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Phys. Rev. Lett. **107**, 207205 — Published 11 November 2011

DOI: 10.1103/PhysRevLett.107.207205

Coherent spin precession via photo-induced antiferromagnetic interactions in $La_{0.67}Ca_{0.33}MnO_3$

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Abstract

Pronounced spin precessions are observed in a geometry with negligible canting of the magnetization in ferromagnetic $La_{0.67}Ca_{0.33}MnO_3$ thin films using the time-resolved magneto-optical Kerr effect. The precession amplitude monotonically decreases with increasing field, indicating that the coherent spin rotation may be triggered by a transient exchange field and not by demagnetization and/or anisotropy field modulation. We attribute the transient exchange field to emergent antiferromagnetic interactions due to charge transfer and modification of the kinetic energy of e_g electrons under optical excitation.

PACS numbers: 75.78.Jp, 75.30.Et, 75.47.Lx, 76.70.Hb

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Control of coherent spin rotation in ferromagnets received intense investigations in the past decade for fast magnetization switching in novel magnetic recording and spintronic devices [1, 2]. Recently, optical excitations of coherent spin precessions have been demonstrated in various ferromagnetic (FM) metals, providing a new approach for fast spin manipulation. The precessions are triggered by effective field pulses generated via fast demagnetization [3, 4], or transient modulation of anisotropy constants [5, 6, 7]. The demagnetization occurs due to fast dissipation of spin angular momentum via efficient spin lattice thermalization [8], whereas the anisotropy constant modulation may be caused by the alteration of spin-orbit interactions due to hot electrons and electron phonon thermalization [5], creation of itinerant carriers [6], or coherent acoustic phonons [7].

Despite the different excitation mechanisms, both scenarios require canting of the magnetization, *i.e.,* an angle between magnetization and the external applied magnetic field to induce the coherent rotation. This geometry is also essential for spin precession in exchange biased magnetic structures [9]. In such systems, the instant laser heating destroys the exchange induced pinning of the FM spins at the interface, and thereby triggers the rotation of the magnetization to the new direction. Djordjevic *et al.* recently proposed a new excitation mechanism, where localized spin flip may decay into short wavelength spin wave excitations and then transfer to low energy spin wave modes [10]. The canted geometry also may not be important for ferrimagnetic materials in which the spin precession is excited via inverse Faraday effect using circular polarized light [11, 12].

In this letter, we report observations of pronounced spin precessions in a geometry with negligible canting of the magnetization in FM $La_{0.67}Ca_{0.33}MnO_3$ (LCMO) thin films on $NdGaO_3$ (NGO) substrates. Using time-resolved magneto-

optical Kerr effect (TR-MOKE) measurements, we observed large spin precessions over a wide range of magnetic fields (0-2.5 T) applied along the in-plane easy axis. The precession amplitude monotonically decreases with increasing field, in contrast to the precession induced by transient anisotropy field in the state of canted magnetization. These observations indicate that the coherent spin rotation may be triggered by a transient exchange interaction and not by demagnetization and/or anisotropy field modulation. The picosecond modulation of the exchange interaction results from the emergence of antiferromagnetic interaction caused by charge transfer and alteration of kinetic energy of e_g electrons in LCMO under optical excitation.

The LCMO films are epitaxially grown by pulsed-laser deposition on NGO substrates. The films exhibit an in-plane uniaxial anisotropy of ~0.2 T at thickness of 60-100 nm [13]. TR-MOKE measurements are performed with a Ti:sapphire amplifier laser system providing 150-fs pulses at 1-kHz repetition rate. We use 800-nm pump pulses with fluence of ~1 mJ/cm^2 to excite the magnetization in our samples, and the time evolution of the magnetization is monitored with 400-nm probe pulses, where the Kerr rotation is at a maximum [14], with fluences on the order of 0.1 mJ/cm^2. The transient Kerr rotation $\Delta\theta$ is measured in a cross polarization configuration. Rotation of the analyzer through the extinction angle leads to opposite sign of $\Delta\theta$. For transient reflectivity (ΔR) measurements we use the same pump and probe wavelengths as in TR-MOKE.

