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Ultrafast mid-infrared dynamics in the colossal magnetoresistance pyrochlore $Tl_2Mn_2O_7$

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Abstract: An optical pump, mid-infrared (IR) probe system is used to investigate ultrafast temperature-dependent dynamics of the colossal magnetoresistance pyrochlore $Tl_2Mn_2O_7$. The dynamics change appreciably near the Curie temperature (T_c), indicating a dependence on ferromagnetic ordering.

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Colossal magnetoresistance (CMR) materials, in which the electrical resistance changes drastically with application of a magnetic field, have received a great deal of attention in recent years, both due to fundamental interest in this phenomenon as well as potential applications in areas such as magnetic sensing and recording. The best known example of these materials are the ferromagnetic perovskite manganites such as $La_{1-x}Sr_xMnO_3$ [1, 2], in which the CMR mechanism originates from the combined effects of double exchange and Jahn-Teller distortions. Ultrafast optical spectroscopy has contributed to an understanding of the manganites by differentiating between the relative contributions of the lattice and spin degrees of freedom [3, 4].

The pyrochlore $Tl_2Mn_2O_7$ also exhibits CMR below $T_c \sim 120$ K, with ferromagnetic ordering due to Mn quasi-localized spins, but the CMR mechanism is expected to be very different since double exchange and Jahn-Teller effects are negligible in these compounds due to the low carrier density ($0.7 \cdot 5 \times 10^{19} \text{ cm}^{-3}$) and absence of the Jahn-Teller ion Mn^{3+} [5, 6]. Optical conductivity measurements revealed a transition from an insulator-like to a metallic structure as the temperature was tuned below T_c , with a corresponding increase in the carrier density and change of the reflectivity with temperature near the plasma edge (~ 0.1 eV-see inset to Figure 1(a)) [7]. In this work, we use an optical pump, mid-IR probe system to investigate ultrafast dynamics in $Tl_2Mn_2O_7$ near the plasma edge of the reflectivity as a function of temperature. We observe distinct changes as the temperature is tuned through T_c , in particular a large increase in the peak amplitude and offset of the signal, corresponding to an increase in the fractional carrier density near T_c that may be attributed to photoinduced enhancement of ferromagnetic ordering.

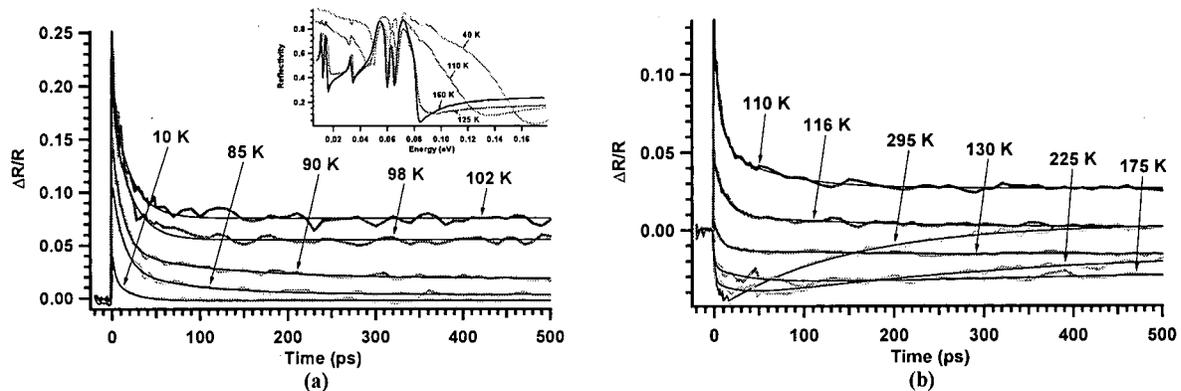


Fig. 1. Temperature-dependent measurements on $Tl_2Mn_2O_7$ with an 800 nm pump and 10 μm probe. (a) Dynamics for $T < T_c$. The inset shows the reflectivity as functions of temperature and frequency, reprinted with permission from ref. [7]. (b) Dynamics for $T > T_c$.

The output of a 1 kHz regenerative amplifier producing 2 mJ, 60 fs pulses at 800 nm is split into two beams to excite the sample and pump an optical parametric amplifier (TOPAS, Light Conversion Systems). The signal and idler beams from the TOPAS are mixed in a nonlinear crystal to generate the tunable 3-20 μm mid-IR probe. The sample preparation is described in reference [5]. The sample was pumped in reflection at 800 nm (1.55 eV) with a fluence of $460 \mu\text{J}/\text{cm}^2$, exciting a carrier density of $2.65 \times 10^{20} \text{ cm}^{-3}$. The pump only excites electrons in the minority spin band (the energy gap in the majority spin band is ~ 2 eV) [8], which are believed responsible for CMR in $\text{Tl}_2\text{Mn}_2\text{O}_7$ [9]. Temperature-dependent measurements with a 10 μm (0.125 eV) probe are shown in Figure 1.

