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Probing nanoscale inhomogeneities in transition metal oxides with ultrafast mid-infrared spectroscopy

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Abstract

Transition metal oxides are a rich area of investigation for experimentalists and theorists alike due to their coupling of multiple degrees of freedom with similar interaction strengths. This complexity leads to intrinsic phase inhomogeneities that are believed to play a significant role in the fascinating phenomena observed in these systems. We use ultrafast mid-infrared spectroscopy to probe quasiparticle dynamics in the colossal magnetoresistive oxides, $Nd_{0.5}Sr_{0.5}MnO_3$ and $Tl_2Mn_2O_7$. Our results demonstrate for the first time that ultrafast spectroscopy is sensitive to the presence of nanoscale phase inhomogeneities, strongly indicating the universality of phase coexistence in complex transition metal oxides.

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The contribution of intrinsic phase coexistence to various phenomena such as high-temperature superconductivity and colossal magnetoresistance in transition metal oxides is an intense area of contemporary research [1]. Colossal magnetoresistance (CMR) has been observed in many oxides, with the best known example being the perovskite manganites. However, CMR has also been observed in other systems, such as the pyrochlore $Tl_2Mn_2O_7$, in which the double exchange and Jahn–Teller effects that are believed to contribute to CMR in the manganites are nonexistent [2]. Therefore, the primary origin of CMR is still a matter of intense debate, with nanoscale phase inhomogeneities believed to play a significant role in this effect. A deep understanding of their contribution to CMR therefore necessitates the development of advanced characterization techniques for spatially and temporally resolving the properties of these nanoscale entities.

Ultrafast optical spectroscopy has recently emerged as a powerful method for examining quasiparticle dynamics in transition metal oxides with femtosecond time resolution over a wide range of photon energies [3–5]. In this work, we use ultrafast mid-infrared (IR) spectroscopy to temporally resolve quasiparticle dynamics in the colossal magnetoresistive oxides $Nd_{0.5}Sr_{0.5}MnO_3$ (NSMO) and $Tl_2Mn_2O_7$ (TMO). These measurements reveal the presence of nanoscale inhomogeneities in these different systems, indicating their important role in the CMR effect.

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Optical pump, mid-IR probe experiments were performed in reflection on TMO and NSMO with an 800 nm pump and probe wavelengths of 5 and 13 μm, respectively (experimental details are described in Refs. [4,5]). The use of mid-IR probe wavelengths in both systems was critical in observing dynamics due to phase inhomogeneities; the 5 μm probe was tuned near the plasma edge of the reflectivity in TMO [6], where the photoinduced reflectivity changes were largest, and the 13 μm probe was tuned near a Jahn–Teller polaron absorption peak in NSMO [7].

We will first discuss carrier dynamics in NSMO, since the physics of manganites has been extensively studied with different experimental techniques [3,7,8]. NSMO is a paramagnetic (PM) insulator at high temperatures, transitioning to a ferromagnetic (FM) metallic state below $T_C = 250$ K and to a charge-and-orbital-ordered (COO) insulating state below $T_{CO} = 160$ K. Here we will focus on quasiparticle dynamics for $T > T_{CO}$; the dynamics for $T < T_{CO}$ will be discussed in a separate publication. The normalized photoinduced reflectivity change, $\Delta R/R$, was measured as a function of the pump-probe delay t from 10 to 300 K. We extracted $\Delta R/R$ ($t = 0$) from our data (Fig. 1), which is due to the excitation of single polarons out of bound polaronic states, as evidenced by the overlap of the probe wavelength with the polaron absorption peak [7]. The excellent match between the temperature dependence of $\Delta R/R$ ($t = 0$) and that of the number of uncorrelated lattice polarons as measured by X-ray diffraction [8] further supports this argument (Fig. 1). Our data therefore demonstrates that sub-picosecond dynamics in NSMO are due to the excitation and subsequent redressing of uncorrelated lattice polarons; the redressing time obtained from our time-resolved data is $\backsim 300$ fs. The existence of these polaron excitations above T_{CO} directly shows the inhomogeneous nature of the majority PM and FM phases, in agreement with studies that point to the influence of phase separation on CMR in manganites [1,7,8].

