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Photoinduced spin dynamics in $La_{0.6}Sr_{0.4}MnO_3$ observed by time-resolved magneto-optical Kerr spectroscopy

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Photoinduced effect in a ferromagnetic perovskite $La_{0.6}Sr_{0.4}MnO_3$ was investigated by femtosecond pump-probe spectroscopy. By utilizing magneto-optical Kerr rotation as a probe, the photoinduced dynamics of the spins was observed independently of the quasiparticle relaxation dynamics. The low-energy optical conductivity decreases immediately after photoexcitation without changing magnetization, and subsequently the demagnetization occurs in a strongly temperature-dependent manner. The photoinduced demagnetization shows critical slowing down with the time constant up to 1.4 ns when the spin temperature after photoexcitation reaches near the Curie temperature. Such a slow response of a spin system indicates good thermal insulation between quasiparticles and a spin system, suggesting small spin-orbit interaction.

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Ultrafast control of a spin state has been attracting much attention in recent years. Since the conventional control of a spin state by an external magnetic field cannot exceed a precession cycle of the spin moment which is of the order of nanoseconds, an optical mean such as irradiation of a femtosecond laser pulse may be promising for ultrafast manipulation of a spin state. However, an optical field can hardly change a spin state directly, and hence a special mechanism is required for such a control. Investigation for ultrafast control of a spin state has been performed on typical ferromagnetic metals,[1,2] dilute magnetic semiconductors,[3] and so on.[4–6] Colossal magnetoresistive manganites, which show drastic change of resistivity upon the magnetic ordering as a consequence of strong coupling among spin, charge, orbital, and lattice degrees of freedom,[7] are also of great interest for this purpose.

Perovskite manganite $La_{0.6}Sr_{0.4}MnO_3$ shows the ferromagnetic metal phase below $T_C = 370$ K. The origin of ferromagnetism is double-exchange interaction: Because of the strong Hund's-rule coupling between e_g electron spin and t_{2g} electron local spin, carrier e_g electrons gain kinetic energy when localized t_{2g} spins show ferromagnetic alignment. This interaction leads to strong correlation between magnetization and charge transport properties, as well as optical absorption in a low-energy region (<1.0 eV).

Photoinduced effects in femtosecond temporal region for a ferromagnetic phase of perovskite manganites have been previously investigated by conventional pump-probe spectroscopy.[8–12] The first report about photoinduced demagnetization is given by Matsuda *et al.* on $(Nd_{0.5}Sm_{0.5})_{0.6}Sr_{0.4}MnO_3$.[8] They reported that the photoinduced absorption, which shows the rapid change within 1 ps and the following gradual change up to about 200 ps, reflects the photoinduced demagnetization. More detailed studies were reported on $La_{0.7}Ca_{0.3}MnO_3$ by Lobad *et al.*,[9–11] and Averitt *et al.*[12] They attributed the ultrafast component of photoinduced absorption within 100 fs after photoexcitation to electron-lattice thermalization, and the subsequent slower component, the time constant of which changes from 20 ps to 100 ps depending on temperature, to spin-lattice thermalization. In these studies, however, the interpretation and evaluated time scales for the demagnetization were based on the change in optical absorption, and thus the spin dependent parts have not been discriminated in the absorption changes.

In the present work, photoinduced dynamics of magnetization in a single crystal of $La_{0.6}Sr_{0.4}MnO_3$ was investigated by time-resolved magneto-optical Kerr effect spectroscopy, in which magneto-optical Kerr rotation was employed as a reliable probe of magnetization. The photoinduced dynamics of the magnetization was detected independently of the quasiparticle relaxation, which was detected by conventional transient reflection spectroscopy. Namely, one can discriminate the spin dynamics from the charge dynamics by comparing the results of time-resolved magneto-optical Kerr effect and transient reflection spectroscopy. It was found that after a 200-fs pulse photoexcitation the magnetization decreases with an unexpectedly long time constant, which exceeds 1 ns near the ferromagnetic phase transition temperature. Such a slow response of the spin state contrasts with the instantaneous response of the quasiparticles upon photoexcitation.