The initial increase in reflectivity at all temperatures is due to a shift of the plasma edge to higher energies resulting from the large excited carrier density; measurements performed at other pump fluences (not shown) as well as simulations based on a Drude model for the reflectivity support this argument. The fast initial relaxation in $\Delta R/R$ is due to a decrease in carriers that contribute to the Drude-like response. However, near T_c , the peak amplitude and offset from zero of the signal at long time delays increase significantly. This dependence is shown in Figure 2. This suggests that the photoexcited carriers interact with the quasi-localized Mn moments, possibly enhancing the ferromagnetic alignment of Mn spins.

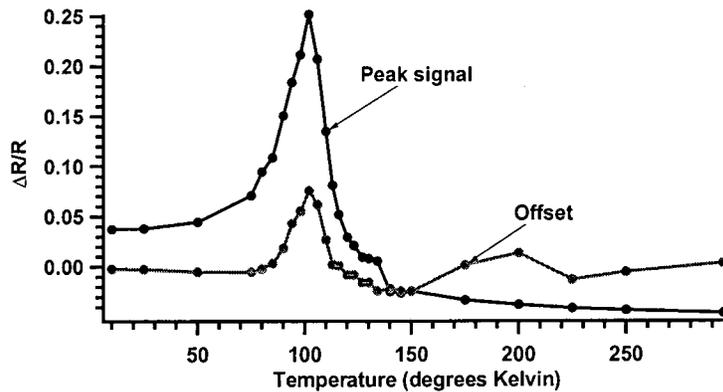


Fig. 2. Peak amplitude and offset from zero of the pump-probe data shown in figure 1 as a function of temperature. There is a large reflectivity enhancement near T_c , indicating that a greater fraction of the photoexcited carriers contribute to the charge transport (i.e., Drude response). The peak change in R occurs below T_c due to heating effects at this pump fluence.

This can be seen more clearly in Figure 3, where we have plotted the photoinduced carrier density at long times and compared it to the maximum possible change in carrier density (given by $n(10 \text{ K}) - n(T)$, with n the carrier density extrapolated from the data in ref. [7]). At low temperatures the magnetization, M , is saturated; therefore the excess carrier density cannot enhance the alignment of Mn spins and the carriers rapidly decay. Above T_c , thermal fluctuations prevent the excess carriers from interacting with the Mn moments, and again the carriers rapidly relax, shifting the plasma edge back towards its equilibrium value. In the vicinity of T_c , however, it appears that a fraction of the initially excited carrier density enhances ferromagnetic ordering. Time-resolved magneto-optical Kerr effect measurements are required to fully clarify this.

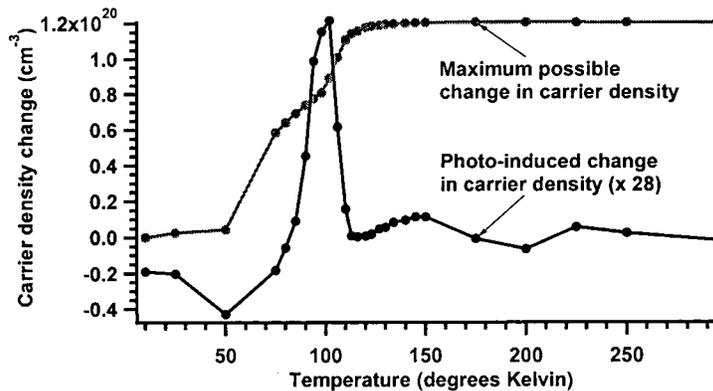


Fig. 3. Photo-induced carrier density at long time delays compared with the maximum possible carrier density change extrapolated from the data in ref. [7]. The negative values at low temperatures (< 80 K) in the experimental plot are due to lattice heating effects (and the resultant decrease in carrier density), and the magnitude is multiplied by 28 for comparison with the calculated plot.

In optical conductivity measurements [7], it was shown that n is proportional to M^2 , where a decrease in temperature or increase in magnetization led to an increase in n . This behavior was most pronounced near T_c and may be related to a shift of the minority spin band below the Fermi energy [9]. It appears that in the present work, a photoinduced increase of carriers leads to an enhancement of M near T_c . We note, however, that at maximum, only 3.6% of the excited carriers contribute to the increase in reflectivity near T_c at long times. Overall, these dynamics greatly differ from those measured in the manganites under similar excitation conditions [3].

Experiments were also performed with a pump wavelength of 400 nm (3.1 eV), obtained by frequency doubling the 800 nm light in a BBO crystal. The pump fluence was $316 \mu\text{J}/\text{cm}^2$, exciting a carrier density of $2.5 \times 10^{20} \text{ cm}^{-3}$. Temperature-dependent measurements at a probe wavelength of $10 \mu\text{m}$ are depicted in Figure 4. From these measurements, it is clear that the induced changes in reflectivity with a 400 nm excitation are significantly different from that measured with an 800 nm excitation (Figure 1). As Figure 4 shows, at all temperatures there is no long lived excess carrier density as was seen with an 800 nm excitation. The dynamics cannot be linked to the Drude response or magnetization in this case. We believe that this is due to excitation of carriers in the majority spin band that were inaccessible with the 1.55 eV pump. These carriers are not expected to contribute to CMR, and therefore different photo-induced dynamics are anticipated. It is worth noting that in the manganites, no significant difference was detected in the time-resolved signals when comparing 400 and 800 nm excitations [10].

