

Spin–lattice interaction in colossal magnetoresistance manganites

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The metal–insulator transition and underlying spin dynamics in $\text{La}_{0.7}\text{D}_{0.3}\text{MnO}_3$ ($\text{D}=\text{Ca}, \text{Sr}$) are investigated using optical pump–probe spectroscopy at 1.5 eV. Our measurements, which span the ferromagnetic–paramagnetic transition temperature, reveal that the dynamics of the optically induced spectral weight transfer follow the temperature dependence of the magnetic specific heat. This dependence reflects the intrinsic interdependence between the optical conductivity and magnetism in the manganites allowing for the determination of the spin-lattice coupling magnitude.
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The metal–insulator transition (MI) in the manganite perovskites ($\text{Re}_{1-x}\text{D}_x\text{MnO}_3$, where Re is a rare earth such as La or Nd and D is a divalent alkali such as Sr or Ca) was extensively studied in the 1950's and 1960's¹ and theoretically interpreted using the double exchange model (DEX).² More recently, it has been realized that double exchange alone cannot account for the observed changes in resistivity as a function of temperature (or as a function of applied magnetic field) in the manganites. The highly correlated nature of the spin, lattice, charge, and orbital degrees of freedom lies at the heart of the phase transition.³ Below T_c , this strong correlation leads to metallicity and ferromagnetic order ($x=0.2\text{--}0.4$). Commensurate with the MI transition is a dramatic spectral weight transfer (SWT) of the optical conductivity from higher energies (~ 1.5 eV) to lower energies (below ~ 1 eV).⁴

The double exchange interaction (J_{DEX}) that induces ferromagnetism in the manganites is an *effective* interaction that is unlike the familiar Heisenberg direct exchange interaction.⁵ The double exchange interaction is related to the kinetic energy of the e_g electrons and their hopping between neighboring Mn atoms. This leads to the strong correlation between transport and magnetization.⁶ This correlation is reflected in the prevailing CMR theories that attribute the MI transition to either the magnetization induced increase in the electron conduction bandwidth,⁷ the magnetization related current carrier density collapse,⁸ or a magnetization dependent localization length or barrier.⁹

Despite the tremendous surge in interest in this class of materials, the investigation of the ultrafast dynamics of the MI transition has been lacking. In previous work we have demonstrated that optical transient absorption is a sensitive technique for investigating the MI transition by monitoring the temporal evolution of the spectral weight transfer in $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ (LCMO) upon photoexcitation.¹⁰ In this letter we present data on $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ (LSMO) (in addition to LCMO) and model the results using a two-temperature

model for the spin–lattice interaction thus showing that DSWT is associated with photoinduced demagnetization. Our results are consistent with the fact that magnetism and transport in the manganites are interdependent in an intrinsic manner.

The epitaxial $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ (LCMO) and $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ (LSMO) films (~ 75 nm thick) used in the study were prepared by pulsed laser deposition on LaAlO_3 substrates using a XeCl excimer laser.¹¹ The oxygen background pressure was 400 mTorr and the heater block temperature was 1000 K. After the deposition the sample was annealed at 1170 K under flowing oxygen. From magnetization data, the LCMO film had a paramagnetic insulator (PI) to ferromagnetic metal (FM) transition at 270 K, while the LSMO film exhibited a paramagnetic metal (PM) to FM transition at 355 K. The films were excited and probed with 55 fs/1.5 eV pulses at various temperatures above and below T_c . A low repetition-rate (1 kHz) system was used for the measurements to avoid a residual background signal due to long-lived excitations. The absorption coefficient of the films was $\sim 9 \times 10^4 \text{ cm}^{-1}$. The excitation fluence of $\sim 30 \mu\text{J}/\text{cm}^2$ generated an excited carrier density of $\sim 10^{19} \text{ cm}^{-3}$. The pump and probe beams were both linearly polarized and orthogonal to each other. Transient reflection and transmission were measured simultaneously, allowing for the extraction of absorption and refractive index dynamics. No Fabry–Perot effect was detected in these highly absorptive films and the transient transmission was found to faithfully reflect the absorption dynamics. Therefore we present induced transmission changes normalized to the film transmission $\Delta\mathcal{T}/\mathcal{T}$.

