Time domain dynamics of the asymmetric magnetization reversal in exchange biased bilayers

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Abstract: We have explored the dynamics of magnetization reversal asymmetry in exchange biased FeF₂/Fe bilayers using picosecond time resolved Kerr magnetometry. The data reveal an increase in the characteristic precession frequency with decreasing temperature, even above the Néel temperature of the antiferromagnet, which we interpret in term of the previously observed anisotropy enhancement due to antiferromagnetic spin fluctuations. Below the Néel point the magnetization precession is strongly suppressed due to the damping provided by exchange coupling to the antiferromagnetic layer. Dynamic hysteresis loops measured at a fixed delay between field pulse and probe pulse reveal distinct reversal asymmetry that is not observed in the corresponding static loops. The asymmetry takes the form of a suppression of the Kerr rotation signal in the part of the hysteresis loop where nucleation of reverse domains is energetically favorable. This is due to restricted coherent rotation due to the tendency to form reverse domains. The temperature dependence of this dynamic asymmetry is found to be non-monotonic, with interesting correlations with the coercivity. This is discussed in terms of the known coercive mechanisms in these materials.

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A rich phenomenology accompanies the simple exchange shift of the hysteresis loops in antiferromagnet (AF) / ferromagnet (F) bilayers [1,2]. Almost 50 years of research on the exchange bias problem [3] has revealed some fascinating additional phenomena including coercivity enhancements [1,2,4-9], high field rotational hysteresis [1,2,10], training effects [1,2,11], memory effects [1,2,12,13], positive exchange bias [1,2,14,15], exchange bias in the paramagnetic state [16,17], and even a measurement technique dependence [1,2,18-20]. Recently, a lot of attention has been paid to the magnetization reversal mechanisms in AF/F bilayers [21-32], primarily due to the realization that the reversal modes can be asymmetric, i.e. the mechanisms of magnetization reversal are different on the two sides of the same hysteresis loop. This is an interesting consequence of the existence of an exchange induced anisotropy and, due to the fact that it could be expected to further elucidate the origins of exchange bias, this effect has been investigated in numerous systems using methods such as magneto-optical microscopy [21], polarized neutron reflectometry [11,22,23], anisotropic magnetoresistance [23,30,31], magnetic viscosity [26,32], Lorentz microscopy [24], and decoration techniques [25].

Although many systems show some form of reversal asymmetry, the transition metal difluoride AFs have been shown to exhibit a particularly simple form of the effect where the reversal occurs largely by rotation on the left side of the hysteresis loop (the descending field branch) and by nucleation of reverse domains on the right side (the ascending field branch) [22,23,30-33]. This was understood in terms of the interplay between the four-fold anisotropy in the F layer due to perpendicular coupling to a twinned AF, and the unidirectional anisotropy [22,23]. In samples with sufficient crystalline perfection this allowed for the hysteresis loop to be tailored such that the coherent rotation of the magnetization could occur in two distinct stages [23]. Krivorotov et al were able to understand this phenomenon in greater detail using a novel application of the anisotropic magnetoresistance to measure the anisotropy of the exchange coupled F layer [31]. These measurements reveal three-fold terms in the anisotropy expansion, which were demonstrated unequivocally to be the origin of the symmetry breaking in the magnetization reversal. These ideas were expanded [33] to develop a model for the
complex anisotropy that is capable of independently extracting the interfacial coupling strength and the density of uncompensated spins, which are responsible for the exchange bias [34-37].

