Coherent Magnetization Rotation and Phase Control by Ultrashort Optical Pulses in CrO₂ Thin Films

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We have applied photoexcitation by ultrashort laser pulses to single crystal thin CrO₂ films to trigger coherent transient magnetization rotation on a subnanosecond time scale, in macroscale single domains. Moreover, by applying the photoexcitation by pairs of temporally separated pump pulses, the transient precession of the magnetization can be phase controlled, depending on the time separation between the pulses. The mechanism behind the photoexcitation originates from the modulation of the magneto-crystalline anisotropy by nonthermal hot electron spins.

The use of photons to excite spin degrees of freedom is an old subject in atomic and semiconductor physics. In semiconductors, where band structure allows a coupling to spins by photons via interband transitions, optical techniques have been used recently to create transient populations of nonequilibrium spins for study and control of their transport, phase, and entanglement [1]. By contrast, little such work exists in ferromagnetically ordered metals with long-range order in the ground state of the many-electron system. Optics is used in these systems as a probe of the magnetization vector **M** via the magneto-optical Kerr effect, but there are few examples of light-induced nonequilibrium phenomena [2]. In fact, magneto-optical disk storage uses laser light simply as a means of lattice heating thin ferromagnetic films above the Curie temperature [3]. Recently, the use of ultrashort laser pulses to provide intense photoexcitation has led to demonstration of nonthermal quenching of the magnetic order on a picosecond time scale in thin Ni films [4] as well as transient magnetization oscillations in an exchange-coupled NiFe/NiO bilayer [5] and spin waves in ferromagnetic Ni thin film [6]. Likewise, such techniques were recently used to study spin dynamics in a half-metallic ferromagnet Sr₂FeMoO₆ in connection of optically induced magnetic phase transition [7].

In this Letter, we demonstrate that it is possible to employ ultrashort, pulsed laser excitation of a half-metallic, ferromagnetic CrO₂ thin film, to directly trigger coherent transient magnetization rotation on a subnano-second time scale, where the macroscopic photoexcited volume acts as a single precessing magnetic domain. To highlight such optical control in CrO₂ films further, we show below that, by applying the photoexcitation in pairs of time sequenced pump pulses, the phase of the transient precession of the magnetization vector can be manipu-lated, constructively or destructively, depending on the temporal separation between the impulses of excitation. As argued below, we evoke for the physics of the "photo-trigger" mechanism the modulation of the magnetocrys-talline anisotropy by hot electrons, this mechanism opening the door to modulation of the collective, long-range magnetic order via one-electron/spin excitation by photons in half-metallic systems in general.

We employed single crystal films of CrO₂ which were grown epitaxially on TiO2 single crystal substrates by chemical vapor deposition [8]. A half-metallic ferromag-net, CrO₂ has an energy gap in its density of states for the minority spin electrons, reported from optical measure-ments to be ≈ 2 eV [9]. Large spin polarization (∼ 96%) in CrO₂ single crystal films has been demonstrated [10], and studies show how static magnetization reversal occurs by coherent rotation with the entire sample acting as a single domain [11]. We chose a photon energy of excitation below this gap in order to optically excite the majority spin electrons only. This is a unique feature of the half-metallic systems and we show next how ultrashort pulse optical excitation leads to a perturbation of the ferromag-netic state, resulting in coherent rotation of the magneti-zation vector **M**(t) with a rotation amplitude much larger than can be expected in other ferromagnetic materials, while preserving its magnitude.

The crystal structure of CrO₂ is tetragonal and for our (100) oriented CrO₂ single crystal films the b and c axes of the unit cell lie in the film plane, with the uniaxial magnetocrystalline anisotropy energy dominated by $\kappa_1 = \sim 2.7 \times 10^5$ erg/cm³ and its corresponding anisot-ropy field $H_K = 2\kappa_1/M_S = 800$ Oe. Typical saturation magnetization is ~ 600 emu/cm³ with the easy axis the magnetization showing nearly 100% remanence, a measured Kerr rotation of $\theta_K = 12$ mdegrees, switching

sharply at fields typically ~ 200 Oe in the approximately 3000 Å thick films. In our experiments excitation pulses from a mode-locked Ti:sapphire laser ($\tau_p = 120$ fsec, $h\nu = 1.4$ eV, or $\lambda = 825$ nm, pulse energy 2.1 nJ) were directed normal to the sample and absorbed within the CrO_2 layer. To reduce average lattice heating, we used a pulse repetition rate of 1.9 MHz for an average power of approximately 4 mW focused to a spot of about 20 μm. The optically induced changes in the magnetization of the samples were extracted from the measurement of transient changes in the (longitudinal) Kerr effect in the CrO_2 film, labeled below as $\theta'_K(t)$, by using time delayed weak (< 0.2 mW) probe pulses also in the near infrared ($h\nu = 1.4$ eV). The Kerr instrumentation employed a polarization-sensitive optical bridge and a low-noise differential detector [12].

