness. Note that the hysteresis loop at 10 K reveals a slight indication of reversal asymmetry, in that the apparent ''switching widths'' for the magnetizing and demagnetizing branches are different. Specifically, the left side of the loop (where rotation occurs) has a broader switching than the right (where domain processes occur). This seems intuitively correct given that coherent rotation generally results in reversal over wide field ranges while domain wall motion is usually responsible for almost discontinuous switching.

Time dependent measurements were made on both sides of the hysteresis loop after saturating the magnetization in the opposite direction. For example, the data shown in Fig. 2(a) represent the time dependence of the moment measured after application of −500 Oe followed by application of a field of 35 Oe, i.e., the coercive point on the right-hand side of the loop. This is the side of the loop for which the magnetization reversal occurs by reverse domain nucleation and wall propagation. Figure 2(a) shows that the time dependence roughly follows a logarithmic form over the entire time range investigated, as expected for a system with a wide range of energy barriers to reversal. Domain wall nucleation and propagation in an imperfect film where many suitable nucleation sites and pinning centers are available would be expected to be in this category. Figure 2(b) shows the same plot for the time dependence on the left-hand side of the loop. Here the measurement is made by saturating the film in a field +500 Oe, then applying a field of −125 Oe close to the coercive point on the left-hand side of the loop. This is the side of the loop for which the magnetization reversal occurs by coherent rotation. Here the time dependence of the moment shows slight deviations from the ln(t) behavior of the S-shaped form discussed in the introduction. This is consistent with a small distribution of barriers to reversal. It is not immediately clear why the reversal by coherent rotation would lead to a time dependence consistent with a small distribution of energy barriers. We will return to this point later in the paper. However, it is clear that we are observing an indication of reversal asymmetry from our time-dependent magnetization measurements. It is noted at this point that the total change in moment over the time period

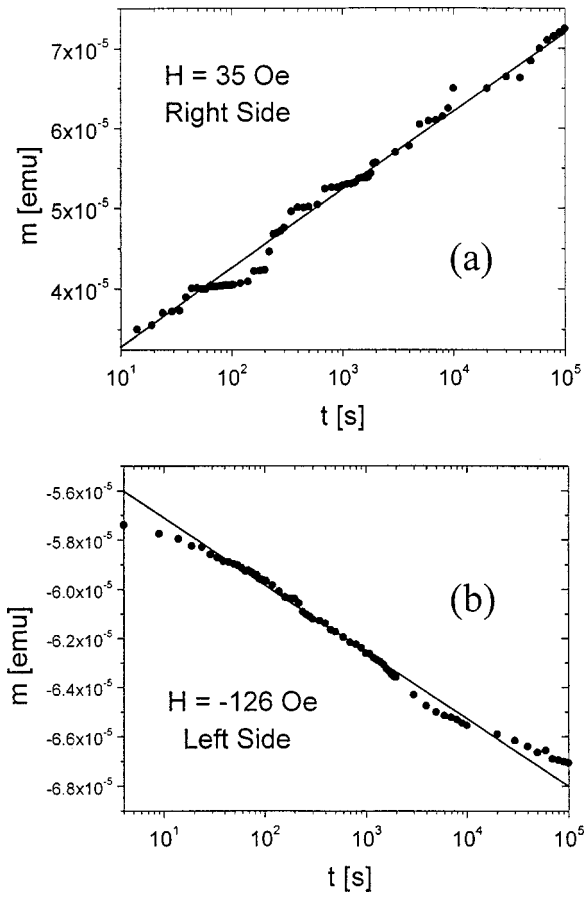

FIG. 2. Time dependence of the magnetic moment m at $T=10$ K for the right and left side of the hysteresis loops after cooling in a field $H_{FC}=+2$ kOe. $H=+35$ Oe in (a) and -126 Oe in (b). The solid lines are fits to Eq. (3) to obtain the viscosity S. A field of magnitude 500 Oe was used to saturate the films before measurement of the time dependence.

probed in this experiment is rather low [a factor of 2 in Fig. 2(a)] due to the low measurement temperature of only 10 K.[14] This makes a detailed examination of the form of the time dependence problematic. Throughout the rest of the paper we simply fit to a logarithmic dependence to extract a magnetic viscosity $S(H)$ (the gradient of the lines in Fig. 2) that simply provides a measure of the magnitude of the magnetization change per unit time. No detailed information is deduced from the exact functional form of $m(t)$, just from the field dependence of S.