Despite this interest in the magnetization reversal mechanisms and the general interest in magnetization dynamics [38,39], sweep rate dependent coercivity [40,41], and thermal stability in exchange biased systems, very little work has been done to investigate the dynamics of these reversal mechanisms. Frequency domain techniques such as Brillouin light scattering [42-44] and ferromagnetic resonance [45-47] have been applied, but have focused primarily on investigations of the spin wave damping and ferromagnetic anisotropy, as opposed to direct study of the magnetization reversal mechanisms. In the time domain, picosecond (ps) time resolved measurements have been used to probe the response of AF/F bilayers to ultra-short laser pulses designed to dynamically disturb or temporarily “unpin” the exchange coupling [48], but again magnetization reversal modes were not studied specifically. In this paper we describe the results of ps time resolved Kerr effect measurements [49,50] to probe the dynamics of the asymmetric reversal modes in FeF$_2$/Fe bilayers. In addition to an increase in the characteristic precession frequency with decreasing temperature (consistent with an increase in the anisotropy) we also observe a strong suppression of precession below the Néel temperature ($T_N$) of the AF layer. Measurement of the field dependence of the Kerr rotation at a fixed delay between pump and probe pulses allows us to measure dynamic hysteresis loops, which exhibit distinct differences to static loops. Specifically, we observe disagreement between static and dynamic responses in the ascending field branch of the hysteresis loop, where the static reversal is dominated by the nucleation of reverse domains. We attribute this to a reduced capability for coherent rotation, and therefore precession, at the magnetic field values where the nature of the F anisotropy dictates a preference for domain nucleation. The temperature dependence of this dynamic asymmetry is intriguingly correlated with the temperature dependence of the coercivity. This is discussed in terms of prior experimental and theoretical work on the origins of the coercive mechanisms in various temperature regimes.
The samples were deposited by high vacuum electron beam evaporation using methods described previously [4,14,15,22,23]. Samples of structure MgO(100) / FeF$_2$(110) / Fe (poly) / Al were used in this work with layer thicknesses of 800, 120 and 30 Å, respectively. (The Al is a capping layer to prevent oxidation.) The heterostructures were characterized by wide-angle X-ray diffraction and grazing incidence X-ray reflectivity. The FeF$_2$ layer has a twinned “quasiepitaxial” structure [23], the Fe overlayers are polycrystalline with strong (110) texture, and the interfacial roughness is of order 6 Å. For comparison MgO(100) / ZnF$_2$(110) / Fe (poly) / Al samples were also grown (with similar layer thicknesses) to determine which phenomena are due to the exchange coupling between the F and AF layers (ZnF$_2$ is not magnetically ordered but is isostructural with FeF$_2$). Temperature dependent hysteresis loop measurements were performed by SQUID magnetometry, prior to measurement by time resolved Kerr magnetometry. The geometry for the time-resolved Kerr magnetometry measurements is shown in figure 1. The technique measures the change, $\Delta M$, in the out-of-plane component of the magnetization induced by an ultra-fast magnetic field pulse $H_P$. The temporal width of this pulse is approximately 120 ps at half-maximum, and its magnitude is $\sim 5$ Oe. The pulsed field is applied along the MgO [001] direction and is perpendicular to the applied d.c. field, which is along the [010] direction. The sample is placed with the ferromagnetic film on top of a tapered micro-stripline mounted on the cold-finger of a helium flow cryostat. To implement the measurement, a 76 MHz train of 150 fs pulses from a mode-locked Ti:sapphire laser operating at a wavelength of 800 nm is split into two beams. One beam is focused on a fast photodiode to generate the magnetic field pulses. The second beam is delayed by a variable time $\Delta t$ and is focused to a $\sim 25$ µm diameter spot on the sample. The optical spot is slightly smaller than the width of the stripline. The polar Kerr rotation $\theta_K$ of the reflected probe beam is measured using a balanced detector. $\theta_K$ is measured as a function of the time delay $\Delta t$ between the pump and probe pulses, and the results at each time step are averaged over approximately $10^7$ laser pulses.