In previous work¹⁰ we showed that independent of the excitation energy, the measured response probed at 3 eV is weakly temperature dependent while that probed at 1.5 eV changed dramatically in amplitude and time constant as a function of temperature. Kramers-Kronig requires that refractive index changes at 3 eV reflect the absorption change at other frequencies (those measured at 1.5 eV among them). The lack of temperature dependence in the dynamics probed at 3 eV can be understood by assuming that the observed dynamics at 1.5 eV is compensated with correlated, yet opposite, changes in the Drude low energy part of the spectrum. This was termed dynamic spectral weight transfer

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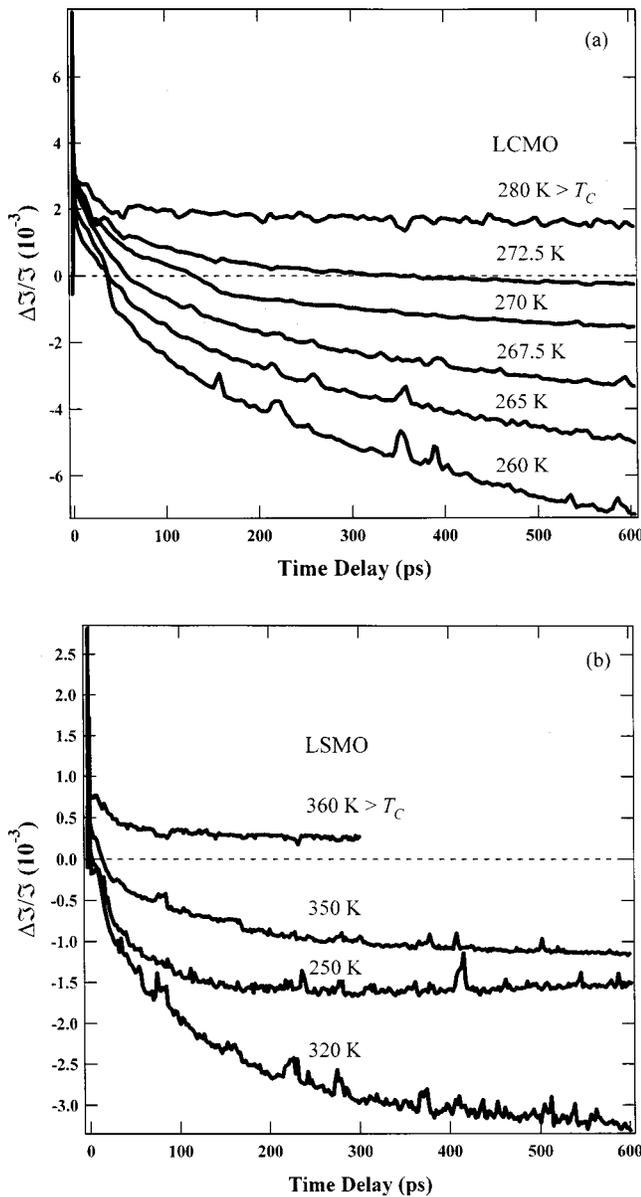


FIG. 1. Fractional transmission transients at 1.5 eV in LCMO (a) and LSMO (b).

(DSWT) representing the photoinduced partial transfer, below T_c , of optical conductivity from the low-energy (<1 eV) part of the spectrum to the high-energy region (~ 1.5 eV). In recent 1.5 eV-pump/terahertz-probe experiments we measured transient conductivity changes with time constants whose temperature dependence quantitatively correlates with, yet is opposite in amplitude to, the dynamics probed at 1.5 eV.¹² In this context, the increase of resistance upon photoexcitation in the FM state reported by Zhao *et al.*,¹³ indicates a suppression of the Drude spectral weight and is consistent with DSWT. However, the nanosecond dynamics presented in Ref. 13 reflect the recovery of the SWT rather than its onset.

The induced change in transmission is plotted in Fig. 1(a) for LCMO and Fig. 1(b) for LSMO at different temperatures near their respective transition temperatures. Both the LSMO and LCMO films exhibit qualitatively similar dynamics. The dynamics in Fig. 1 exhibit two components: an ultrafast (<1 ps) component and a much slower component

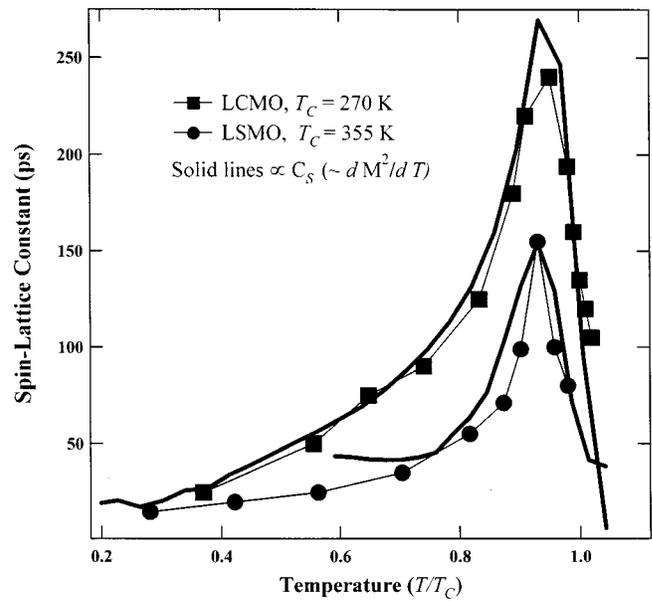


FIG. 2. Spin-lattice relaxation constant (τ_{SL}) for LCMO (■) and LSMO (●) as a function of the temperature normalized to T_c . The scaled magnetic specific heat (solid lines) is superimposed on τ_{SL} for both LCMO and LSMO.