The time-resolved longitudinal Kerr effect $\theta'_K(t)$ probes the off diagonal component of the conductivity tensor σ_{xy}, which is proportional to net spin polarization and the magnetization. Two distinct contributions are found in such pump-probe experiments, distinguished by their different time constants and field dependence [5,13]. First, following the subpsec impulse of photoexcitation, the hot electron and spin distributions are displaced from thermal equilibrium, leading to modulation in θ_K because of single particle spin occupancy effects and modulation in the net spin polarization. Recent studies of these processes in thin Ni, Co, and CoPt FM films have given information about the dynamics of spin relaxation [4,13]. The proportionality of the longitudinal Kerr effect to the in-plane component of magnetization makes the transient experiment also sensitive to photoinduced changes in the *direction* of $\mathbf{M}(t)$ due to its coherent rotation. As shown next, the one-electron occupancy effects lead to a slow (~ 100 psec) featureless decay of the transient Kerr traces, atop which well defined oscillations appear, signaling the occurrence of coherent magnetization rotation. The slowly decaying background in $\theta'_K(t)$ is largely insensitive to external magnetic field, for example, and is easily subtracted to highlight the contribution of interest, i.e., the Kerr signal proportional to $\mathbf{M}(t)$.

Figure 1 compares the transient Kerr and transient reflectivity $\Delta R(t)$ signals at room temperature, under an applied magnetic field of $H = 500$ Oe along the hard-axis direction of a CrO_2 film. The presence of the oscillations in the Kerr trace provides an immediate clue to the presence of coherent magnetization rotation, further articulated by the magnetic field's orientational dependence as discussed next. In Fig. 1(a), the vertical axis contains a projection of the time dependent magnetization $\mathbf{M}(t)$ along the c axis of the unit cell, following photoexcitation at $t = 0$ psec. The featureless transient reflectivity signals such as in Fig. 1(b), originating from the diagonal elements of the dielectric tensor, are seen to be independent of the magnetic field. They are dominated by a slow decay which, apart from an initial fast hot
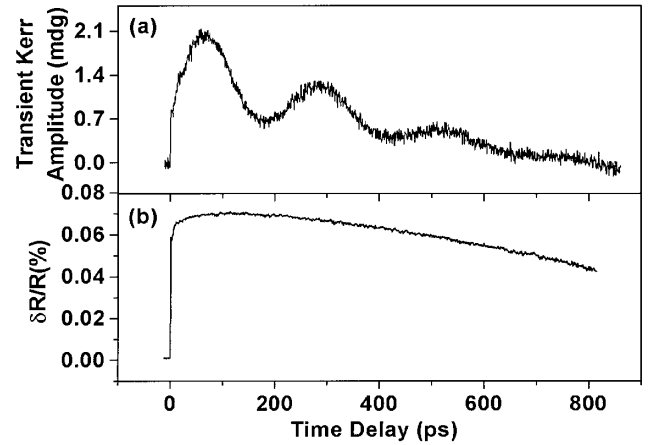

FIG. 1. (a) Transient Kerr signal and (b) transient reflectivity of a 3000 Å thick CrO_2 thin film at room temperature and in an external magnetic field of $H = 500$ Oe applied in the hard-axis direction.

electron decay (electron-phonon relaxation on a psec time scale), involves phonon thermalization and heat conduction to the substrate, respectively [14]. The estimated maximum lattice temperature increase following each excitation pulse is below $30\,°C$, compared with the Curie temperature of CrO_2 of $T_C = 100\,°C$. The slowly varying background in $\theta'_K(t)$, in turn, involves a composite of relaxation processes including the spin-lattice relaxation time and diffusive processes across the film.