Figure 1(a) shows the time evolution of ΔR from a 100-nm thick LCMO film at 25 K. ΔR exhibits a fast response with a rise time comparable to the laser pulse width and a slow response with decay time of ~100 ps. The latter component is associated with the slow spin-lattice relaxation [15].

The time evolution of $\Delta\theta$ is shown in Fig. 1(b). Here, the external field H is applied perpendicular to the sample plane and the probe beam is incident at an angle of ~5 degrees with respect to the sample normal, permitting sensitive detection of the out-of-plane magnetization component M_z. In contrast to the fast electronic response, the magnetic response shows no instantaneous change on the timescale of 1ps. The major feature of the magnetic response manifests as strong oscillations which correspond to the coherent precessions of the ferromagnetic spins. We therefore conclude that no significant demagnetization occurs before the magnetization rotation is launched. This finding is consistent with the observations of negligible demagnetization in ferromagnetic $La_{0.6}Sr_{0.4}MnO_3$ films at temperatures significantly lower than its Curie temperature [14]. The weak demagnetization is due to the half metallic nature in manganites where the Elliot-Yafet scattering is blocked [16].

As a comparison, Fig. 2(a) shows the laser induced magnetization dynamics in an Fe film at an external field of 550 Oe applied along the in-plane hard axis at room temperature (RT). The transient Kerr rotation clearly shows a fast response which occurs within 1ps. This response originates from the fast demagnetization due to the spin flip process associated with the spin lattice interactions [8]. The demagnetization decays within ~50 ps, while the subsequent magnetization precessions persist for much longer time (>500 ps) [17]. The excitation of the magnetization precessions in the Fe film can be reasonably explained by the modulation of the anisotropy fields caused by the demagnetization [3, 4]. Under the laser interaction, the fast demagnetization modifies the magneto-crystalline anisotropy, generating a transient effective field H^{tr} perpendicular to the static effective field H_{eff}, and thus the magnetization precessions are triggered.

Hence, the precession is significantly weaker and difficult to observe if the external field is applied nearly close to the easy axes along which the magnetization is pinned, as shown in Fig. 2(b). These observations clearly indicate that the canting of the magnetization and fast demagnetization are of crucial importance in exciting spin precession in the Fe film, typical for 3d transition ferromagnetic metals.

In contrast, Fig. 1(b) shows that fast and large demagnetization is not present in the spin precession dynamics of LCMO films. In the following, we will present further measurements of the magnetization dynamics when H is applied along the uniaxial easy axis in the film plane, i.e., a configuration with negligible canting of magnetization. The results are shown in Fig. 1(c). Similar to the case where the magnetization is pulled out of plane, no fast demagnetization occurs when spins are aligned within the film plane, while pronounced spin precession is present over a wide range of magnetic fields. Particularly, a large precession is also excited at zero field and persists for over 1 ns, as shown in Fig. 1(d). We note beating patterns in the transient Kerr spectra at zero field. Fourier transform reveals two spin waves modes at frequencies of ~11 GHz and ~15GHz (Fig. 1(e)). The mode with lower frequency corresponds to the Kittel mode, as the detailed analysis of the magnetic field dependence of the precession frequency shows [13]. The higher-frequency mode may correspond to a surface Damon-Eshbach-type mode or a higher-order standing spin wave mode within the ferromagnetic film [18, 19]. The slow decay of the precession indicates that the dephasing process is weak, and thus indicates a well defined easy axis along which all exchange-coupled spins are aligned at zero external field. In this case canting of magnetization does not exist, and therefore modification of the in-plane anisotropy may not be the main contributor to the transient field that launches the spin precession shown in Fig. 1(c).