Figure 2 shows the temperature dependence of the exchange bias ($H_E$) and coercivity ($H_C$) for the FeF$_2$/Fe (left panel) and ZnF$_2$/Fe bilayers (right panel) after field
cooling in 2 kOe. The data shown are from conventional SQUID magnetometry (solid points) and dynamic hysteresis loops measured by the time resolved Kerr effect (open points), as discussed later in the paper. The temperature dependence, and the absolute magnitudes of $H_E$ and $H_C$, are similar to that observed in previous work on samples with similar thickness and structure. In FeF$_2$/Fe $H_E$ displays a monotonic increase with decreasing temperature below $T_N$ as well as a broad peak in $H_C$ near $T_N$. The ZnF$_2$/Fe data reveal zero exchange bias, as expected, and a weak monotonic increase in $H_C$ with decreasing temperature. The peak in $H_C(T)$ near $T_N$ has been observed before in these difluoride systems [1,51], as well as other materials [52-56], and has been studied theoretically [9]. The existence of the peak near $T_N$ is due to the losses that occur in the AF layer when the F magnetization is reversed as the Néel point is approached. The basic idea is that the fraction of total energy loss that occurs in the AF part of the AF/F bilayer increases near $T_N$. As $T_N$ is approached from below the AF anisotropy is rapidly decreasing, meaning that reversal of the ferromagnet can induce more spin reorientation in the AF layer, and the coercivity increases. This continues until $T_N$ where the AF order is lost and the effect is destroyed, meaning that the coercivity begins to decrease again. Hence the broad peak roughly centered around $T_N$. Various scenarios have been proposed for the exact mechanism for energy loss in the AF. This can be pictured as a consequence of the reduction in anisotropy near $T_N$ in uniform AF layers, leading to an increase in the “dragging” of the AF spins with the F during reversal [1]. Alternatively, taking into account realistic inhomogeneities in the AF the energy losses could arise from reorientation of “loose” or weakly pinned AF spins at grain boundaries, point defects, or twin boundaries. Finally, it has been proposed by Stiles and McMichael [9] that irreversible energy losses take place in the AF layer and that these are responsible for the peak in $H_C(T)$. Clearly these three scenarios are closely related and a consensus exists that the peak is due to loss of energy, by some mechanism, inside the AF layer.

Time resolved measurements on the ZnF$_2$/Fe “control” layers are discussed first in order to provide an example of the behavior typical of a thin Fe layer, with no exchange coupling present. The data of figure 3 show the time evolution of the Kerr rotation from 295 K down to 10 K, for ZnF$_2$/Fe bilayers (left panel) and FeF$_2$ / Fe
bilayers (right panel). Starting with the 295 K ZnF$_2$/Fe data we observe a clear coherent precession of the magnetization vector in response to the tipping pulse. Note that the (in-plane) tipping pulse is oriented perpendicular to the applied d.c. field of 400 Oe. The resulting torque tips the magnetization vector out of plane and the Kerr signal observed is sensitive only to this out of plane component. These data therefore only probe the fraction of the magnetization that coherently rotates out of plane in response to the pump pulse. The ZnF$_2$/Fe precession has a very weak temperature dependence, as illustrated more clearly in figure 4, which plots the temperature dependence of the characteristic precession frequency. This frequency is obtained via Fourier transformation of the raw Kerr rotation data, and is directly related to the applied d.c. magnetic field and the anisotropy field of the F layer. Specifically, the precession frequency is given by [57]

$$\omega = \gamma \left| \sqrt{H_0 + g(\phi)4\pi M_s} \right| H_0 + [1 + f(\phi)]4\pi M_s$$

where $\gamma$ is the gyromagnetic ratio, $H_0$ is the external d.c. field (applied in the plane of the thin film), $M_S$ is the saturation magnetization, and $g(\phi)$ and $f(\phi)$ are functions of the azimuthal angle $\phi$ of the magnetic field relative to the crystal axes and describe the nature of the F anisotropy. For an isotropic thin film, $g(\phi) = f(\phi) = 0$, but they will be non-zero in the presence of magnetocrystalline or exchange-induced anisotropies. In the cases considered here, $f(\phi) \ll 1$ [58]. For a constant applied d.c. field $H_0$, as in this case, the data of figure 4 on ZnF$_2$/Fe therefore imply an almost temperature independent anisotropy, consistent with the weak temperature dependence of the coercivity shown in the right panel of figure 2.