Magneto-optical Kerr rotation θ and ellipticity η are related to dielectric tensor ϵ via the relation $\theta + i\eta = \epsilon_{xy}/(1-\epsilon_{xx})\sqrt{\epsilon_{xx}}$, in an isotropic material. The off-diagonal element ϵ_{xy} is nearly proportional to the magnetization, and also depends on the oscillator strength of the optical transition. Thus ϵ_{xy} can be a good probe of the magnetization, as far as the electronic structure of the system around the probe photon energy is not changed drastically. Furthermore, θ and η depend not only on ϵ_{xy} but also on the diagonal element ϵ_{xx}. Accordingly, it is to be confirmed that the contribution from change in ϵ_{xx} is small enough to utilize θ and η as an accurate probe of the magnetization.

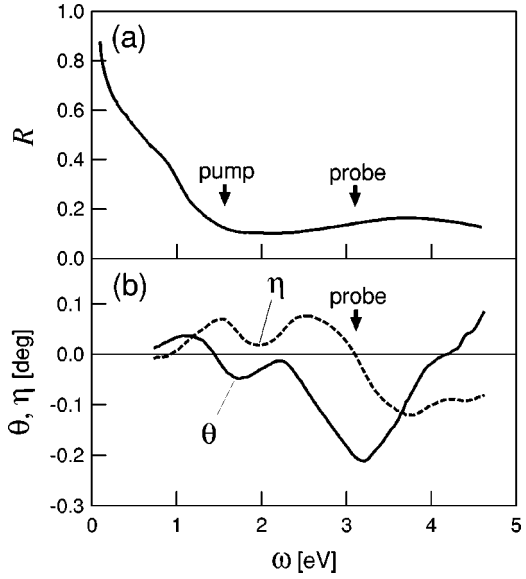

FIG. 1. (a) Reflectivity and (b) magneto-optical Kerr rotation (θ) and ellipticity (η) spectra of $La_{0.6}Sr_{0.4}MnO_3$ at 296 K. The arrows denote the pump and probe energies in time-resolved magneto-optical Kerr spectroscopy.

A bulk single crystal of $La_{0.6}Sr_{0.4}MnO_3$ was grown by a floating zone method. A specimen of typically 5 mm in diameter and 1 mm in thickness was cut from the crystal rod and polished for optical measurement. To remove possible mechanical strain of the surface due to the polishing procedure, the sample was annealed at 1300 K in an oxygen atmosphere.

Figure 1 shows the spectra of reflectivity R and magneto-optical Kerr rotation θ and ellipticity η for $La_{0.6}Sr_{0.4}MnO_3$ at 296 K. The spectral structures in θ and η at 1.5–2 eV are assigned to the plasma edge enhancement, which originates from the diagonal-term contribution $1/(1-\epsilon_{xx})\sqrt{\epsilon_{xx}}$. In fact, the spectrum of off-diagonal term ϵ_{xy} (not shown) does not show any prominent structure in this region. Thus, θ and η in this region should be sensitively affected by small changes in ϵ_{xx}, so that θ or η in this energy region is not suitable as a probe for the magnetization change. On the other hand, the structure around 3 eV is a genuine magneto-optical (off-diagonal term) contribution, which corresponds to the charge-transfer transition from O $2p$ to Mn $3d$ states, and the diagonal-term contribution is small. Therefore, the photon energy of 3.1 eV, which can be easily generated by second harmonics of Ti:sapphire laser light, was adopted in this study for the probe.

The differential reflection $\Delta R/R$ spectra as measured by a conventional pump-probe method are shown in Fig. 2(a). An amplified mode-locked Ti:sapphire laser with the repetition rate of 1 kHz and the central wavelength of 800 nm (1.55 eV) was used for the pump light source. The probe light was generated by the following optical parametric amplifier equipped with second and fourth harmonic generators. The pump and probe pulse duration are less than 200 fs. The pump pulse intensity was $\sim 700\ \mu J/cm^2$, which corresponds to the excitation density of $\sim 10^{20}\ cm^{-3}$ or $\sim 10^{-2}$/Mn site. The spectral feature of $\Delta R/R$ in this energy region (0–3 eV)