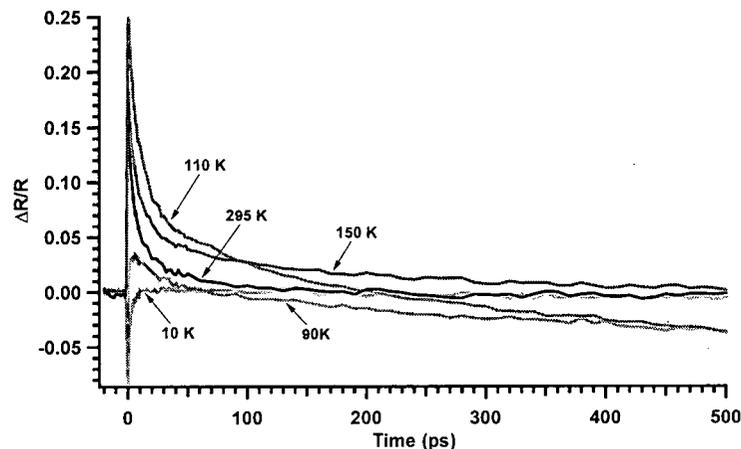


Fig. 4. Temperature-dependent measurements on $\text{Tl}_2\text{Mn}_2\text{O}_7$ with a 400 nm pump and $10 \mu\text{m}$ probe.

In conclusion, we have investigated ultrafast dynamics of the pyrochlore $\text{Tl}_2\text{Mn}_2\text{O}_7$ with an optical pump, mid-IR probe system. We find that with 800 nm excitation, a large increase in the peak amplitude and offset of the signal is observed near T_c , corresponding to a long-lived carrier density increase which may be due to reduced spin scattering resulting from dynamically enhanced ferromagnetic ordering. Future work will include time-resolved magneto-optical Kerr measurements to unambiguously resolve the spin dynamics.

References

1. S. Jin, T. H. Tiefel, M. McCormack, R. A. Fastnacht, R. Ramesh and L. H. Chen, "Thousandfold change in resistivity in magnetoresistive La-Ca-Mn-O films," *Science* **264**, 413-415 (1994).
2. A. J. Millis, "Lattice effects in magnetoresistive manganese perovskites," *Nature* **392**, 147-150 (1998).
3. R. D. Averitt, A. I. Lobad, C. Kwon, S. A. Trugman, V. K. Thorsmølle and A. J. Taylor, "Ultrafast conductivity dynamics in colossal magnetoresistance manganites," *Phys. Rev. Lett.* **87**, 17401-1-17401-4 (2001).
4. R. D. Averitt and A. J. Taylor, "Ultrafast optical and far-infrared quasiparticle dynamics in correlated electron materials," *J. Phys. Condens. Matter* **14**, R1357-R1390 (2002).
5. Y. Shimakawa, Y. Kubo and T. Manako, "Giant magnetoresistance in $\text{Tl}_2\text{Mn}_2\text{O}_7$ with the pyrochlore structure," *Nature* **379**, 53-55 (1996).
6. M. Subramanian, B. H. Toby, A. P. Ramirez, W. J. Marshall, A. W. Sleight and G. H. Kwei, "Colossal magnetoresistance without $\text{Mn}^{3+}/\text{Mn}^{4+}$ double exchange in the stoichiometric pyrochlore $\text{Ti}_2\text{Mn}_2\text{O}_7$," *Science* **273**, 81-84 (1996).
7. H. Okamura, T. Koretsune, M. Matsunami, S. Kimura, T. Nanba, H. Imai, Y. Shimakawa and Y. Kubo, "Charge dynamics in the colossal magnetoresistance pyrochlore $\text{Tl}_2\text{Mn}_2\text{O}_7$," *Phys. Rev. B* **64**, 180409-1-180409-4 (2001).
8. S. K. Mishra and S. Satpathy, "Electronic structure and exchange interactions in the manganese-based pyrochlore oxides," *Phys. Rev. B* **58**, 7585-7592 (1998).
9. H. Imai, Y. Shimakawa, Y. V. Sushko and Y. Kubo, "Carrier density change in the colossal-magnetoresistance pyrochlore $\text{Tl}_2\text{Mn}_2\text{O}_7$," *Phys. Rev. B* **62**, 12190-12194 (2000).
10. A. I. Lobad, A. J. Taylor, C. Kwon, S. A. Trugman and T. R. Gosnell, "Laser induced dynamic spectral weight transfer in $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$," *Chem. Phys.* **251**, 227-236 (2000).