The right panel of figure 3 shows that the situation is very different for FeF$_2$/Fe bilayers. A similar behavior is observed at 295 K, well above $T_N$, but as the temperature decreases the characteristic precession frequency shows a marked increase, even above $T_N$. This is shown explicitly in figure 4. The simplest possible interpretation of these data is that, even at $T > T_N$, the anisotropy of the F layer is increasing with decreasing temperature. Such an effect has been observed before in the FeF$_2$/Fe system, both in the
temperature dependence of the coercivity and the anisotropy determined from FMR measurements, where it was interpreted as an enhancement in the F anisotropy due to AF spin fluctuations [46]. More interestingly, the time resolved Kerr response of figure 3 shows a dramatic suppression of the precession signal below $T_N$, where the AF order sets in and the F layer becomes exchange coupled to the AF [47]. In fact, the amplitude of the precession signal decreases by a factor of 3 on cooling from 100 K to 10 K and at the lowest temperatures no precession signal can be observed beyond 500 ps. This is consistent with previous measurements of the FMR linewidth [46,47] and the aforementioned theoretical notions on the origin of the coercivity enhancement in AF/F bilayers [9] and is responsible for a minimum temperature at which the precession frequency data for FeF$_2$/Fe can be acquired. We believe that the precession signal is decreased by the additional damping provided by the exchange coupling to the AF spins [47]. The mechanisms by which this occurs will be discussed in more detail below. It should be noted at this stage that it is also possible that inhomogeneous dephasing is responsible for the suppressed precession signal [59]. In either case the additional damping is absent in ZnF$_2$/Fe bilayers (see fig 3), which show no significant change in the precession signal as a function of temperature. In this case the precession can be described by a Gilbert damping parameter, $\alpha = 0.013 \pm 0.002$, which is temperature independent.

As mentioned in the introduction the motivation for our work is to use the time resolved Kerr technique to probe the dynamics of the magnetization reversal mechanisms, which are known to be asymmetric in these systems. One simple way to probe the field dependence is to acquire isothermal “dynamic hysteresis loops” by measuring the field dependence of the Kerr rotation at a fixed delay between the tipping pulse and the probe pulse, as shown in figure 5 at $T = 40$ K. These data were taken at a small delay of 30 ps, which corresponds to the first peak in the precession signal. As noted in figure 1, the change $\Delta M$ in the magnetization during the pulse is approximately equal to the integral of the torque during the probe pulse. In small fields, the measured Kerr signal should therefore be proportional to the magnetization averaged over the optical spot size, provided that the magnetization can rotate coherently during the pulse.
At large magnetic fields, significant precession occurs during the pulse and the Kerr rotation measured at very short delays decreases. When the precession frequency exceeds the bandwidth of the pulse at very large fields, the dynamic magnetization approaches zero. The net result of these effects is a signal with the appearance of a conventional hysteresis loop superimposed on a decaying background at higher fields. It must be stressed though that the hysteresis loops measured in this manner are only sensitive to the part of the magnetization that is able to undergo coherent rotation on ps time scales, an essential difference to conventional “static” hysteresis loops.

In examining these data it is first important to note that the dynamic loops measured in this fashion give $H_E$ and $H_C$ values very similar to those measured by conventional (static) magnetometry, as shown by the agreement between open and solid points in figure 2. It is likely that the small deviations are due to the use of different cooling fields for the two data sets (1.1 kOe for dynamic and 2 kOe for static), or unavoidable differences in magnetic field alignment in the two experiments. The unusual phenomenon revealed by the data of figure 5, which is not observed in the corresponding static loops, is the existence of a distinct asymmetric appearance to the dynamic hysteresis loops. [Note that our prior work has shown that the static hysteresis loops appear symmetric despite the strong asymmetries revealed by more direct probes of the reversal mechanisms such as PNR [22] and AMR [23,30,31,33]]. The asymmetry observed in the dynamic loops below $T_N$ (i.e. figure 5) is observed on the ascending branch of the loop (the right side), and at negative magnetization. This is the point at which the numerous prior investigations of the static reversal asymmetry [22,23,30-33] have shown that the angular dependence of the anisotropy energy constrains the system to nucleate reverse domains. It can be seen from figure 5 that the asymmetry manifests itself as a suppression of the Kerr rotation indicating a reduced ability of the system to coherently rotate the magnetization vector out of plane in response to the tipping pulse. We suggest that this is a direct consequence of the known static reversal asymmetry which dictates that at this point on the hysteresis loop (i.e. the lower right hand quadrant) the favored reversal mode is the nucleation of reverse domains, which would not
contribute at all to the Kerr rotation signal. The key point here is that the magnetization is unable to respond by coherent rotation on the ps time scale. Note that the magnitude of the Kerr rotation is close to zero at an applied d.c. field of $-H_E$, indicating that at this point the coherent precession is completely suppressed. This is consistent with previous PNR measurements [22] detecting no coherent rotation on the right side of the hysteresis loop for FeF$_2$/Fe, directly implying that domain reversal is completely dominant.