The field dependence of the viscosity S in the vicinity of the right-hand side of the loop is shown in Fig. 3. These data were obtained by saturating the film at -500 Oe, applying the measuring field, taking the time dependence, then returning to the saturation field, and repeating the whole process at the next measuring field.[15] As expected we observe a strong peak (full width at half-maximum ≈ 6 Oe) in $S(H)$ at H_C. It is interesting to note that the peak clearly has a Lorentzian-type line shape, although to the best of our knowledge no quantitative theory of the line shape for $S(H)$ data is available. The most striking feature aside from this peak is the existence of the diffuse ''bump'' in $S(H)$ between 0 and ~ 23 Oe. This indicates that a significant (in comparison to the noise level) amount of thermal activation is occurring at fields unusually far from the coercive point. We propose that this observation can be easily explained within the framework previously discussed in the context of the AMR measurements in Ref. 8. The AMR hysteresis loops have an asymmetric form where a significant MR response is observed on the right-hand side of the loop, far from the coercive point. This is analogous to the observation of the ''bump'' in Fig. 3. Essentially, the existence of the unidirectional anisotropy axis favors the formation of reverse domains even far from the coercive field. Hence the effective barrier to reversal is lowered and $S(H)$ has an elevated value compared to the high-field side of Fig. 3.

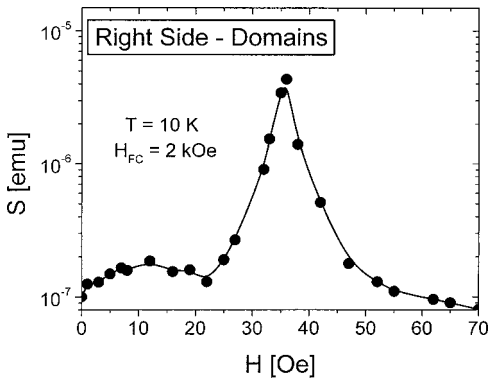

FIG. 3. Applied field dependence of the magnetic viscosity on the right side of the hysteresis loop at $T=10$ K and $H_{FC}=2$ kOe. The solid line is a guide to the eye. Note that this figure and Fig. 4 are plotted over the same range in S and over the same field range of 70 Oe allowing easy comparison.

A dramatically different form of $S(H)$ is observed on the left-hand side of the loop as shown in Fig. 4. The peak we observe at the coercive field is much sharper than that in Fig. 3 having a full width at half maximum of the order of ~ 1 Oe. Again this is consistent with the AMR measurements[8] that revealed a much sharper MR response on the left-hand side of the loop than on the right. Presumably this is due to the fact that the reversal on the left side occurs by coherent rotation of the magnetization vector as determined by polar-

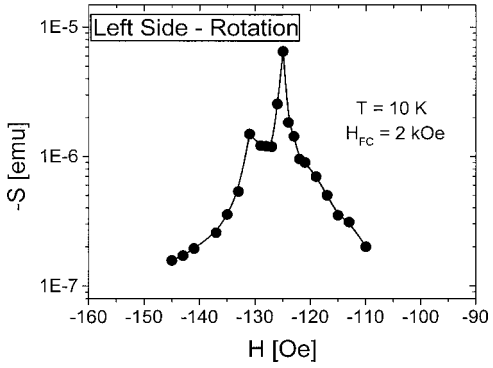

FIG. 4. Applied field dependence of the magnetic viscosity on the left side of the hysteresis loop at $T=10$ K and $H_{FC}=2$ kOe. The solid line is a guide to the eye. Note that this figure and Fig. 3 are plotted over the same range in S and over the same field range of 70 Oe allowing easy comparison.

