Electronic Phase Manipulation on The Femtosecond Time Scale

Nathan Traynor- The University of Tennessee PHYS 599

Abstract

Over the past several decades an elusive goal of ultrafast science has been to achieve control of matter using light. In this talk I will focus on work claiming to induce an insulator-metal transition to a hidden metallic state in a manganite through selective pumping of a phonon mode[1]. In this work the authors use femtosecond laser pulses at 71meV (the energy of a phonon resonance) to pump $Pr_{0.7}Ca_{0.3}MnO_3$. By monitoring both the sample conductivity and reflectivity after pumping, a metallic state is found to emerge within 300 fs (the experimental resolution) of the pump pulse impinging on the sample. For comparison, the recombination of photoexcited electron-hole pairs across a Mott gap in Na_2IrO_3 is has been experimentally shown to take place within 200 fs[2]. Thus, it is reasonable that a metallic state in $Pr_{0.7}Ca_{0.3}MnO_3$ could form in a time shorter than 300 fs.

This metallic state is novel as it changes the conductivity of the sample by a factor of ~10^5 and no corresponding change is present when altering the sample temperature[3]-it is a hidden phase. This work has the potential to open a new paradigm in technology applications by leveraging strong correlations to selectively control electronic properties on ultrafast time scales. Moreover, generating novel ultrafast responses could open new avenues for studying the interactions that drive the formation of novel ground states in strongly correlated systems.

Overview

Manganites have been one of the most studied classes of materials in modern condensed matter physics. This class of materials has extremely rich phase diagrams, hosting various charge and magnetic order phases at different doping concentrations and temperatures[4]. Further, the manganites exhibit many insulator-metal transitions that can be driven by temperature changes, magnetic fields, and strong optical excitations[5]. The observed behavior of manganites is technologically interesting. However, these materials are also interesting from a fundamental science perspective, as their properties arise due to the strong correlations, and a delicate balance, between the various degrees of freedom (spin, lattice, orbital, charge) determining the ground state[6].

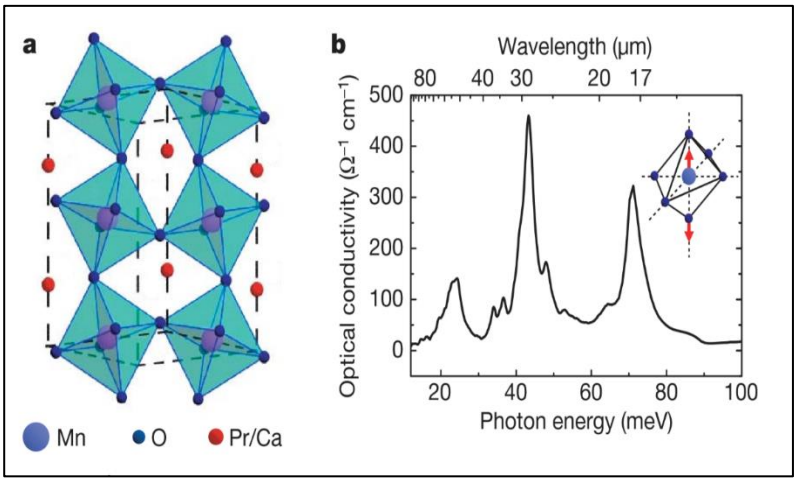

Figure 1: a) The crystal structure of $Pr_{0.7}Ca_{0.3}MnO_3$. The oxygen octahedra rotation (or orthorhombic distortion) is exaggerated for easier visualization. b) Optical conductivity data of $Pr_{0.7}Ca_{0.3}MnO_3$. The relative displacements for the mode at 71 meV are shown in the inset. (Figure from Ref. [1])

$Pr_{1-x}Ca_xMnO_3$ is a rather unique compound among the manganites as it displays robust insulating properties in the range of $0 \leq x \leq .5$ and $T \leq 400K$ [7]. However, it has been found that $Pr_{0.7}Ca_{0.3}MnO_3$ (crystal structure shown in Figure 1) borders a hidden metallic phase that can be induced through application of a magnetic field[7] or through irradiation with strong laser light pulses[8,9]. In the case of irradiation with laser light at energies larger than the insulating band gap, the origin of the insulator-metal transition is generally attributed to the melting of a charge ordered state due to hot carrier injection, and not simple heating of the lattice[10].

