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Novel laser-induced dynamics in exchange-biased systems

A. Porat[1], S. Bar-Ad[1(a)] and I. K. Schuller[2]

[1] *School of Physics and Astronomy, Sackler Faculty of Exact Sciences, Tel Aviv University, Tel Aviv 69978, Israel*
[2] *Physics Department, University of California, San Diego - La Jolla, CA 92093-0319, USA*

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Abstract – Ultrafast optical excitation of a ferromagnet/antiferromagnet (Ni/FeF_2) exchange bias bilayer produces novel magnetization dynamics unlike ever observed before. An unexpected precession of the magnetization, in reverse magnetic fields that exceed the exchange bias, originates from a reorientation of frustrated spins at the interface. As the laser-excited interface approaches the blocking temperature, an exchange bias reversal can also be induced with a single excitation pulse, showing that not only the ferromagnet but also the antiferromagnet is strongly affected by the optical perturbation. This non-trivial response cannot be extrapolated from the known slow dynamics of magnetic bilayers, and provides important information on the physics of the interlayer coupling.

Ultrafast lasers are an extremely powerful research tool, which lends itself to direct investigations of the impulse response of physical systems. This has been implemented in studies of diverse phenomena, from molecular motion and chemical reactions on the femtosecond time scale, to many-body correlations between quasiparticles in condensed-matter systems, to coherent spin manipulation in semiconductors. In recent years it has been widely implemented in studies of magnetic materials, where demagnetization, magnetization reversal and spin precession may be induced through thermal excitation or direct transfer of angular momentum, on picosecond and even sub-picosecond time scales [1–8]. These studies are motivated by a fundamental interest in the strongly non-equilibrium conditions that intense femtosecond laser pulses excite in magnetic systems, as well as by efforts to push the fundamental limit of the magnetization reversal time, which has important consequences for magnetic recording and information processing.

A particularly promising new direction for these types of studies is the ultrafast response of nanostructured magnetic systems in proximity configurations, *i.e.* dissimilar magnetic systems in contact with each other. Exchange Biased (EB) ferromagnetic-antiferromagnetic bilayers are archetypal systems of this type, in which the interfacial coupling between a ferromagnet (FM) and an antiferromagnet (AFM) results in a strong unidirectional exchange anisotropy [9]. Such bilayers have shown many unexpected phenomena over the years [10], and are thus likely to also have a distinctive ultrafast temporal response. We demonstrate this in a study of a Ni/FeF_2 bilayer, in which the coupling across the FM-AFM interface is antiferromagnetic, the interfacial AFM moments are known to couple to the external field, and the magnetic order is particularly sensitive to the effects of temperature and the external field [11]. This causes the bilayer to react in a very unusual fashion to a fast optical excitation. In a reverse magnetic field in which the FM is fully saturated, an ultrashort pump pulse triggers an unexpected precession of the FM, which lasts a few hundreds of picoseconds. When the optical excitation is weak, after the decay of the precession the bilayer reverts to a behavior expected based on slow dynamics. However, when the optical excitation is intense enough to heat the FM-AFM interface to the blocking temperature, the response of the bilayer is dominated by the AFM rather than the FM. In fact, a complete reversal of the AFM can be induced by a single excitation pulse. As a consequence the possibility arises of "writing" information into the AFM. This is unlike other ultrafast dynamics experiments in EB systems, in which the ultrafast dynamics is dominated by the response of the FM, and can be extrapolated from the slow dynamics of the system.

A 50 nm thick layer of FeF_2 was grown epitaxially on a (110) MgF_2 substrate at 300 °C, followed by a 21 nm

(a) E-mail: shimshon@post.tau.ac.il

thick layer of Ni, grown at 150 °C, and coated *in situ* with a 4 nm layer of Al, to prevent the Ni from oxidation. X-ray diffraction shows that the Ni layer is polycrystalline, and the sample has uniaxial anisotropy with the easy axis along the FeF_2 easy axis (001). The blocking temperature of the sample (*i.e.* the Néel temperature of the AFM) is $T_B = 78\,\text{K}$. Before cool-down the sample is magnetized along the easy axis which defines positive fields. The cool-down in a low positive field ($0 < H_{FC} < 500\,\text{Oe}$) produces a negative EB field $H_{EB} = -1430\,\text{Oe}$ [12]. In this field range the results are independent of the magnitude of H_{FC}.