In the state of canted magnetization, anisotropy modulation will indeed generate a transient field, triggering the spin precession. The anisotropy modulation may arise from the thermal modification of magnetocrystalline energy at elevated electron temperature [5]. Although the modulation of anisotropy field H_a is independent of the applied field H, the transient torque on the magnetization becomes stronger with increasing H as it pulls the magnetization away from H_a, thus leading to larger precession angles at higher fields. When the magnetization is nearly aligned along H at the field strength stronger than H_a, the torque starts to saturate and the precession angle decreases with increasing H. Thus, one can expect that the precession amplitude reaches a maximum at the field approximately equal to the static anisotropy field. The top left panel of Fig. 3(a) shows the field dependence of precession amplitude obtained when H is applied at an angle of 45^o to the easy axis, a state with canted magnetization. We note that the amplitude is sharply enhanced with increasing field and then slowly reduced at fields larger than ~0.3 T, consistent with our expectation. Similar results are obtained from the other LCMO samples, as well as the Fe film with the field applied along the hard axis and the direction at an angle of $\sim 10^o$ to the easiest axis (Fig. 3(b)). The modulation of the magnetic in-plane anisotropy in an oxide ferromagnetic material CrO_2 also leads to similar dependence of precession amplitude on H applied along the hard axis, and no precession is detected if H is oriented close to the easy axis [5, 20].

In contrast, the precession with the applied field along the easy axis of LCMO shows the opposite dependence, i.e., the amplitude decreases with increasing field for all three LCMO samples, as shown in the right panels of Fig 3(a). This result confirms our previous conclusion that the transient anisotropy field, either due to demagnetization or modification of anisotropy constant, is not the major contributor

to launch the spin precession when H is applied parallel to the easy axis. We thus propose a transient exchange field H_{ex}^{tr} that triggers the spin precession.

In order to obtain the field dependence of H_{ex}^{tr}, we simulate the precession of the magnetization and follow its out-of-plane component [21]. In the simulation, the magnitude of H_{ex}^{tr} is adjusted so that the calculated spin precession angle coincides with the nominal precession angle obtained from the measured precession amplitude (Fig. 3(a)). We thus determine the field dependence of the magnitude of H_{ex}^{tr} for the 60-nm thick film, as shown in Fig. 4(a). We note that H_{ex}^{tr} is reduced with increasing applied field. This dependency is a characteristic of the transient exchange field in manganites as discussed below. In contrast, the transient anisotropy field H_{a}^{tr}, which is perpendicular to the magnetization, due to demagnetization or reduction of the anisotropy constants is enhanced with increasing field. We present a calculated result in Fig. 4(b), which corresponds to H_{a}^{tr} in a configuration with slight canting of magnetization, *i.e.,* an angle of 5 degrees between the applied field H and the uniaxial anisotropy field H_u, and 10% reduction of H_u (0.2 T) after laser interaction [22].

The transient exchange field may result from the modification of the exchange coupling because the optical excitation may change the balance between the FM metallic and antiferromagnetic (AF) insulating states in the film. The coexistence of distinct FM metallic and AF insulating phases in doped manganites has long been established [23-25], and the AF domain may be melted in the presence of an external field. In the case of LCMO, several reports indicated the presence of the AF phase down to the lowest temperatures [26-28]. Very slow photoinduced drop in conductivity and demagnetization have been observed in LCMO and interpreted as the photoinduced shift in the FM/AF phase balance triggered by the photoexcited

carriers that promote the cooperative Jahn-Teller distortions and formation of AF insulating clusters [29]. An ultrafast drop in conductivity upon photoexcitation with femtosecond pulses was observed by Averitt *et al*., which is also consistent with the promotion of antiferromagnetic correlations [15].

We speculate a formation of nanometer- to sub-micrometer-scale regions with transient AF interaction after the intense optical excitation, similar to the size of AF domains observed in quasi-static phase separated manganites [23-26]. Since the AF regions are randomly distributed in LCMO films, we would expect a spatial inhomogeneity of the transient exchange field which may lead to excitations of the Damon Eshbach mode or the standing spin waves [18, 19]. Because application of the magnetic field may suppresses the AF interaction, the magnitude of H_{ex}^{tr} is reduced at higher fields as shown in Fig. 4(a).

The reorientation of the spins to form AF clusters may take a long time due to slow spin-lattice thermalization process, a general feature in manganites [30], to dissipate spin angular momentum. However, the exchange interaction may be modified in a much faster time regime. This is because the local carrier concentration and its kinetic energy play a crucial role in determining the magnetic phases [31, 32]. Both photoinduced charge transfer between neighboring Mn ions [33, 34] and the thermalized electrons may suppress the double exchange interaction and promote the AF coupling. Therefore, the exchange field pulse may be generated within electron phonon relaxation time, much faster than the emergence of the demagnetization caused by real formation of AF clusters.

