Ultrafast Time Resolved Photoinduced Magnetization Rotation in a Ferromagnetic/Antiferromagnetic Exchange Coupled System

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Ultrashort pulse laser techniques are applied to study optically induced modulation in exchange biased ferromagnetic/antiferromagnetic (FM/AF) thin bilayer films (NiFe/NiO). Photoexcitation of the FM/AF interface with subpicosecond laser pulses induces large modulation in the unidirectional exchange bias field (H_{ex}) on an ultrashort time scale. The "unpinning" of the exchange bias leads to coherent magnetization rotation in the permalloy film which is time resolved by the experiment and corresponds to a large modulation in the magnetization component ($\Delta M_Z/M_S \sim 0.5$), on a time scale of 100 psec. [S0031-9007(99)09042-0]

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A problem of contemporary interest at both fundamental and applied levels concerns the ultimate speed "limit" for magnetization reversal under external control. Recently, there have been several types of experiments probing the question. For example, Lederman *et al.* [1] studied the thermal activation switching of small Stoner-Wolfarth-type FeO particles, while Doyle and co-workers [2] and Freeman with co-workers [3] applied a short transient magnetic field pulse created by a microstripline (on a nanosecond time scale) to study magnetization switching in magnetic thin films. Most recently, Siegmann and co-workers [4] applied high energy beam generated picosecond magnetic field pulses on CoPt films.

A different avenue to the study of fast spin and magnetization in ferromagnetic (FM) metals has emerged in which ultrashort laser pulses create a nonequilibrium distribution of electrons and their spins on a subpicosecond time scale [5,6]. The ultrafast optical approach raises the prospect of inducing and investigating basic magnetization switching and relaxation phenomena with the system driven by the absorbed photons. As a specific material test system, we have chosen the exchange coupled NiFe/NiO FM/AF bilayer, characterized by its distinct unidirectional magnetic anisotropy [7] which has found important recent use in giant magnetoresistive and spin-valve sensors [8]. The magnetic characteristics of such coupled FM/AF systems show both an effective exchange bias field H_{ex} (shifted hysteresis loop) and an increased coercivity (H_C). The idea in this paper, sketched in Fig. 1, is to create spin excitations by photons at the FM/AF interface so as to abruptly reduce the exchange coupling. When the magnetization of the FM layer is initially biased antiparallel to the external applied field H_A, the optically induced "unpinning" of the exchange is shown below to activate ultrafast switching of the internal field H_{ex}, which provides the driving force for the study of subsequent coherent magnetization dynamics. These dynamics, studied here in the reversible regime of (large) magnetization modulation, depend on the microstructure at the FM/AF interface. In practical terms, the sensitivity of the exchange interaction to photoexcitation has allowed us to obtain significant modulation in the magnetization of the NiFe/NiO system ($\Delta M_Z/M_S \sim 0.5$) on a 100 psec time scale.

Our samples were dc magnetron sputtering polycrystalline $Ni_{81}Fe_{19}$/NiO bilayers on glass substrate, with $H_{\mathrm{ex}} \sim 100$ Oe [9]. The transparency of NiO makes it possible to photoexcite the interface between the FM and AF directly. The low blocking temperature ($T_b \approx 220\,^{\circ}\mathrm{C}$) of the NiO/NiFe system is convenient for breaking the exchange bias *nonthermally*. We seek to create conditions across the FM/AF interface such that the *interfacial* spin/electron temperature $T_{e,s}(t)$ is elevated close to or above T_b, but kept below the Curie point, while the lattice temperature $T_l \ll T_b$. The hot electrons are unable to diffuse into the insulating NiO AF layer. In addition, any domain wall motion is slow on the time scales considered here so that the bulk magnetic structure of NiO is "frozen."

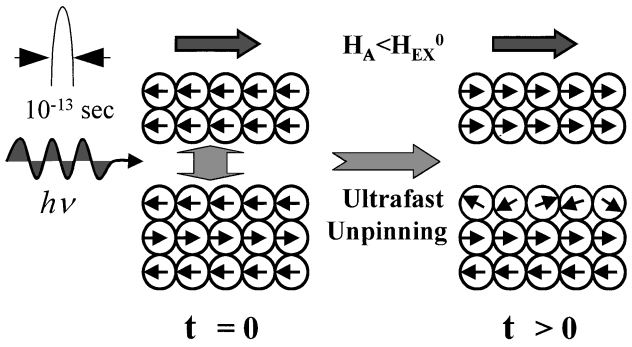

FIG. 1. The concept of optically induced "unpinning" of the exchange bias (field) at FM/AF exchange coupled interface. With the magnetization of the FM layer is initially biased antiparallel to external field, ultrashort pulse photoexcitation triggers a coherent rotation process.