The typical pump-probe setup uses $\sim 60\,\text{fs}$ pulses at 800 nm ($h\nu = 1.5\,\text{eV}$) generated by a 1 KHz Ti:Sapphire regenerative amplifier. The intense pump beam and the weak probe beam are focused to 300 μm and 50 μm diameter spots on the sample, respectively, to assure that a homogeneously excited region is probed. The sample is placed in a continuous-flow liquid-He optical cryostat, at the center of a 2400 Oe Helmholtz coil. The magnetization dynamics are measured using the longitudinal Magneto Optical Kerr Effect (MOKE). In this configuration, two components of $\vec{M}$ are simultaneously detected, M_x and M_z, *i.e.* the magnetization components along the easy axis and out of the sample's plane, respectively (see the inset in fig. 1(b)).

Figure 1(a) shows pump-probe MOKE signals measured as a function of the external magnetic field, with fixed time delays Δt, compared to measurements with the pump beam blocked. Curve A is a hysteresis loop (HL) curve that was measured at room temperature. The other curves were all measured at a temperature of $T = 30\,\text{K}$, and are representative of the results obtained for $4.2\,\text{K} < T < 36\,\text{K}$. Curve B is a typical EB curve obtained without pump excitation, and shows that the magnetization reversal occurs at $H_{EB} = -1430\,\text{Oe}$, in agreement with data in the literature [12]. Excitation with a pump fluence of $1.4\,\text{mJ/cm}^2$ results in a clear change of the MOKE response. At $\Delta t = 330\,\text{ps}$ (curve D in fig. 1(a)) the switching of the magnetization, from the $+\hat{x}$-direction to the $-\hat{x}$-direction, occurs at a lower magnetic field ($H_{ext} \sim -1250\,\text{Oe}$) compared to the no-pump case. This suggests a temperature rise at the interface of $\approx 10\,\text{K}$ (this estimate is based on EB curves that were measured at different temperatures in equilibrium conditions). Measurements at negative Δt (corresponding to an actual delay of the probe of 1 ms relative to the preceding pump pulse) reveal a MOKE signal which is identical to the no-pump signal (curve B). This indicates that between successive pump pulses the system returns to its initial state.

Surprisingly, at $\Delta t = 60\,\text{ps}$ (curve C), two strong excursions of the signal are observed at $H_{ext} \sim -1400\,\text{Oe}$ and $H_{ext} \sim -1750\,\text{Oe}$. These excursions represent Kerr rotations which are larger than the rotations observed at saturation without pump excitation. This may be explained by an out of plane tilt of the magnetization (*i.e.* a non-vanishing M_z). We have therefore carried out similar measurements with the probe set at zero incidence angle