(~ 20 – 200 ps). In the following, we focus on the slow component, which we attribute to the spin-lattice thermalization. The fast component will be discussed in a future publication. The component of interest is represented by the decrease in transmission (increase in absorption) that develops below T_c . The time constant for this component is plotted in Fig. 2 for LCMO and LSMO as a function of the temperature normalized to their respective T_c . The time constant in LCMO peaks at ~ 260 K with a value of ~ 250 ps decreasing to ~ 20 ps at low temperatures. The extracted time constant for LSMO also peaks below T_c .

In their work Zhao *et al.* attempted to explain the observed conductivity dynamics in LCMO through the electronic nature of the optical transitions at 1.5 eV including the photoionization of the Jahn–Teller distortion and other spin–flip transitions.¹³ The fact is that the assignment of optical transitions is still a controversial subject and our data¹⁰ argue against this approach. The two important observations from our work in Ref. 10 are that (1) the measured optical response probed at 1.5 eV is vastly different from that probed at 3 eV where it is insensitive to the MI transition, and (2) the response measured at either probe energy, below and above T_c , is independent of the excitation used whether at 1.5 or 3 eV. The first observation suggests that the 1.5 eV photon probes the dynamics of the e_g conduction electrons while the 3 eV photons do not probe these electrons. The second observation reflects the fact that the long time scale dynamics is determined by the interaction of the phonons with the elementary excitations of the spin system, the magnons. The exact nature of the electronic excitation (i.e., initial and final states involved) on the other hand is expected to govern the dynamics within the first few picoseconds. The success of the two-temperature model, as shown below, further supports this conclusion.

To model the energy transfer between the lattice and magnetization (spin system), we invoke the two-temperature

model. We assume that the highly excited electronic excitation has shed its excess energy and thermalized within the first couple of picoseconds. For weak lattice heating and for time delays beyond 5 ps the spin–lattice interaction can be modeled as $\partial(\Delta T_{\text{SL}})/\partial t = -G_{\text{SL}}/\beta(\Delta T_{\text{SL}})$, where $\Delta T_{\text{SL}} = T_L - T_S$ is the spin-lattice temperature difference, G_{SL} is the spin lattice energy coupling constant and $\beta = C_S C_L / (C_S + C_L)$ with the spin and lattice specific heat given by C_S and C_L , respectively. Heat conduction terms are ignored since they are weak and operate in the nanosecond regime. The spin-lattice relaxation time is given by $\tau_{\text{SL}} = \beta/G_{\text{SL}}$.

Using a mean field approximation, the spin specific heat is given by¹⁴ $C_S = -\alpha \partial M^2 / \partial T$, where M is the measured magnetization and $\alpha = 3SRT_c / [2(S+1)M_o^2]$ with $S = 2x + 3/2(1-x)$ is the average Mn spin and M_o is the saturated low- T value of M . The calculated spin specific heat peaks below T_c at a value of 30 and 41 J/(K mole) for LCMO and LSMO, respectively. The value for LCMO agrees with the measured anomalous contribution to the specific heat near T_c .¹⁵ This contribution is attributed to magnetic correlation. The calculated spin specific heat (appropriately scaled) is superimposed on the measured τ_{SL} in Fig. 2 for both LCMO and LSMO (solid lines). This demonstrates that the temperature dependence of the spin-lattice relaxation time in the critical region close to T_c originates from the magnetic specific heat while the spin-lattice energy coupling $G_{\text{SL}} (= \beta/\tau_{\text{SL}})$ displays no strong temperature dependence. Furthermore, from our data we estimate the spin-lattice coupling $G_{\text{SL}} \sim 9.5$ and ~ 20 W/(K mole) for LCMO and LSMO, respectively (β was calculated by taking the lattice specific heat close to T_c as $C_L \sim 100$ J/(K mole) and neglecting its temperature dependence in comparison to that of C_S .) Microscopically, spin-lattice relaxation occurs through the coupling of spins to the anisotropic fluctuations of the crystal field produced by the phonons. This coupling is mediated by the spin–orbit interaction. For comparison, the spin lattice relaxation in rare-earth Gd was measured¹⁶ and theoretically modeled¹⁷ to be around 100 ps consistent with our result. This can also be considered as a magnetostriction effect.

In conclusion, we have measured the ultrafast dynamics of the metal–insulator transition in $\text{La}_{0.7}\text{D}_{0.3}\text{MnO}_3$ ($\text{D}=\text{Ca, Sr}$) as a function of temperature. We modeled the slow component with a two-temperature model representing the energy exchange between the lattice and the spin system. The temperature dependence of the spin-lattice relaxation was shown to follow that of the spin specific heat with no

significant change in the spin-lattice coupling constant. This behavior is a reflection of the interdependence of the optical conductivity, represented by the SWT and magnetization in the manganites.

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