This finding is corroborated by the temperature dependence of the magnetization dynamics shown in Fig. 5. At temperature lower than 150 K, the magnetization dynamics show negligible demagnetization. With increasing

temperature, a slow demagnetization (>500 ps) is clearly observable. However, the spin precession is launched nearly at the same time at all temperatures lower than the Curie temperature T_c (~270 K). The reduction of the precession amplitude at higher temperatures is mainly caused by the decrease of the saturated magnetization. The reduction of the magnetization also results in a pronounced decrease of precession frequency at temperatures approaching to T_c, as shown in the inset of Fig 5. This result therefore supports our model that the precession is triggered by the transient exchange field in the absence of spin lattice thermalization.

A recent study reveals a transient anisotropy field due to the optical induced transition between easy in-plane and easy out-of-plane anisotropy states in $La_{2-2x}Sr_{1+2x}Mn_2O_7$ bilayered manganites [6]. However, we point out that the $La_{0.67}Ca_{0.33}MnO_3$ film is not located at the boundary of two ferromagnetic phases with different anisotropies, and therefore no transient field due to the emerging of anisotropy field along different directions is expected to be present in our LCMO films. Such a transient field also would be independent on the external field, different from H_{ex}^{tr} which we obtain from the simulation of the field dependence of the precession amplitude as shown in Fig. 4(a).

In summary, we have observed pronounced photoinduced magnetization precessions in $La_{0.67}Ca_{0.33}MnO_3$ films with negligible canting of magnetization. The excitation mechanism is ascribed to a transient modulation of the exchange coupling due to the promotion of AF interaction by photoinjected carriers.

This work was sponsored in part by DOE (Grants No. DE-FG02-04ER46127 and No. DE-FG02-08ER46531), the National Natural Science Foundation of China (Grants No. 60908005 and No. 11074044), Shanghai Pujiang Program, and SRF for ROCS, SEM.

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21. For the simulation, we assume an in-plane transient field perpendicular to the static magnetization with duration time $\tau = 2\,ps$. The out-of-plane precession angle can thus be expressed as $\alpha = \eta\delta\sqrt{2-2\cos f\tau}$, where η is the ratio of the amplitudes

of the precessional magnetization components perpendicular to the film and normal to the magnetization in the film plane, and $\delta = H_{ex}^{tr} / (H + H_u)$ is the angle between the transient effective field direction and the static equilibrium orientation. The uniaxial anisotropy field H_u is obtained from the field dependence of the precession frequency f using the Landau-Lifshitz-Gilbert (LLG) equation.

22. When H deviates a small angle ϕ from H_u in a LCMO film, H_a^{tr} may be expressed as $\Delta H_u \times H \sin(\phi) / (H \cos(\phi) + H_u)$, where ΔH_u is the reduction of H_u due to optical excitation.

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Figure Captions

Fig. 1 (a) Transient reflectivity ΔR, and transient Kerr rotation $\Delta\theta$ of LCMO film with magnetic field H (0.5 T) applied perpendicular to the sample plane (b), with H applied along in-plane easy axis (c), and with zero field (d) at ~20 K; (e) Fourier transform of (d).

Fig. 2 Transient Kerr rotations of Fe film with magnetic field applied along in-plane hard axis (a) and easy axis (b) at RT.

Fig. 3 Amplitude of precession of (a) 60nm and 100-nm LCMO films with field applied at 45^o to easy axis and along easy axis at 20 K, and (b) Fe film with field applied at 55^o to easy axis and of 10^o to easy axis at RT.

Fig. 4 (a) Transient exchange field and (b) transient anisotropy field in a state of canted magnetization (magnetic field at 5^o to the easy axis) of 60-nm thick LCMO film. The dash line is a guide of the data.

Fig. 5 Transient Kerr rotations of LCMO film with applied field H=0.5 T at various temperatures. The inset shows the uniform precession frequency as a function of the temperature.