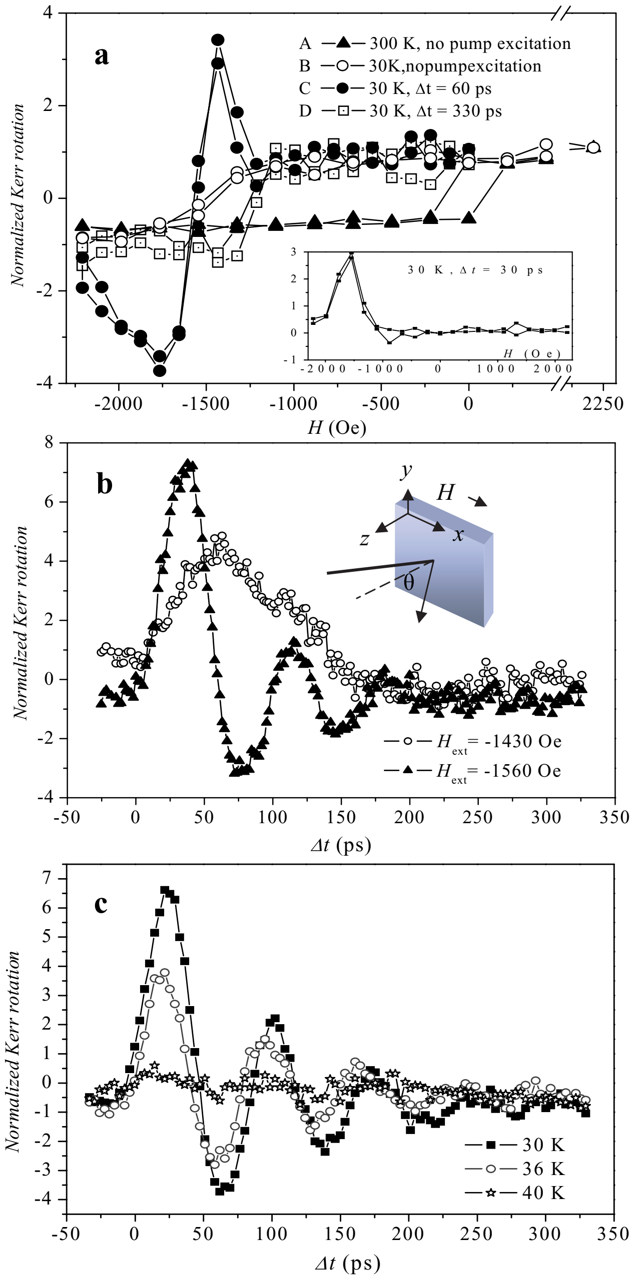

Fig. 1: (Colour on-line) Laser-induced magnetization precession. (a) Pump-probe MOKE measurements as a function of magnetic field, obtained at 30 K, with a pump fluence of $1.4\,\text{mJ/cm}^2$, compared to measurements with the pump beam blocked, at 30 K and at room temperature. The inset shows data measured at zero incidence angle. (b) Time-resolved measurements at 30 K, at different magnetic fields. Inset: the experimental MOKE configuration. (c) Time-resolved measurements at -1560 Oe at different temperatures.

($\theta = 0°$ in the inset of fig. 1(b)). In this configuration the in-plane magnetization M_x does not contribute to the MOKE signal, which therefore reflects the magnetization perpendicular to the sample's plane M_z. The results (see the inset of fig. 1(a)) confirm our hypothesis of a tilt of the magnetization out of the sample's plane.

The interpretation of the transient MOKE excursions signal (curve C in fig. 1(a)) as an out of plane precession is further confirmed by the time-resolved measurements depicted in fig. 1(b). The curve measured at $H_{ext} = -1430\,\text{Oe}$ is representative of data obtained when the external magnetic field is in the range $-1430\,\text{Oe} < H_{ext} < -1330\,\text{Oe}$, *i.e.* slightly above H_{EB} ($-1430\,\text{Oe}$). In this field range the precession appears as a slow rotation of the magnetization vector on a time scale of $\sim 300\,\text{ps}$. On the other hand, in the negative field range below H_{EB} ($-2400\,\text{Oe} < H_{ext} < -1430\,\text{Oe}$) the precession is very pronounced. Typical of this field range is the data measured at $H_{ext} = -1560\,\text{Oe}$, which shows oscillations with a period of $\sim 80\,\text{ps}$ (the period is obviously field-dependent). After 1 ms the magnetization vector always recovers to its initial orientation.

The above results are extremely surprising and unexpected. Earlier laser-induced ultrafast dynamics experiments on NiFe/NiO [13–15], NiFe/FeMn and CoFe/IrMn [16–19], FeF2/Fe [20], and Co/Mn [21] showed that when the FM and AFM layers are decoupled in reverse magnetic fields *smaller* than the exchange bias, a precession of the FM towards the external field is triggered. Such a behavior is indeed expected. In contrast, in our experiment the precession is most pronounced in external fields *larger* than the EB, *i.e.* when $\vec{M}$ has already reversed, and is aligned *parallel* to $\vec{H}_{ext}$. This is totally unexpected, since it is unlikely that a reduction of the EB would trigger a rotation of $\vec{M}$ *away* from $\vec{H}_{ext}$. Also surprising is the fact that a very small temperature increase (10 K as mentioned above) is sufficient to induce the precession, since the heating is insufficient to decouple the FM from the AFM.

The observed precession implies that in the field range where magnetization measurements seem to show that $\vec{M}$ has reversed (*i.e.* $\vec{M} \parallel \vec{H}_{ext}$), this is in fact not so. Either the Ni magnetization is canted, or part of the Ni has not reversed. However, the small temperature rise (from 30 K to 40 K) due to heating by a pump pulse is sufficient to realign the FM Ni completely with the field. This suggests that the data presented above arise from Ni spins which are frustrated by the competing interactions with the external field and the exchange bias. These spins undergo a reorientation transition, from a canted state to alignment parallel to the magnetic field vector, due to an anisotropy change in the above temperature range. Because of this they start precessing after the application of a pump pulse. As the heat dissipates, the frustrated spins settle back in their equilibrium, canted orientation, determined by the competing interactions with the AFM layer and with the external field. Indeed, this scenario is in agreement with slow thermal cycling measurements on similar samples in high magnetic fields, which showed a spontaneous magnetization rotation at an intermediate temperature below the blocking (Néel) temperature [11].