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In our experiments, excitation pulses from a mode-locked Ti:sapphire laser ($\tau_p = 120$ fsec, $h\nu = 1.4$ eV) were directed normal to the sample through the transparent glass substrate and the NiO layer ($E_g > 5$ eV), and absorbed within the NiFe layer [9]. To reduce average lattice heating, we use a pulse repetition rate of 1.9 MHz. The optically induced changes in the magnetization of the samples were recorded by measuring the transient changes in the (longitudinal) Kerr rotation of the NiFe layer, labeled below as $\theta_K'(t)$, using time delayed weak (<0.2 mW) probe pulses in the blue ($h\nu = 2.8$ eV). The Kerr instrumentation employed a polarization-sensitive optical bridge and a low-noise differential detector [6,9].

The time-resolved longitudinal Kerr effect $\theta_K'(t)$ probes the off-diagonal component of the conductivity tensor σ_{xy}, which is proportional to magnetization and the net spin polarization [6,9]. Two contributions are found in our pump probe experiments, distinguished by their different time constants and field dependence. First, following the subpicosecond impulse of photoexcitation, the hot electron and spin distributions are displaced from thermal equilibrium, leading to modulation in θ_K because of spin occupancy effects and modulation in the net spin polarization. Recent studies in thin FM films have given information about the dynamics of spin relaxation [5,6]. The proportionality of the longitudinal Kerr effect to the inplane component of magnetization makes the transient experiment also sensitive to photoinduced changes in the *direction* of **M** due to coherent rotation, a key feature in this paper.

Figure 2(a) displays "snapshots" of the easy-axis transient Kerr hysteresis loops acquired from measurements of $\theta_K'(t)$ on a 100 Å/400 Å NiFe/NiO bilayer sample at a probe delay of $t = 1$, 50, and 200 psec, following the impulse of pulsed laser excitation at $t = 0$ ($\sim$1 nJ focused to a 30-μm-diameter spot). The results are typical of several different samples studied, and should be compared with the static (unperturbed) Kerr loop for the sample (dashed lines in the top panel). First, we find that within about 1 psec the transient Kerr loops show a significant reduction of the $H_{\rm ex}$. Some "softening" of the loop is seen, demonstrating direct electronic access to both the exchange coupling and the spins in the NiFe layer (increased T_S). On the other hand, the coercivity remains nearly unaffected.

With increasing time delay, the photoexcited, coupled FM/AF bilayer systems relaxes via electron, spin, and lattice interaction. Over several tens of psec, the transient Kerr loop, while being reduced in amplitude due to these relaxation processes, begins to display a pronounced change in its shape. An asymmetric distortion sets in against the trend of a monotonically decreasing $\theta_K'(t)$, the effect being concentrated in the lower right corner of the loop, which becomes quite dramatic at $t =$ 200 psec; a more detailed discussion of the time dependence is given below. *We emphasize that in the time-resolved longitudinal Kerr configuration $\theta_K'(t)$ provides a measure of the pump-induced modulation of the magnetization, and contains a contribution which is proportional to the changes of magnetization component in the plane of incidence, ΔM_Z.* The large "negative Kerr loop anomaly" implies directly that coherent magnetization rotation is triggered upon photoexcitation of the bilayer. When calibrated against the static Kerr rotation, this magnetization modulation, induced by each pump laser pulse in Fig. 2(a) and first peaking at $t \sim 150$ psec, corresponds to approximately $\Delta M_Z/M_S \sim 0.4$ under our experimental conditions, implying an average magnetization rotation of about 53°. We have achieved $\sim$100% switching at higher excitation levels; however, in that case irreversible effects take place. Here, the system returns to its initial equilibrium state after each pulse as verified by measuring the static hysteresis loops after each run.

