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Ultrafast Mid-infrared Spectroscopy of the Charge- and Spin-Ordered Nickelate $La_{1.75}Sr_{0.25}NiO_4$

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Abstract. We present the first ultrafast mid-infrared study of charge and spin-ordered nickelates. A sub-picosecond modulation of the optical reflectivity is observed, indicating the filling and subsequent re-establishment of the pseudogap in the time-domain.

1 Introduction

The interplay of charge excitations with spin and lattice degrees of freedom in transition metal oxides leads to novel correlated phases [1], including high-temperature superconductivity in cuprates whose explanation remains elusive. In this context, nickelates represent a particularly interesting system to study charge correlations in antiferromagnetic insulators, since they share chemical and structural similarity to the cuprates yet allow access to carrier correlations without superconductivity at relevant doping levels. In particular, at low temperatures the carriers in nickelates order into static charge and spin stripes on an atomic-scale throughout a large portion of their phase diagram.

Ultrafast spectroscopy provides the important ability to transiently excite correlated systems into non-thermal phases, and the ensuing relaxation dynamics can provide the ability to distinguish otherwise entangled degrees-of-freedom in the time domain [2–4]. In charge-ordered systems, femtosecond spectroscopy in the near-IR and time-resolved photoemission has been applied e.g. to determine the coherent collective dynamics of charge-density waves in $TbTe_3$ and $K_{0.3}MoO_3$ [5,6]. In the nickelates, equilibrium investigations have revealed a pseudogap phenomenology, exposed in the optical response as a strong suppression of the mid-infrared conductivity followed by the formation of an energy gap below the stripe-ordering transition [7–9]. These features yield an optical probe of low-energy charge correlations in the nickelates. Here, we report the first ultrafast mid-infrared study of nickelates, which tracks the transient redistribution of spectral weight in the low-energy conductivity to provide insight into the dynamical relationship between short-range correlations and the establishment of long-range charge order.

2 Experimental Setup

In our pump-probe experiments, a femtosecond near-IR pulse centered at λ = 800 nm was used to photo-excite carriers across the low-energy conductivity gap of the stripe-ordered nickelate crystal. Transient mid-IR reflectivity changes are then detected with a time-delayed probe pulse (Fig. 1). The

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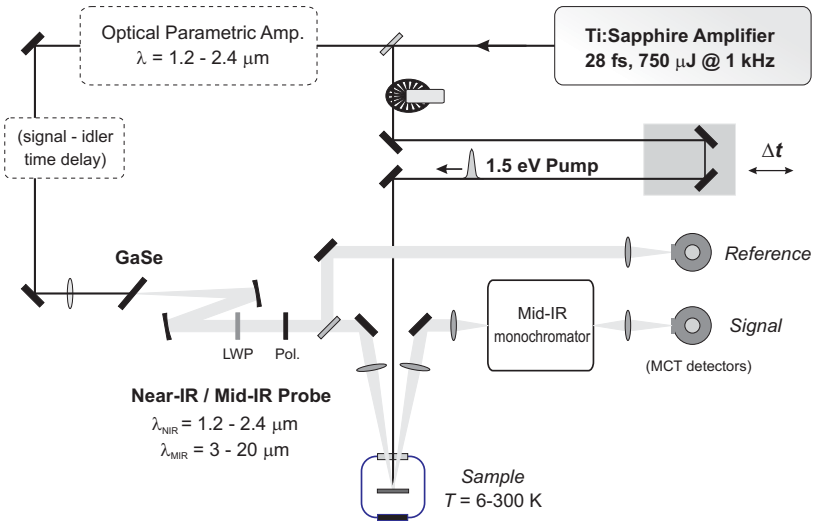

Fig. 1. Scheme of the experimental setup used for the time-resolved reflectivity experiment.

probe was broadly tunable in the near-infrared and mid-infrared spectral range and additionally spectrally resolved after the sample. Such pulses were generated by a ∼ 30 fs, 1-kHz Ti:sapphire amplifier via optical parametric amplification and difference frequency mixing. The combination of these two generation schemes allows studying the relevant spectral regions for the spectral weight transfer in nickelates. The time-resolved reflectivity experiment is combined with Fourier-transform infrared (FTIR) spectroscopy measurements of the equilibrium reflectivity at different temperatures. We report results obtained on a floating-zone grown $La_{1.75}Sr_{0.25}NiO_4$ (LSNO) single crystal, cut and polished to expose the bc-plane. Reflectivity is probed with a light polarization along the b-axis direction, while excitation of the sample in the time-resolved experiment is obtained using a pump pulse polarized 55 degrees off the b-axis with fluence 1.5 mJ/cm^2.