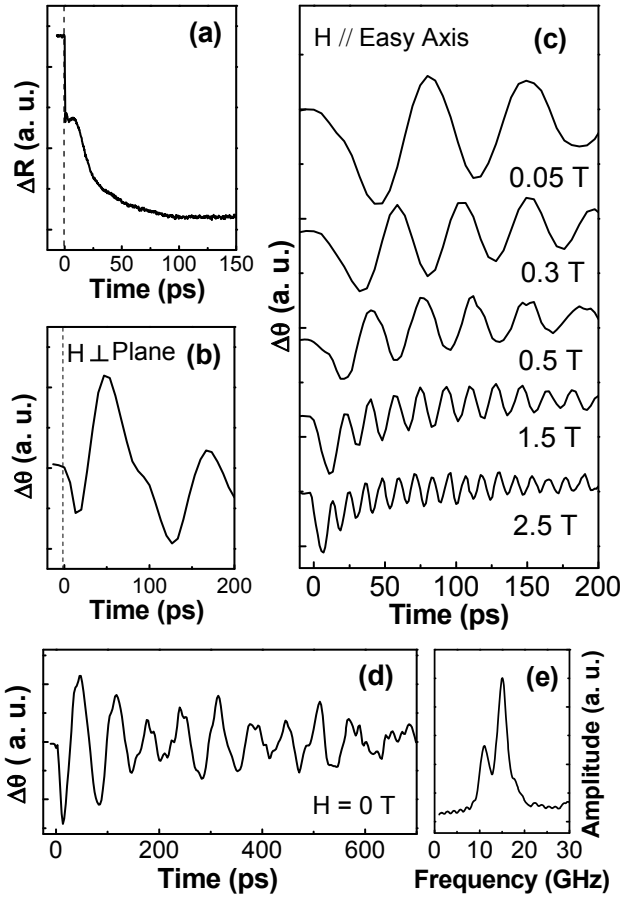
(a)
ΔR (a. u.)
0
50
100
150
Time (ps)
H ⊥ Plane
(b)
Δθ (a. u.)
0
100
200
Time (ps)
H // Easy Axis
(c)
0.05 T
0.3 T
0.5 T
1.5 T
2.5 T
Δθ (a. u.)
0
50
100
150
200
Time (ps)
(d)
Δθ (a. u.)
H = 0 T
0
200
400
600
Time (ps)
(e)
Amplitude (a. u.)
0
10
20
30
Frequency (GHz)

Figure 1

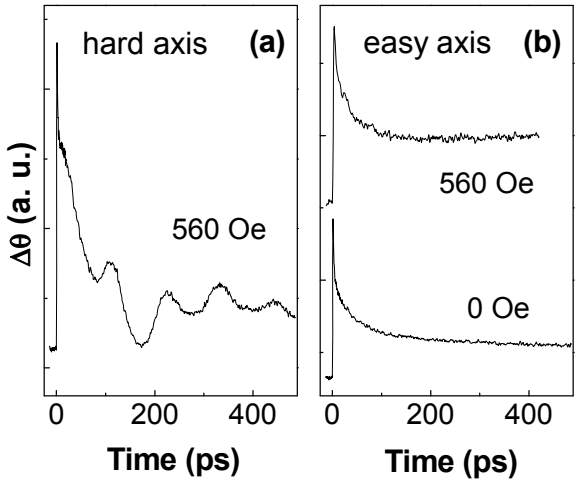
hard axis
(a)
easy axis
(b)
560 Oe
560 Oe
0 Oe
Δθ (a. u.)
0
200
400
Time (ps)
0
200
400
Time (ps)