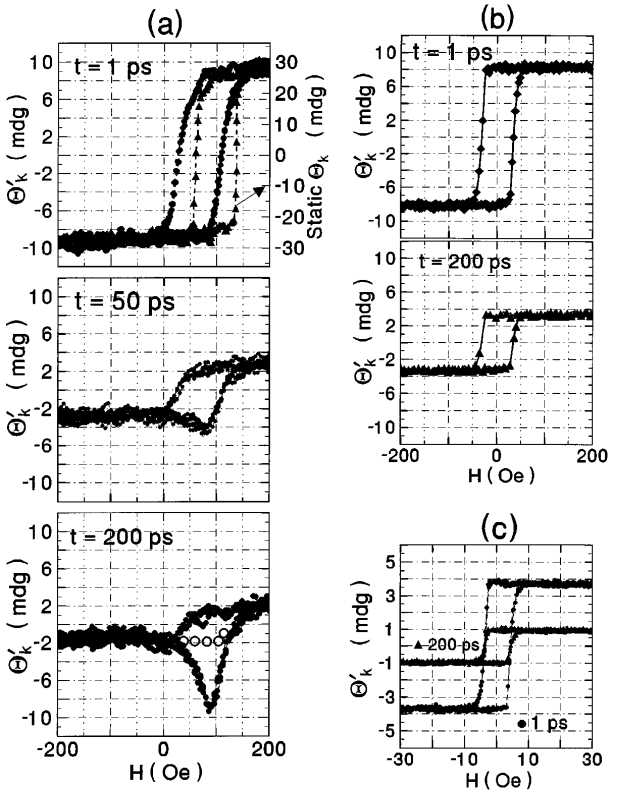

FIG. 2. (a) Easy-axis transient Kerr loops for a photoexcited NiFe/NiO exchange biased bilayer at $t = 1$, 50, and 200 psec following pulse photoexcitation at $t = 0$. The open circles inserted in the bottom trace show expected behavior in the *absence* of coherent magnetization rotation. (b) Transient Kerr loops at $t = 1$ and 200 psec for a thermally quenched NiFe/NiO bilayer companion sample. (c) Transient Kerr loops at $t = 1$ and 200 psec for a NiFe thin film. Both control samples show no sign of magnetization rotation.

Important supporting information was provided by two types of control samples which showed no evidence for magnetization rotation, even in a considerably higher excitation regime than in Fig. 2(a). First, Fig. 2(b) shows two transient Kerr loop snapshots for a NiFe/NiO bilayer sample from the same wafer, whose exchange bias had been intentionally quenched by thermal annealing.

Neither were anomalies seen in a 100 Å thick NiFe single epitaxial layer, shown in Fig. 2(c). Accordingly, we interpret the data in Fig. 2(a) as direct evidence for magnetization switching on an ultrafast time scale ($\sim$100 psec), triggered by photoinduced unpinning of the exchange coupling. The microscopies of the unpinning are presumed to involve the uncompensated spins at the NiO surface and the adjacent ferromagnetic spins in NiFe [10]. Note that the large magnetization modulation is observed only in easy-axis configuration where the external field H_A is *antiparallel* to both the built-in $H_{\rm ex}$ and the static magnetization of the FM layer, i.e., in the "third quadrant" of the hysteresis loop. This is consistent with the constraints imposed by the exchange coupled system as shown below.

The time evolution of the transient magnetization in the bilayer is illustrated in Figs. 3(a) and 3(b), for both easy and hard axis directions, respectively, at $H_A = 80$ and -220 Oe. [The calculations leading to Figs. 3(c) and 3(d) will be discussed below.] Focusing on Fig. 3(a), the $\theta_K'(t)$ traces show a very short ($<$1 psec) initial hot spin transient due to the contribution of occupancy factor/spin effects already mentioned. For the applied field of $H_A = -220$ Oe, optically induced magnetization switching is not possible since in negative full saturation M is already parallel to H_A and $H_{\rm ex}$. On the other hand, for $H_A = 80$ Oe where the conditions for magnetization switching are optimized, one clearly identifies a strongly damped oscillating feature with an approximately 150 psec rise time and 300 psec period. Figure 3(b) shows the corresponding case for H_A oriented along the hard axis, where the time evolution of magnetization component along the applied field [$\Delta M_Y(t)$] is probed. The oscillatory nature of $\theta_K'(t)$ is more accentuated with a period of 280 psec. The distinct time dependent feature, for which we found no counterpart in the control samples, give direct insight into the magnetization dynamics that are triggered by the photomodulation of the exchange coupling.