Similar dynamic loop measurements performed between 120 K (above $T_N$) and 10 K (figure 6) reveal an unexpected temperature dependence of the dynamic reversal asymmetry. As expected, the asymmetry disappears above $T_N$, but the temperature dependence is non-monotonic below this, with the maximum asymmetry occurring at 40 K, and the effect becoming noticeably weaker at 10 K. This is shown more clearly in figure 7 which plots the temperature dependence of the dynamic asymmetry parameter, $\Gamma$. This quantity is defined by $\Gamma = [1 - (\theta_{\text{ascend}} / \theta_{\text{descend}})]$, where $\theta_{\text{ascend}, \text{descend}}$ are the Kerr rotations at $H = -H_E$ on the ascending and descending branches of the hysteresis loops, respectively. The asymmetry parameter is therefore 0 for a completely symmetric loop and 1.0 in the extreme case where the Kerr rotation becomes exactly zero at $H = -H_E$ on the ascending branch (right side) of the dynamic loop. The temperature dependence of the coercivity of this sample from static hysteresis loop measurements is shown for comparison, and reveals an intriguing correlation between the dynamic reversal symmetry and the coercivity. Specifically, the dynamic reversal asymmetry is maximized at the point at which the coercivity reaches a minimum. As previously discussed, the peak in $H_C(T)$ at $T_N$ is thought to be a signature of energy losses in the AF layer due to the reversal of the F layer [1]. This could be due to dragging of “loose” or weakly pinned AF spins when the F layer reverses, or, as a result of irreversible losses in the AF layer when the F layer reverses magnetization due to the exchange coupling between the two [9]. This effect is reduced with decreasing temperature below $T_N$. At lower temperatures the model of Stiles and McMichael [9] suggests that the coercivity is then dominated by losses within the F layer due to inhomogeneities in coupling to the AF. In essence, inhomogeneous local barriers to magnetization reversal exist in the F layer due to spatial inhomogeneities in the AF/F coupling strength. This effect leads to the usual increase in
coercivity with decreasing temperature. The different temperature dependencies of the two coercive mechanisms therefore results in the existence of a minimum in $H_C$ at some intermediate temperature. It is at this point that the dynamic reversal asymmetry is maximized. Although the non-monotonic temperature dependence of the dynamic asymmetry is not entirely understood it is clear from these data that at temperatures below 40 K, where the coercivity begins to increase again with decreasing temperature, the dynamic asymmetry is reduced. Within the framework of the Stiles and McMichael coercivity model this suggests that at low temperatures, when the losses are confined to the F layer and do not occur in the AF, the asymmetry is decreased. It is possible that in this regime, when the static field is increased beyond the point where reverse domain nucleation occurs on the ascending field branch, the magnetization within the individual domains can respond coherently to the field pulse, restoring the symmetry in the dynamic loops.

We have also carried out time resolved Kerr measurements on MnF$_2$/Fe, a lower anisotropy counterpart to the AF/F system discussed here. This system displays a number of differences in comparison to FeF$_2$/Fe including; (i) a weaker damping of the precession signal below $T_N$, (ii) smaller dynamic reversal asymmetry, and, (iii) a precession frequency which is only enhanced below $T_N$. The first observation is likely due to the smaller exchange induced anisotropy, and therefore exchange bias (by a factor of $\sim 4$), in MnF$_2$/Fe compared to FeF$_2$/Fe. This lower value of the exchange induced anisotropy is also related to the second observation, which is consistent with previous measurements indicating that the static reversal asymmetry is stronger in FeF$_2$/Fe [22], leading to the expectation that the dynamic reversal asymmetry would also be stronger. The final observation is consistent with prior FMR data [45] showing an increase in resonance frequency only below $T_N$. The absence of F anisotropy enhancements above $T_N$ is due to the lower susceptibility of the MnF$_2$ AF, which would reduce the spin fluctuation effects. Note that the coercivity enhancement at $T >> T_N$ has only been observed in FeF$_2$/FM systems [46,60].
In summary, we have investigated the time domain dynamics of the magnetization reversal asymmetry in exchange biased FeF$_2$/Fe. In addition to an increase in F anisotropy above the Néel temperature of the AF, the data reveal a strong suppression of the magnetization precession below $T_N$ due to the increased damping provided by exchange coupling to the AF layer. Dynamic hysteresis loops show a distinct asymmetry, which does not occur in the corresponding static loops, which we believe is due to a reduced ability for coherent rotation at the magnetic fields where reverse domain nucleation is known to be favorable. This dynamic reversal asymmetry has a non-monotonic temperature dependence, which, although it is not entirely understood, displays some notable correlations with the temperature dependence of the coercive mechanisms.