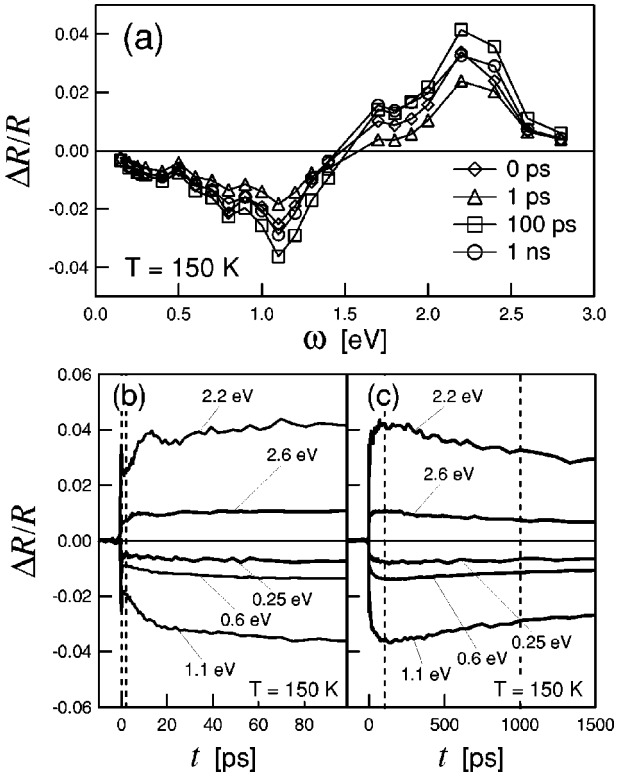

FIG. 2. (a) Differential reflectivity $\Delta R/R$ spectra for various probe delay times. The pump photon energy is 1.55 eV and the pump intensity is $\sim 700\ \mu J/cm^2$. (b), (c) Temporal behavior of $\Delta R/R$ for various probe photon energies as presented in different temporal ranges. Vertical dashed lines denote probe delay times in (a).

appears to be dominated by a shift of the plasma edge to the lower energy, indicating that the system becomes resistive upon photoexcitation. Figures 2(b) and 2(c) show temporal behaviors of $\Delta R/R$ at various probe photon energies as presented in narrow (0–100 ps) and wide (0–1500 ps) temporal ranges, respectively.

In $\Delta R/R$ at the probe energies of 2.2 eV and 2.6 eV, oscillating structures are observed at the delay time of 0–40 ps. They are attributed to the modification of the reflectivity by a shockwave, which is generated by a sudden volume change induced by intense photo-excitation.[13] This interpretation of the oscillation is demonstrated by the fact that the period of the oscillation is almost proportional to the probe wavelength. In the time evolution of $\Delta R/R$, besides the oscillating signal, three characteristic responses are observed in different time domains; a spikelike instantaneous response within the time resolution of 200 fs and its tail within 1 ps, a delayed rise-up signal within the time constant of ~ 10 ps, and a slow decay with the decay time $\tau_r > 1$ ns. Figures 2(b) and 2(c) show that the temporal behavior of $\Delta R/R$ is essentially identical irrespective of the probe energy. It corresponds to the fact that the spectral shape of $\Delta R/R$ is almost the same at all the probe delay times as seen in Fig. 2(a). These results suggest that the transient reflectivity changes in the measured energy region are dominated essentially by a single mechanism, i.e., the plasma-edge shift due to spectral

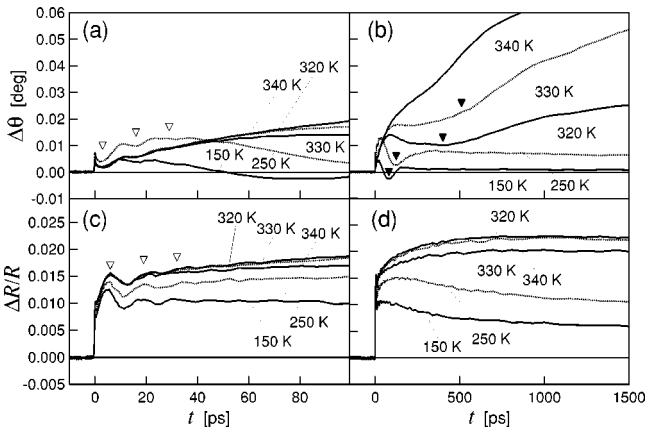

FIG. 3. Temporal behavior of $\Delta\theta$ (a), (b) and $\Delta R/R$ (c), (d) plotted in different scales of temporal range at various temperatures. The pump and probe photon energies were 1.55 eV and 3.1 eV, respectively. The pump intensity was $\sim 500\ \mu J/cm^2$. Open triangles in (a) indicate the oscillation due to the interference between the lights reflected from the surface and the shockwave running normal away from the surface, while closed triangles in (b) indicate the oscillation due to the photoinduced precession of magnetization (see text).

weight transfer from the low-energy (<1.5 eV) to the high-energy region (>1.5 eV).