After subtracting the single particle background in $\theta'_K(t)$, we investigated the details of the optically triggered coherent transient magnetization rotation, as shown in Fig. 2(a), where the dependence of the measured oscillations on the orientation of the externally applied field relative to the hard axis is shown, with $\theta = 0°$ indicating the B field along the hard axis. In this example, a field of $B = 500$ Oe was chosen. The amplitude of the oscillations, their frequency, and damping rate depend on the orientation of the external field. As one moves away from the hard axis, the effect decreases. We have found it
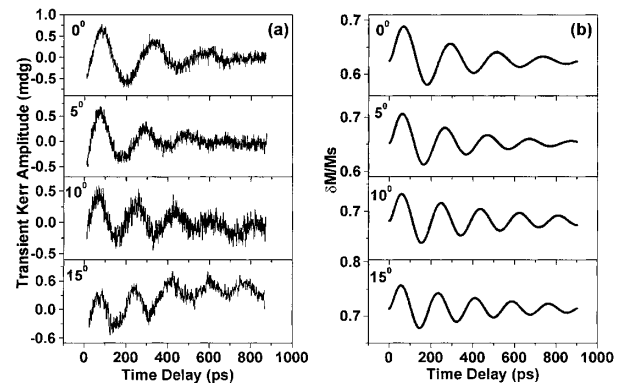

FIG. 2. (a) Transient Kerr signal as a function of the angle θ of the applied field (here $H = 500$ Oe) and (b) results of calculation according to the coherent rotation model discussed in the text.

difficult to detect oscillations for $\theta > 45°$ given the signal-to-noise ratio limit in our experiments of about $\theta'_K(t) \sim 0.1$ mdeg. The oscillations can be easily seen over a range of externally applied fields, ranging roughly from 100 to 1000 Oe, but the example shown represents a near optimal case for maximizing their amplitude. Note that the damping rate decreases as the externally applied field moves away from the hard axis towards the easy axis. Similar anisotropy of the intrinsic damping of the magnetization was also found in Ni [15] and Co [16] thin films via ferromagnetic resonance (FMR). Here, the effect could be due to underlying magnetocrystalline anisotropy or thin film shape effects and needs to be studied further. Figure 2(b) shows the results of a model simulation, based on the Landau-Lifshitz-Gilbert (LLG) gyroscopic equations of motion, to which we return below.

As a means of further verifying and exploiting the transient coherent magnetization rotation phenomena, we applied a sequence of two equal amplitude pulses of pump excitation to the thin CrO_2 films. The time delay between the pulses was varied continuously to study the impact of two consecutive trigger events through the transient Kerr effect. Figure 3(a) shows the result obtained for three different time delays between the two pump pulses, referenced to the case of a single pump pulse only (top trace), with an external magnetic field of $H = 500$ Oe applied in the hard-axis direction. The traces demonstrate how the precession of $\mathbf{M}(t)$ is interrupted by the second pump pulse, the effect ranging from nearly destructive ($\tau \approx 140$ psec) to constructive interference ($\tau \approx 230$ psec). We refer to this ability to impulsively influence the phase and amplitude of the magnetization rotation as "coherent control" by the ultrashort optical pulses. In this work we have not yet "fine-tuned" the conditions for full constructive/destructive coherence control; the model discussed next shows that this is possible for specific values of the ratio of the pulse energies of the two excitation pulses.
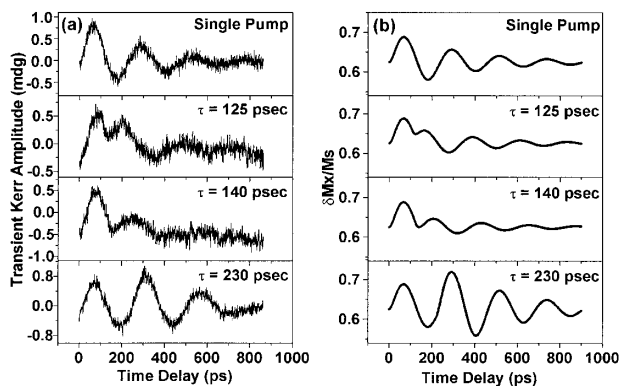

FIG. 3. (a) Transient Kerr signal in the presence of two pulses of excitation, whose time separation is indicated (top trace corresponds to a single pulse only). The magnetic field of 500 Oe is applied in the hard-axis direction. (b) Result of the model calculation.