ized neutron reflectometry.[4,9] The most striking feature of the data in Fig. 4 is the suppression in $S(H)$ between -126 Oe and -130 Oe followed by the possible second sharp peak at -130 Oe. Once more we suggest that this feature is easily understood in terms of the model previously advanced on the basis of the neutron reflectometry and magnetotransport measurements. The basic picture for reversal on this side of the loop is that the magnetization vector coherently rotates to a local easy axis 90° from the H_{FC} direction. The magnetization is then in a metastable state for a range of magnetic field, before the barrier to complete reversal is surmounted. In samples with low values of the full width at half maximum of the high-angle x-ray rocking curve width ($<1.8°$) this can give rise to ''kinks'' in the hysteresis loops[9] and constant values of the MR over this limited-field region where the magnetization vector has fallen into the local-potential minimum. The suppression in $S(H)$ occurs because the magnetization vector falls into this potential minimum resulting in a local maximum in the effective barrier to reversal. As the field is increased to larger negative values, $S(H)$ duly increases before falling to extremely low values ($<10^{-7}$) far from the coercive point.

Finally, the time dependence of the magnetization on the coherent rotation side of the loops has a form that is consistent with a small distribution of energy barriers. This is worthy of further comment. For a perfect sample, free from defects and mosaic spread, the pure coherent-rotation mechanism would lead to an Arrhenius-law time dependence, i.e., a single energy barrier given by the energy required to move the magnetization vector out of the local minimum provided by the 90° easy axis. For real samples, with finite full widths of the high-angle rocking curves, effects such as mosaicity and twinning must lead to the presence of multiple barriers to reversal and therefore the observed time dependence. This is consistent with the neutron-reflectivity observation[4] of a coherently rotated fraction of less than 100% at the coercive point on the left side of the loop.

An important point is that all of these asymmetric features disappear above the Néel temperature of the antiferromagnet, where the exchange-induced anisotropy disappears. At 300 K we simply observe symmetric peaks in $S(H)$ at H_C.

In summary we have used time-dependent measurements of the magnetization to probe the reversal asymmetry known to exist in the exchange-biased hysteresis loops of MnF_2/Fe bilayers. The side of the loop characterized by domain nucleation and propagation shows a logarithmic time dependence with a field dependence of the viscosity that is easily understood in terms of the tendency to form reverse domains at fields far from the coercive point. On the other hand, for the side of the loop where coherent rotation takes place, the magnetization as a function of time shows deviations from a logarithmic time dependence. This is indicative of a narrow distribution of barrier heights. The field dependence of the viscosity on the coherent rotation side of the loop reveals the local easy axis observed previously in polarized-neutron reflectivity and magnetoresistance measurements. This manifests itself as a suppression in the viscosity over a limited range of fields near the coercive point.

These results indicate that this technique is a simple and instructive method for investigating the asymmetry in magnetization reversal mechanisms, which is beginning to be more extensively investigated in exchange-biased materials. Although we have interpreted the data in this paper in terms of the exact nature of the magnetic anisotropy in our system, measurements on other materials should provide equally valuable information. The ultimate goal of such studies will be to understand the extent to which these asymmetric mechanisms are general phenomena in exchange-biased bilayers.

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[14] The temperature of 10 K was chosen to ensure that the exchange anisotropy has reached its saturation value well below $T_N = 67.3$ K.

[15] A finite time period is required to move from the saturating field

(−500 Oe) to the measuring field (this is in the range 0–70 Oe). We first point out that this time is rather short (of the order of 60 s), while the actual period of time spent at any field close enough to the coercive point to induce significant magnetization change is much shorter. Moreover, given that the field change only varies from 500 Oe to 570 Oe over the whole of Fig. 3, it is important to note that this time period is practically constant for all data.