In the time-resolved magneto-optical Kerr spectroscopy, polarization change of the probe light was detected by a balance detection technique.[6] The sensitivity of change in polarization rotation/ellipticity was as high as about 10^{-3} deg. The pump and probe photon energies were 1.55 eV and 3.1 eV, respectively, which were provided by an amplified mode-locked Ti:sapphire laser and its second harmonics. The pump intensity was $\sim 500\ \mu J/cm^2$. An external magnetic field of ~ 0.25 T was applied normal to the sample surface by a permanent magnet.

Temporal behavior of the photoinduced change in magneto-optical Kerr rotation $\Delta\theta$ is shown in Figs. 3(a) and 3(b). Simultaneously measured $\Delta R/R$ at the same probe photon energy (3.1 eV) is also shown in Figs. 3(c) and 3(d). Since the sign of θ at 3.1 eV in absence of photoexcitation is negative [see Fig. 1(a)], a positive sign of $\Delta\theta$ corresponds to the decrease of the magneto-optical Kerr rotation. The transient increase of the reflectivity at this photon energy is mostly due to the spectral weight transfer from the low-energy region (<1.5 eV) as described above.

The oscillating behaviors with the period of ~ 15 ps observed in both $\Delta\theta$ and $\Delta R/R$ [open triangles in Figs. 3(a) and 3(c)] share the common origin, namely, the interference effect caused by the shockwave, as already discussed in Fig. 2(b). This component does not originate from the change in magnetization but the change in ϵ_{xx}. Another oscillating component observed in $\Delta\theta$ with the period of 100–500 ps [closed triangles in Fig. 3(b)], which has no counterpart in $\Delta R/R$, can be assigned to precession of the magnetization.[14,15] When the magnetization is initially tilted from the external field (normal to the surface) due to magnetic anisotropy, the photoexcitation may change the anisotropy, and accordingly the magnetization starts to precess around the effective magnetic-field direction. The low-energy spectral weight is independent of the direction of the magnetization, and hence $\Delta R/R$ should not be influenced by this precession as observed. The crystal magnetic anisotropy in $La_{0.6}Sr_{0.4}MnO_3$ is quite small,[16] therefore the shape anisotropy near the surface of the sample (with the magnetization tending to be parallel to the surface) may be responsible for this precession behavior. The increasing period of the precession with temperature is ascribed to the reduction in the anisotropy and/or the total moment value.

Besides these oscillating signals, there is a clear difference between $\Delta\theta$ and $\Delta R/R$: $\Delta R/R$ has a steplike response while $\Delta\theta$ does not. Comparing the temporal behaviors of $\Delta\theta$ and $\Delta R/R$, it is noted that the $\Delta R/R$ response is composed of the magnetization-dependent part which shows the same dynamics as $\Delta\theta$, as well as the magnetization-independent one which rises in a steplike manner and decays with the decay time $\tau \sim 1$ ns. In addition, the former strongly depends on temperature, but the latter does not. The low-energy spectral weight as measured by $\Delta R/R$ response decreases within pump pulse duration (<200 fs) after photo-excitation. As there is no energy gap in this system, it is most likely that photo-excited carriers relax to the thermal state within a few picoseconds. Thus it is reasonable to consider that there remains no change in carrier density after this relaxation process. Judging from the absence of the fast response in $\Delta\theta$ within the time domain less than a few picoseconds, we can conclude that the thermalization of quasiparticle excitation will take place without accompanying loss of magnetization.

The decay of the magnetization-independent part corresponds to the cooling process of the quasiparticle system by thermal diffusion. The slower response of the magnetization indicates the good thermal insulation between the quasiparticles and the spins. Although the double-exchange interaction strongly ties the kinetic motion of electrons and the local spin moments, this interaction preserves the total magnetization of the system so that the double-exchange interaction cannot contribute to the demagnetization. It is natural to consider that the observed slow demagnetization process is dominated by the thermalization of the spin-lattice system through spin-lattice interaction. A principal origin of this interaction is the spin-orbit coupling. The time constant of the photoinduced demagnetization observed in the manganite is one or two order of magnitude longer than those of other ferromagnets such as Ni and Sr_2FeMoO_6 previously reported.[6,14,17] It suggests that the spin-orbit coupling is small in the perovskite manganite. This is consistent with small magnetocrystalline anisotropy in this material.