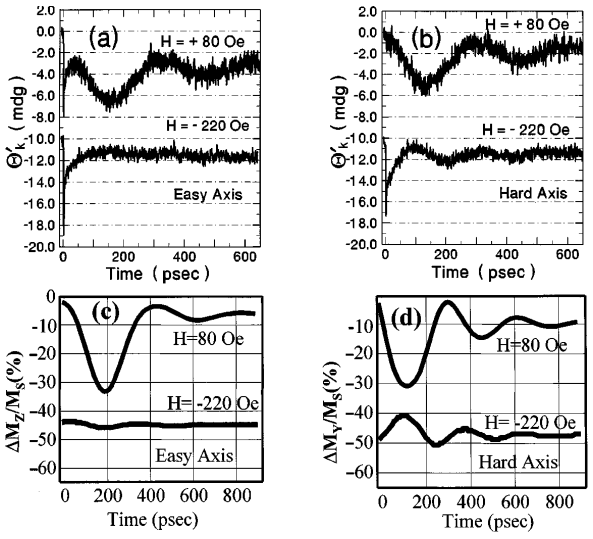

FIG. 3. Time-resolved transient Kerr effect $\theta_K'(t)$ for the photoexcited NiFe/NiO bilayer in an external field of $H_A = 80$ Oe, and $H_A = -220$ Oe. (a) applied along the easy axis, and (b) along the hard axis. Traces (c) and (d) show the results of a model calculation described in the text.

For a physical description of a possible optically induced magnetization reversal, we first consider the energetics of the coupled NiFe/NiO system by a "mesoscopic" two-level model, used recently to describe the stability of exchange bias against thermal fluctuations [10]. In this model, which recognizes the importance of the surface morphology and (columnar) microstructure of the AF medium (NiO) [11], an ensemble of independent single-domain AF grains (typical size of 10 nm) interacts with the FM layer. The energy per unit area for the coupled system is written as [10,12]

$$E = 2(AK)^{1/2}[1 - \cos(\theta - \psi)] + (K_I + K_U t_f)\sin^2\theta + K_{\rm AF} t_{\rm AF}\sin^2\psi - H_A M_S t_f \cos\theta\,, \quad (1)$$

where the first term is the energy associated with the exchange bias field $H_{\rm ex} = 2(AK)^{1/2}/M_S t_F$; the second term represents the uniaxial anisotropy (K_u of the FM layer and the uniaxial interfacial anisotropy K_I [12]); the third term is the Zeeman energy. The angle between a positive applied field and the (sublattice) magnetization of the FM and AF layer is given by θ and ψ, respectively. Figure 4(a) shows the "configurational coordinate" diagram for the NiFe/NiO bilayer with fixed values of $\psi = 180°$, and $H_A = 80$ Oe (along the easy axis), but with varying $H_{\rm ex}$. The initial magnetization bias is set antiparallel to the applied field (i.e., $\theta = -180°$); hence, by modulating the exchange coupling only, the relative energy for the two stable magnetization states is varied. With a decrease in $H_{\rm ex}$ from 100 to 50 Oe, taken here to be rapidly time varying due to the optically induced modulation, the state for parallel magnetization ($\theta = 0°$) becomes energetically favored over the antiparallel ($\theta = -180°$) configuration. For the parameters appropriate to our case, note how the initial energy barrier disappears, so that magnetization of the FM layer can begin to coherently rotate towards its new, energetically favorable, direction. By contrast, Fig. 4(b) shows how, when initial magnetization is parallel to the external field ($H_A = 200$ Oe), the parallel magnetization remains always energetically favorable. This prediction is consistent with our experimental results, including the qualitative shape of the transient Kerr hysteresis loops. Note that the model is used here to describe nonadiabatic behavior in that all other energy terms in Eq. (1), including those due to magnetocrystalline anisotropy and domain wall structure, are assumed to be frozen on the time scale of interest ($\ll$1 nsec). Finally, Figs. 4(c) and 4(d) show the "configurational" coordinate diagrams for the hard axis case with $H_A = 80$ and 200 Oe, respectively. In both cases, the magnetization vector can rotate towards the direction of its new in-plane energy minimum upon the modulation of the exchange coupling.