Figure 3(b) shows the results of a simulation which is based on the LLG equations of motion for $\mathbf{M}(t)$ [17], adapted for the case of our photoexcited CrO_2 thin films. This approach to modeling was noted in connection of Fig. 2(b). We apply the formalism by inserting a total effective field into the LLG equations of the form $\mathbf{H}_T(t) = \mathbf{H}_A + \mathbf{H}_D + \mathbf{H}_K(t)$. In the geometry of choice, the static applied field has the components $\mathbf{H}_A = (H_A \cos\beta, H_A \sin\beta, 0)$, a uniaxial time varying anisotropy field $\mathbf{H}_K(t) = [0, H_K(t)\cos\theta, 0]$, and an effective demagnetization field $\mathbf{H}_D$ due to the shape anisotropy of the thin FM film. The thin film lies in the x-y plane and the easy c axis of the unit cell is taken to be along the y direction, with θ as the angle between $\mathbf{M}(t)$ and easy axis, while H_A is applied at an angle β with respect to the hard axis. The optically induced modulation of the anisotropy field, whose physics is discussed below, is entered as a time dependent single exponential driving term with a modulation depth m of the form $\mathbf{H}_K(t) = H_K^0[1 - m\exp(-t/t_0)]$. The rise time of $H_K(t)$ is approximated as a step function and the relaxation time $t_o \approx 15$ psec corresponds roughly to the empirical electron-lattice relaxation time in CrO_2 extracted from the rise time of the oscillatory portion of the transient Kerr traces in our experiments. Spin-lattice relaxation times of this order of magnitude have been obtained from time-resolved measurements on other ferromagnetic thin films using effective temperature models to decode the relaxation processes that involve the spin, electronic, and lattice degrees of freedom [2,13].

The calculated traces in Figs. 2(b) and 3(b) were both obtained by inputting the known values of M_S and κ_1, while $m = 0.3$ and the dimensionless damping coefficient $\alpha = \sim 0.04$–0.06 in the LLG equations reflect values for these two adjustable parameters for a best fit. As the level of photoexcitation is estimated to create a nonequilibrium spin density of approximately 10^{20} cm^{-3}, the value of m seems reasonable. Recently, FMR experiments have been conducted in single crystal thin CrO_2 films, yielding a value as low as $\alpha = 0.0023$ from linewidth studies [18]. However, we note that the physical situation in our case differs from that in a typical FMR experiment since the initial tipping angle of $\mathbf{M}(t)$ from equilibrium here is fairly large ($> 5°$), and we field bias the film in near the hard-axis direction. However, the subject clearly needs further exploration for identifying the microscopic spin deexcitation channels involved in the damping of the coherent rotation of $\mathbf{M}(t)$. We note that significantly large tipping angles or even irreversible magnetization reversal are possible in materials with higher magnetocrystalline anisotropy and at higher optical excitation levels.

We now outline physical arguments for the photoexcitation pathway, which hinge crucially on the modulation of the magnetocrystalline anisotropy field. We note that theory developed for the single ion magnetic anisotropy constants from general thermodynamic considerations [19] shows how the anisotropy constants $\kappa_2(T)$ and

$\kappa_4(T)$ [and hence $\kappa_1(T) = a\kappa_2(T) + b\kappa_4(T)$] decrease much more rapidly than $|\mathbf{M}(T)|$ with the normalized temperature T/T_C. In the effective temperature picture of the transient experiment, the spin, electronic (orbital), and lattice temperatures are subject to different values [2,4,13]. Excitation by photons below the minority spin gap in a half-metallic system implies that the effective spin temperature T_s remains initially nearly constant, even if the effective electron temperature is high, that is, $T_e > T_s$. That is, the spin degree of freedom is partially isolated from the other degrees of freedom and more so than in a conventional metallic ferromagnet, because spin-flip scattering is substantially suppressed by the gap in the minority spin density of states. Since the anisotropy field originates from the spin-orbit coupling, and the typical spin-orbit coupling in a CrO_2-like case is on the order of 0.1 eV, it can thus be overcome rather easily at modest increase of the effective temperature T_e of the orbital degrees of freedom. That is, $\kappa_1(T_e)$ is subject to change, even while the magnitude of the magnetization vector $|\mathbf{M}(T_s)|$ remains largely unchanged. This supposition is supported by the fact that there is no evidence of thermal quenching of magnetic moment in any of our experiments.

In this Letter, we have emphasized the optical generation of transient, *reversible* magnetization changes which are most readily observed near the hard-axis configuration in the half-metallic CrO_2 thin films. A simple configurational coordinate energy argument [12], constructed by evaluating the total magnetic energy, shows why in this applied field orientation the system is readily susceptible to coherent magnetization rotation by the time varying anisotropy field $\mathbf{H}_K(t) \sim \mathbf{H}_A$ since there are no energetic barriers to overcome, unlike in the easy-axis case (with the two equivalent energy minima). However, if one were to look for the possibility of using the optical approach employed here to induce *irreversible* magnetization switching/reversal, an external field bias near the easy axis pointing in the opposite direction to the magnetization would be preferred, with a value approaching that of the static switching field. For a repetitive optical excitation pulse train, such as employed in our experiments, an additional pulsed magnetic field would be required, to "reset" the magnetization after each event.

Note added.—While this Letter was under review, optically induced coherent spin waves in Ni and permalloy have been reported by van Kampen [20]. These experiments do not utilize the unique aspects of half-metallic systems, which is the subject of the present work.

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