Magneto-optical Kerr rotation θ at 3.1 eV without photoexcitation and the maximum photoinduced change in magneto-optical Kerr rotation $\Delta\theta_{max}$ are shown in Fig. 4(a). The $\Delta\theta_{max}$ forms a peak at 340 K, slightly below the Curie temperature T_C. Note also that $\Delta\theta_{max}/\theta \approx -1$ for $T > 340$ K. This means that the ferromagnetic order is almost totally destroyed by the photoexcitation above 340 K. Provided that the photoexcitation increases the temperature of the spin system, the present photoexcitation of $\sim 500\ \mu J/cm^2$ corresponds to the heating of the spin system

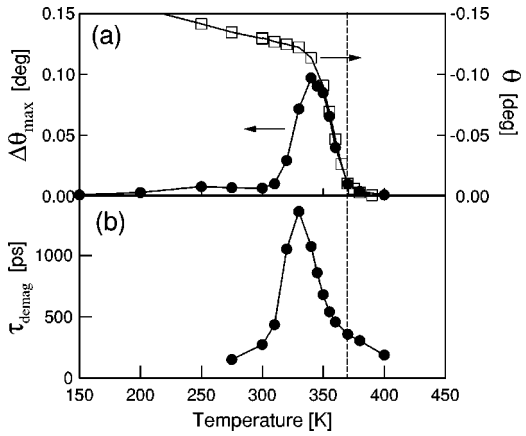

FIG. 4. (a) Magneto-optical Kerr rotation angle θ at the photon energy of 3.1 eV and the maximum values of $\Delta\theta$. (b) Time constant for the photoinduced demagnetization. The vertical dashed line indicates the Curie temperature $T_C = 370$ K.

by ≈ 30 K. The time constant of demagnetization τ_{demag} [Fig. 4(b)] shows a peak at 330 K with a value of 1.4 ns, where the spin temperature should reach near the Curie temperature $T_C = 370$ K by the photoexcitation. This can be interpreted in terms of the critical slowing-down effect as generally observed in the case of second-order phase transition.[6] The value of τ_{demag} is almost one order of magnitude larger than that deduced for similar manganites in previous reports.[8–12] In those works, the demagnetization time constant was evaluated from the temporal behavior of absorption change alone, and thus it is quite likely that the nonmagnetic contribution, which has competing time scale, was not thoroughly excluded when estimating the demagnetization time constant.

The process of the photoinduced demagnetization in $La_{0.6}Sr_{0.4}MnO_3$ is as follows. At first, the temperature of the quasiparticle excitation is raised, while their spin polarization was kept unchanged, by the photoexcitation within the pulse duration of ~ 200 fs. Then the temperature of the lattice system follows it by electron-lattice interaction within a few picoseconds (this can be observed in the tail of the spikelike response in $\Delta R/R$), although the system still retains the initial magnetization At this moment, the low-energy optical (and perhaps dc) conductivity decreases owing to heating of the quasiparticle excitation, but not due to the decrease of magnetization. Subsequently, the spin system relaxes to the new equilibrium state by the spin-lattice interaction. Such an interpretation is essentially the same as that proposed in the previous work.[12] Our studies, however, have demonstrated unambiguously the dynamical aspects of the demagnetization including the precise evaluation of time constant by comparing the response obtained by the magneto-optical measurement with those by the conventional pump-probe reflectivity measurement.

To summarize, we have observed photoinduced dynamics of magnetization in $La_{0.6}Sr_{0.4}MnO_3$ by time-resolved magneto-optical Kerr spectroscopy. The observed clear difference between the photoinduced changes in reflectivity and magneto-optical Kerr rotation indicates that the demagnetization does not occur immediately after the photoexcitation, while the spectral weight in a low-energy region (<1.5 eV) decreases instantaneously (<200 fs) with the total magnetization almost unchanged. The photoinduced demagnetization in $La_{0.6}Sr_{0.4}MnO_3$ shows a critical slowing-down effect with a prolonged time constant of up to 1.4 ns around 330 K, where the spin temperature after the photoexcitation almost reaches the Curie temperature 370 K.

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