Figure 2

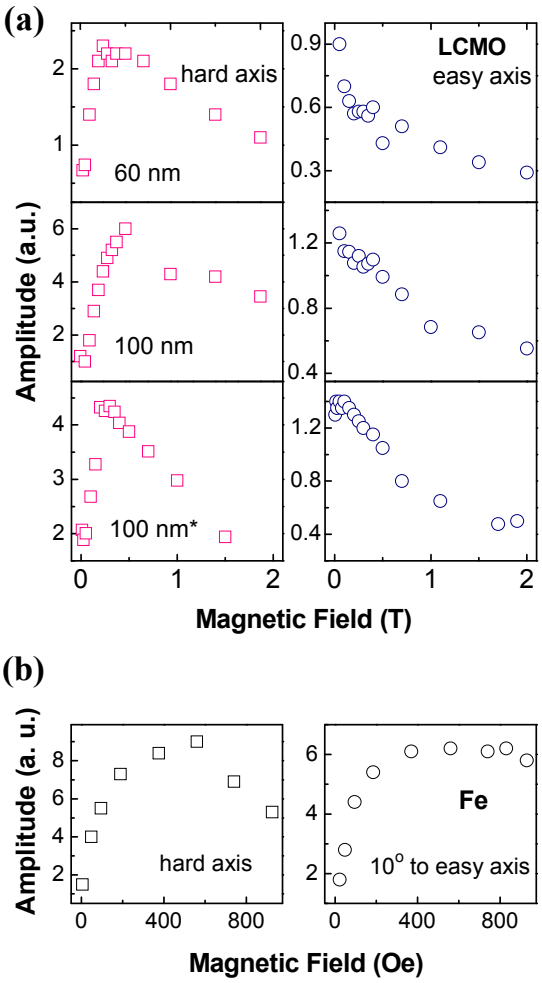
(a)
hard axis
60 nm
100 nm
100 nm*
LCMO
easy axis
Amplitude (a.u.)
Magnetic Field (T)
(b)
hard axis
Fe
10° to easy axis
Amplitude (a. u.)
Magnetic Field (Oe)

Figure 3

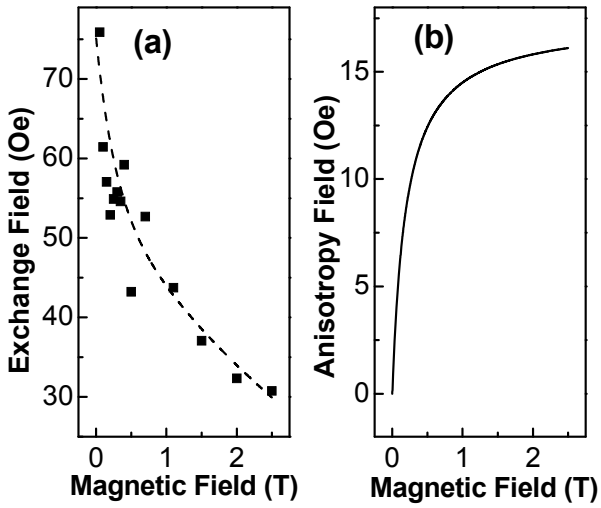
(a)
Exchange Field (Oe)
70
60
50
40
30
0
1
2
Magnetic Field (T)
(b)
Anisotropy Field (Oe)
15
10
5
0
0
1
2
Magnetic Field (T)

Figure 4

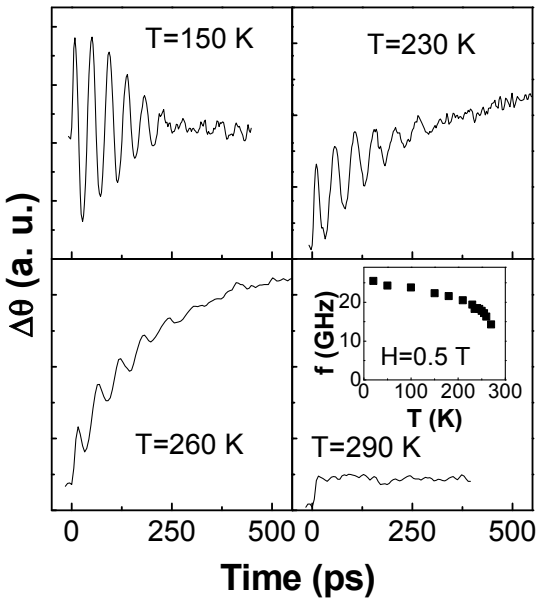

Figure 5