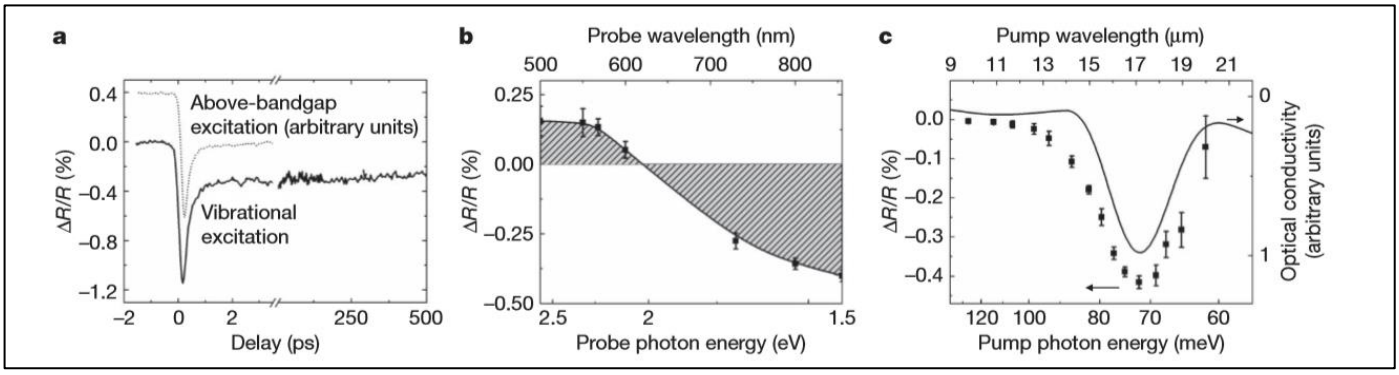

Figure 2: a) Pump-Probe reflectance spectra of $Pr_{0.7}Ca_{0.3}MnO_3$. The above bandgap excitation is generated by pumping with 800nm (1.5 eV) pulse and the vibrational excitation corresponds to a 17.5μm (71meV) pulse. b) Spectral dependence of the change in reflectivity as a function of probe wavelength. The solid line is a spline fit to the data. c) The change in reflectivity signal as a function of pump wavelength. The optical conductivity is convolved with the bandwidth of the pump pulses and plotted on the right axis. (Figure from Ref. [1])

In perovskites of structure ABO_3 the strength of the orthorhombic distortion is quantified by the tolerance factor, $\Gamma = r_{AO}/\sqrt{2}r_{BO}$, where r_{AO} is the average A-O distance and r_{BO} is the average B-O distance[1]. For 3d orbital systems, such as the manganites, the tolerance factor is strongly connected to the electronic state. In these systems transport is dictated by a super-transfer process: electrons hop between Mn *3d* orbitals via O*2p* states[3]. As the tolerance factor varies linearly with the Mn-O-Mn bond angle[11] it is related to the orbital overlap, and thus the hoping matrix element[1].

In Ref [1], the authors begin with optical conductivity measurements, shown in Figure 1. Three dominant perovskite phonon modes are identified in the low energy portion of the spectra. The most important mode for this work is centered at 71 meV (17.5μm) and is assigned to the Mn-O stretching mode (relative displacements shown in the inset of Figure 3b). The authors argue that as this mode modifies the Mn-O distance, it will have a dramatic effect on the tolerance factor and thus on the sample's electronic state.

The authors next describe reflectivity spectra, measuring the change in sample reflectance as a function of time after pumping for two cases: when the sample is excited with a pulse above the band gap (800nm) and when it is excited with light at the phonon resonance (17.5μm). The spectra are qualitatively similar, indicating a metallic state is formed after pumping in both cases. Previous studies have shown the formation of the metallic state by an 800 nm pump[8,9], however the formation of a metallic phase by pumping below the 0.3eV bandgap is a novel result.

The authors also measure the changes in the sample reflectivity 1ps after excitation as both the probe and pump wavelength are changed (Figures 2b and 2c respectively). When the probe wavelength is changed (shown in Figure 2b) there is reduced reflectivity for photon energies below ~2eV. This is indicative of the induced metallic state[10] and may be due to formation of a plasma edge. Also, in Figure 3b the authors show the change in reflectivity as the pump wavelength is changed. When compared to the optical conductivity spectrum (which is convolved with the pump bandwidth for comparison) the peak in both the reflectivity and the optical conductivity are centered around 71meV, showing the selective excitation of the M-O stretching mode corresponds to the maximum reflectivity change of the system.

Finally, the authors then show current and derived conductivity measurements as a function of time after vibrational excitation with a pulse of 71 meV photons (shown in Figure 3). Electrodes were evaporated on to the sample surface and biased. Then, the current following through the sample was measured after vibrational excitation. Upon vibrational excitation, the conductivity of the sample is found to increase by a factor of ~10^5 on

a timescale shorter than the experimental electronics can measure. This result supports the claim that an insulator-metal transition is being induced by the vibrational excitation at 71 meV.

Conclusions

In Ref. [1] the authors clearly show that they have driven an insulator-metal transition to a hidden state of $Pr_{0.7}Ca_{0.3}MnO_3$ by selectively exciting a phonon mode. This is novel as they have generated a hidden electronic state of matter (no corresponding metallic state exists when changing temperature) by coupling to the lattice in a strongly correlated system. This photoinduced transition is not the result of hot carrier injection- it would take 5 photons at 71 meV to span the insulating gap[1]. This work holds great promise for technology applications looking to leverage the strong correlations in oxide materials to drive dramatic material property changes. Further, if similar responses could be generated in other strongly correlated systems, such as the cuprate superconductors, studying these responses would give us another way to investigate how strong correlations drive the formation of novel ground states.

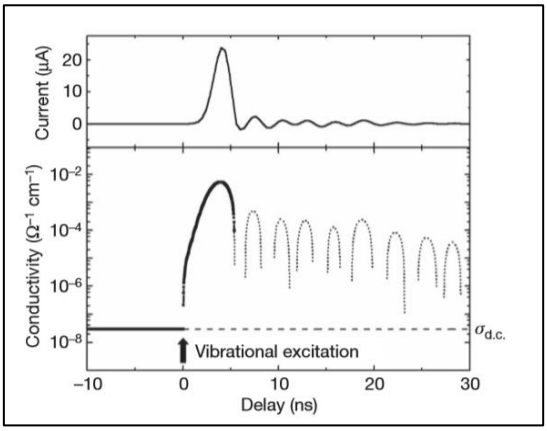

Figure 3: Conductivity measurements of the sample as a function of time after excitation with a 17.5 μm pulse. Oscillations in the current (and thus the conductivity) after the primary excitation are due to electronic ringing and thus not considered a reliable signal (Figure from Ref. [1])

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