The pyrochlore TMO is PM at high temperatures, undergoing an insulator–metal transition to an FM state below $T_C = 120$ K (near which it displays CMR). However, TMO does not support Jahn–Teller polarons, making quasiparticle dynamics in this material very different from those in NSMO. We extracted the photoinduced carrier density Δn at $t = 50$ and 500 ps from our optical pump, mid-infrared probe data and plotted it vs. temperature as shown in Fig. 2 [4]. It is clear from this figure that carriers recombine quickly for $T \ll T_C$; as T approaches T_C the carrier density becomes extraordinarily long-lived, after which it slowly decays at higher temperatures. This can be understood by observing that at the lowest temperatures, electron–hole (e–h) spin singlets are excited in a homogeneous background of t_{2g} spins, recombining rapidly as observed in Fig. 2. However, as T_C is approached from below, pockets of opposite t_{2g} spin develop, providing a path for quasiparticles to spatially separate by migrating to nearby domains. By examining the bandstructure [9], it can be seen that it is energetically favorable for photoexcited holes to cross into domains of opposite majority spin, while there is an energetic barrier for photoexcited electrons. Therefore, as T approaches T_C, electrons and holes can spatially segregate, inhibiting recombination and leading to the long-lived carrier density observed in Fig. 2. At higher temperatures ($T > 170$ K), thermal fluctuations prevent large scale domain formation and increase the probability of electron–hole wavefunction overlap, causing Δn ($t = 500$ ps) to decrease with increasing temperature. These results demonstrate that quasiparticle dynamics in TMO are governed by spin disorder, making it a particularly simple example where the transport properties are determined by intrinsic nanoscale inhomogeneity occurring at a second order phase transition [4,5].

In conclusion, our experiments demonstrate that ultrafast spectroscopy can detect the presence of intrinsic nanoscale inhomogeneities in transition metal oxides.

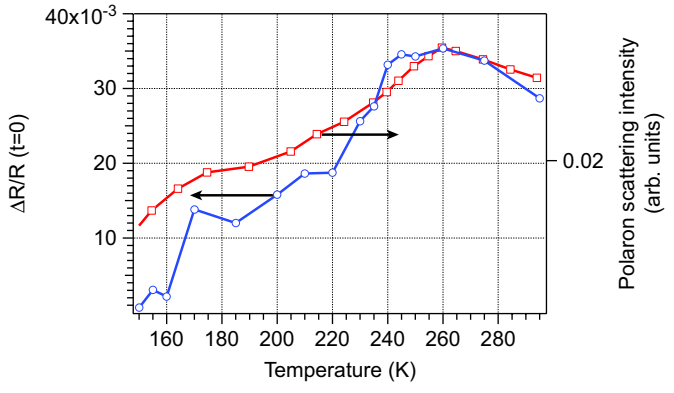

Fig. 1. Amplitude of $\Delta R/R$ measured at 13 μm at $t = 0$ ps (circles) compared to the number of uncorrelated lattice polarons, proportional to the polaron scattering intensity measured by X-ray diffraction (squares) (reprinted from Ref. [8] and scaled for comparison).

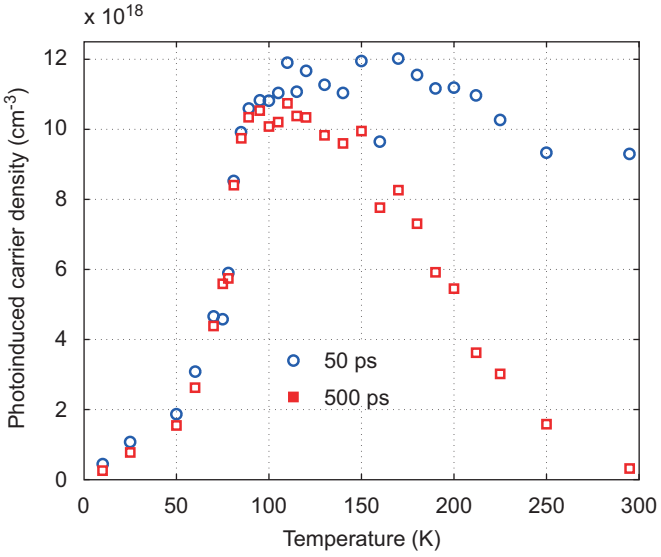

Fig. 2. (a) Photoinduced carrier density Δn at $t = 50$ ps (circles) and $t = 500$ ps (squares) (reprinted from Ref. [4]).

Optical pump, mid-IR probe experiments on NSMO demonstrated the ability to track the excitation and redressing of Jahn–Teller polarons on a sub-pico second time scale. Ultrafast mid-IR spectroscopy was also used to reveal the influence of intrinsic nanoscale inhomogeneity on charge transport in TMO for the first time, giving insight into the mechanism for CMR in this material. Our results strongly support the universality of nanoscale phase inhomogeneities in complex oxides and their influence on the fascinating phenomena observed in these systems.

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