The effects of temperature and pump fluence further confirm that an unusual behavior is observed. The amplitude of the oscillations depends strongly on the sample temperature. While there is no significant change in the range $4.2\,\text{K} < T < 36\,\text{K}$, the oscillations abruptly disappear above $T = 36\,\text{K}$ (fig. 1(c)). This is in agreement with a reorientation transition at 36 K, *i.e.* a change from a canted configuration below 36 K to alignment parallel to the magnetic field vector above 36 K [11]. The behavior as a function of pump fluence is also in agreement with such a transition. At 30 K, the oscillations first appear at $\sim 0.3\,\text{mJ/cm}^2$. Their amplitude sharply increases above this power, reaching a maximum at $1.4\,\text{mJ/cm}^2$, and then slowly decreases until they disappear above $\approx 4\,\text{mJ/cm}^2$.

As the pump fluence is increased and the precession gradually disappears, a dramatic change of the MOKE signal becomes evident. Figure 2(a) shows the time-resolved MOKE signal at 30 K, as a function of the external magnetic field, for a set of fixed Δts, and a pump fluence of $2.8\,\text{mJ/cm}^2$, in comparison with the no-pump MOKE signal. In these measurements a centered hysteresis loop (HL) appears, which does not change as a function of Δt. This is a clear indication of a pump-pulse induced thermal excitation of the FM/AFM interface, which reaches the blocking temperature, resulting in a complete decoupling of the nickel layer from the FeF_2 as the interfacial FeF_2 becomes paramagnetic. The decoupling results in markedly different responses as a function of the externally applied magnetic field: At low fields (below the switching field of the centered hysteresis loop) the decoupled FM maintains its orientation, and the AFM freezes back into the original configuration as the bilayer cools down; On the other hand, at higher fields (above the switching field of the centered hysteresis loop) the FM reverses its magnetization, and the AFM freezes into the *opposite* configuration as the bilayer cools down. Indeed, the width of the centered HL measured at $2.8\,\text{mJ/cm}^2$ is comparable with the coercivity of the nickel, measured in equilibrium at 80 K, just above T_B. Thus the centered HL may represent a complete reversal of the FeF_2 surface magnetization.

To confirm this we carried out the following experiment. Starting with a pristine sample, we measured the no-pump MOKE signal following a momentary exposure (*i.e.* a single pulse up to several seconds at a repetition rate of 1 KHz) of the sample to $2.8\,\text{mJ/cm}^2$ pump excitation, at negative H_{ext} above the coercivity of the centered HL. Figure 2(b) shows the MOKE signal before (curve D) and after (curves E, F) exposures. A partial reversal of the EB occurs, *i.e.* a bifurcated HL appears [12,22], indicating that different parts of the nickel layer are biased in opposite directions. This persistent "magneto-optical writing" effect implies that the FeF_2 layer also reorients. If the same "writing" procedure is repeated at positive magnetic fields, the sample reverts to its original orientation.

At low pump fluences that do not produce a centered HL (below $1.8\,\text{mJ/cm}^2$), "writing" does not occur. As a consequence, when the pump fluence is intentionally