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References

58. $f(\phi) \ll 1$ in the limit where all anisotropy fields are small relative to $4\pi M_S$, where $M_S$ is the saturation magnetization. See ref 57 for details.
59. In the situation where the lateral AF domain size is much smaller than the exchange length in the F the inhomogeneities in the local internal field in the F layer could also result in reduced precession. Although the current measurements are unable to distinguish between these two mechanisms our best estimates for the F exchange length and the AF twin domain size suggest that simple damping is more likely than inhomogeneous dephasing.
Figure Captions

**Figure 1.** Schematic showing the geometry used in the time resolved Kerr effect experiments. The current pulse injected down the stripline directly underneath the sample induces a field pulse labeled $H_p$. This induces an out-of-plane magnetization component (the magnetization vector, $M$, and the out-of-plane component due to the tipping pulse, $\Delta M$, are both labeled), which is probed through the Kerr rotation, $\theta_K$, of the plane of polarization of the probe pulse.

**Figure 2.** Temperature dependence of the exchange bias ($H_E$) (top panel) and the coercivity ($H_C$) (bottom panel) for FeF$_2$ / Fe (AF/F) (left panel) and ZnF$_2$/Fe (non-magnetic/F) bilayers (right panel). Solid points are from conventional SQUID magnetometry while the open points are from dynamic hysteresis loops as discussed in the text.

**Figure 3.** Time dependence of the Kerr rotation, $\theta_K$, for ZnF$_2$ / Fe (left panel) and FeF$_2$ / Fe (right panel). The data were taken from 295 K (top) to 10 K (bottom). The applied d.c. magnetic field is 400 Oe.

**Figure 4.** Temperature dependence of the characteristic precession frequency for FeF$_2$ / Fe (solid points) and ZnF$_2$ / Fe (open points) for applied d.c. magnetic fields of 300, 500 and 700 Oe. These data are extracted from curves of the type shown in figure 3 after Fourier transformation of the raw time dependence data.

**Figure 5.** Dynamic hysteresis loop for FeF$_2$ / Fe at $T = 40$ K. These data were taken at a fixed delay between field and probe pulses of 30 ps.

**Figure 6.** Temperature dependence of the dynamic hysteresis loops for FeF$_2$ / Fe. These data were taken in a similar manner to those shown in figure 5.
Figure 7. Temperature dependence of the dynamic loop asymmetry parameter, as defined in the text (left axis, solid points). The coercivity extracted from conventional magnetometry measurements is shown for comparison (right axis, open points).
\[ \Delta M \approx \gamma M \times H_p \, dt \]
Fig. 2
Fig. 3

The figure shows time-domain signals for two different materials, ZnF$_2$/Fe and FeF$_2$/Fe, at various temperatures: 295 K, 200 K, 100 K, 50 K, and 10 K. Each graph represents the angle $\theta_k$ in arbitrary units as a function of time in nanoseconds (ns) with a constant magnetic field $H = 400$ Oe.
Precession Frequency [GHz] vs. T [K]

- FeF$_2$
- ZnF$_2$

- 700 Oe
- 500 Oe
- 300 Oe

Fig. 4
FeF$_2$ / Fe

$\theta_K$ [arb. units]

$H$ [Oe]

$T = 40$ K

$\Delta t = 30$ psec

Fig. 5
Fig. 7