3 Results and Discussion

The equilibrium reflectivity of the LSNO crystal along the b-axis is shown in Fig. 2 for several representative temperatures, as measured using FTIR spectroscopy. It is characterized by an IR-active phonon mode centered at ∼ 86 meV and a strongly temperature-dependent electronic component that spans across the mid-IR. As the temperature is lowered, the reflectivity below ∼ 0.45 eV decreases while it increases at higher photon energies (see inset of Fig. 2). As shown by Katsufuji et al. for $La_{1.67}Sr_{0.33}NiO_4$ [7], these characteristic reflectivity changes signify a suppression of the low-energy spectral weight (i.e. pseudogap) above the charge-ordering transition followed by the opening of an energy gap in the optical conductivity below T_{CO}.

Fig. 2 shows the transient reflectivity change ΔR/R at low temperature (T = 30 K) and at two different probe wavelengths in the available energy range. The dynamics is characterized by a fast femtosecond decay (∼ 600 fs) and a slowly relaxing component that lasts well beyond the 10-ps time window shown here. The amplitude of the fast component depends strongly on the probe energy and changes sign between the two probing wavelengths. In contrast, the slowly relaxing component that survives at long delay times depends weakly on the probe energy and remains negative. By comparison with the equilibrium reflectivity, the sign change of the fast component demonstrates the filling and subsequent re-establishment of the pseudogap spectral weight on a femtosecond time scale. We have

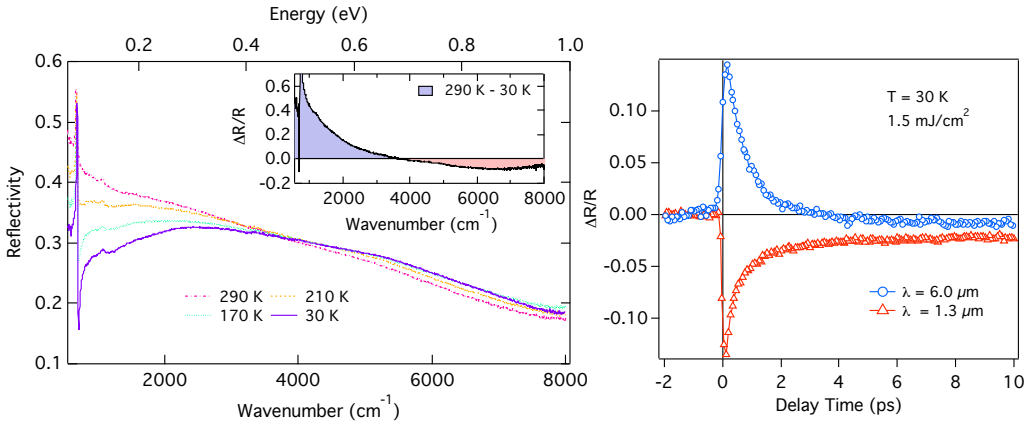

Fig. 2. (Left) Equilibrium reflectivity of our $La_{1.75}Sr_{0.25}NiO_4$ sample at different temperatures. The inset shows the relative difference in reflectivity between 30 K and 290 K. (Right) Ultrafast mid-IR reflectivity change ΔR/R after photoexcitation at 1.55 eV, probed at two different wavelengths (6.0 and 1.3 μm). The pump polarization is oriented 55 degrees off the b-axis and the pump fluence is 1.5 mJ/cm^2.

studied in detail the LSNO response as a function of probe photon energy, pump fluence, temperature and probe polarization, along with the decomposition of the signal into its underlying components (not shown). In particular, the $\sim$ 600 fs dynamics can be ascribed to short-range correlations of the electronic carriers in LSNO. This differs from the picosecond reformation kinetics of charge stripes observed in a recent time-resolved resonant x-ray diffraction study of the same LSNO system [10]. However, an additional slow component is also observed in the mid-IR response below T_{CO}, indicative of the recovery of the long-range order parameter of the charge stripes.

4 Conclusion

In conclusion, we have studied the femtosecond mid-IR response of the strongly-correlated nickelates for the first time. It reveals a strong, sub-ps modulation of the low-energy conductivity, allowing to disentangle local electronic correlations from the slower formation of the long-range order parameter of the stripe phase.

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