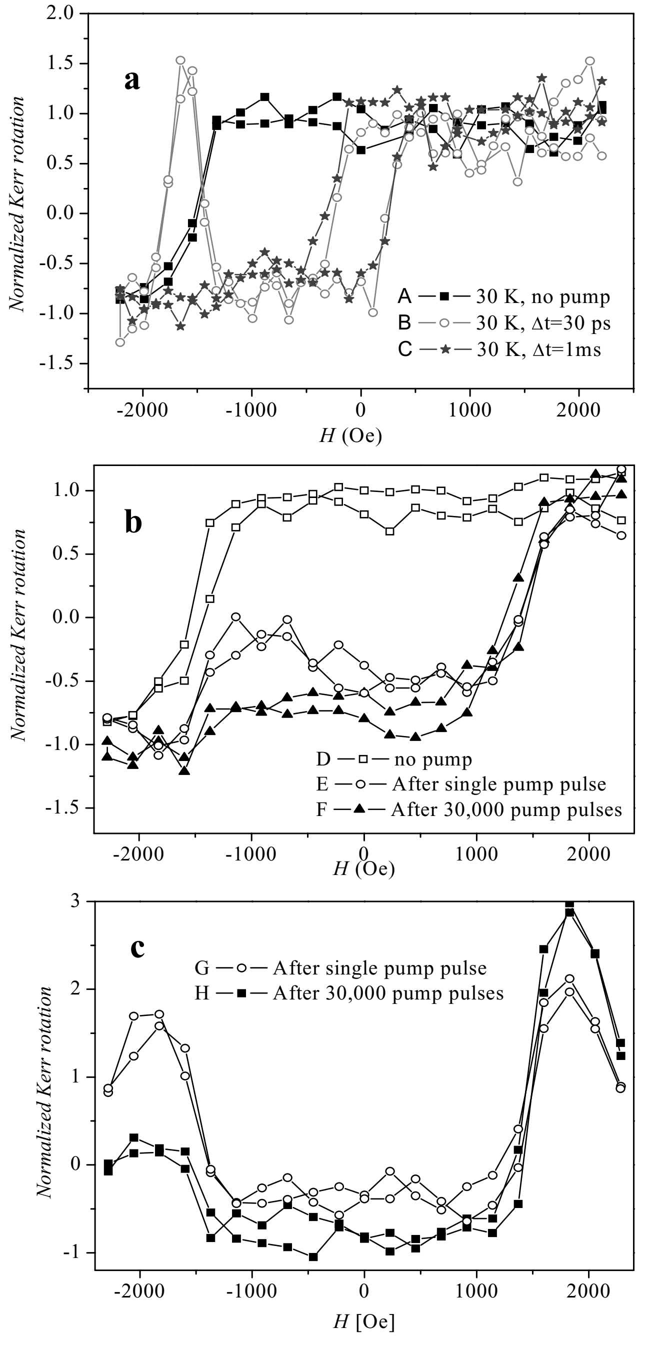

Fig. 2: Laser-induced reversal of the exchange bias. (a) Pump-probe MOKE measurements as a function of magnetic field, obtained at 30 K with a pump fluence of $2.8\,\mathrm{mJ/cm^2}$. (b) Measurements with the pump beam blocked, before and after "writing". (c) Pump-probe measurements at $\Delta t = 30\,\mathrm{ps}$ with a pump fluence of $1.4\,\mathrm{mJ/cm^2}$, after "writing" with a pump fluence of $2.8\,\mathrm{mJ/cm^2}$.

reduced after "writing" in a negative field, a precession signal is detected at both negative and positive magnetic fields (fig. 2(c)). In this case the "written" part of the sample contributes to the precession at positive fields, while the part of the sample which was not "written" contributes to the precession at negative fields. The "writing" also explains the gradual decrease of the oscillations at high pump fluences —only the part which is not "written" contributes to the precession at negative fields. Also note that in the same measurement the "written" parts are "unwritten" (*i.e.* revert to their original orientation) when the field is positive, which explains why the precession is not observed at positive fields (unless the pump fluence is intentionally lowered following "writing" at negative fields). Importantly, the fact that parts of the sample that reverse can nevertheless precess, once the fluence is lowered and the field is positive, also shows that while the "writing" itself is inhomogeneous (either because of the pump intensity distribution or due to inhomogeneous coupling to the substrate), the whole sample contributes equally to the precession. That is, the possibility that the precession is associated with a minority of spins that do not reverse with the bulk of the nickel can be ruled out. Finally, the reversal of the AFM ("writing") is consistent with earlier field-cooling experiments. When the interface temperature approaches the blocking temperature, the frustration leads to a reversal of uncompensated pinned moments in the AFM [23,24], giving rise to a new situation in which the FM feels an opposite exchange bias.

In conclusion, we have observed a strong, unexpected, short time-scale magnetization precession in an exchange biased system. This precession occurs in an unexpected field range, where such a response was not observed before, indicating that exchange bias systems are more complex than hitherto believed. Moreover, the temperature and pump power dependence proves the existence of complex magnetic structures in exchange biased systems even in regions where slow dynamics indicate otherwise. In general, the results show that nanostructured magnetic systems in proximity exhibit an unexpected and rich behavior on short time scales, indicating that they are ripe for discovery of novel phenomena.

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