We now consider the temporal details of the optically induced magnetization "switching" in Fig. 3. The

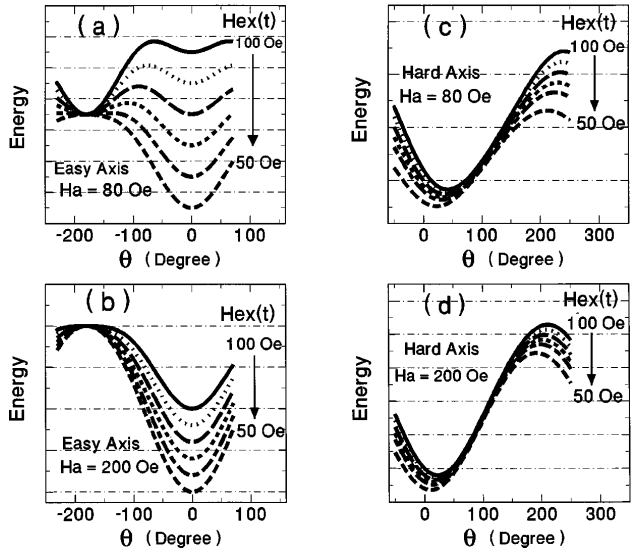

FIG. 4. Energy diagrams for the bilayer system with the exchange bias field $H_{ex}(t)$ modulated between 100 and 50 Oe. The external field of field $H_A = 80$ Oe and $H_A = 200$ Oe is applied along the easy axis in (a) and (b), and along the hard axis in (c) and (d).

temporal shape of the photomodulated exchange bias field $H_{ex}(t)$ consists of a fast rise time due to the unpinning of the exchange coupling by optically excited interfacial spins. Subsequently, $H_{ex}(t)$ returns to its equilibrium state, with a time constant approximately given by the spin-lattice relaxation time. Transient Kerr measurements, performed on quenched NiFe/NiO bilayers and NiFe thin films, suggest that this time constant is on the order of 150 psec at room temperature. If the observed magnetization modulation is due to coherent rotation of local moments defined by each AF grain, the driving term in the relevant equations of motion is provided by $H_{ex}(t)$. On the one hand, as already noted, energetically the disappearance of the barrier for magnetization reversal at $t \sim 1$ psec [Fig. 4(a)] suggests the possibility for a *spontaneous* magnetization reversal at some characteristic frequency of spin reversal υ_0. Presumably, this rate is related to the spin-lattice relaxation time as well [4,5], and could at least partly shape the 150 psec rise time of the $\theta_K'(t)$ signal. With the concurrency recovery of $H_{ex}(T)$, the local magnetization within the grain-sized single domains rotate back. On the other hand, the coherent (macroscopic) magnetization dynamics are generally considered by the Gilbert-Landau-Lifshitz equations of motion in the presence of a time varying magnetic field. We have applied this formalism to test the details of the oscillatory behavior in the $\theta_K'(t)$ traces in Fig. 3(a) and 3(b). The total effective field is taken as $H_T = H_A + H_D + H_K + H_{ex}(t)$, with an applied field H_A, a uniaxial anisotropy field $H_K = (0, 0, H_K \cos\theta)$, and a demagnetization field $H_D = (-4\pi M_X, 0, 0)$ due to the shape anisotropy. The coordinate system is defined similar to that in Ref. [4] such that the thin film lies in the y-z plane, while H_A is applied at angle β to the easy axis. The optically induced modulation of the exchange field is entered as a time dependent driving term of the form $\mathbf{H}_{ex}(t) = \{0, 0, H_{ex}^0[1 - m\exp(-t/\tau_0)]\}$, with the rise time approximated as a step function (<1 psec) and the relaxation time $\tau_0 = 150$ psec corresponding to an empirical spin lattice relaxation time at the FM/AF interface. The results of the calculations for the dynamics of coherent magnetization rotation $\Delta M_{Y,Z}(t)/M_S$ are shown in Fig. 3(c) and 3(d) for a modulation depth $m = 0.6$ and $H_A = 80$ Oe, $H_A = -220$ Oe applied along the easy and hard axes, respectively. A satisfactory fit is obtained for the overall damped oscillatory behavior $\alpha \sim 0.05$, entered as a parameter. The damping is higher than reported for permalloy films [3] but involves the coupled interface system. The model supports semiquantitatively the picture in which the photoinduced modulation of H_{ex} provides the system the possibility for spontaneous magnetization reversal from a simple energy argument, but where the temporal details of the magnetization dynamics are governed by driven equations of motion, with $H_{ex}(t)$ as the time dependent "power supply." The microstructure at the NiFe/NiO interface defines a key spatial scale, bridging between the atomic scale associated with the optically induced spin excitations and the macroscopic magnetization which is measured via $\theta_K'(t)$ in our experiments.

In summary, we have demonstrated an ultrafast, optically modulated magnetization response in a FM thin film, unidirectionally exchange coupled to an AF layer. The approach provides a possibility for very high speed control and coherent switching of magnetization through microscopic access to spins in an exchange coupled system.

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