Probing Short- and Long-range Stripe Correlations in a Nickelate via Multi-THz Spectroscopy

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Abstract: We report optical-pump THz-probe reflectivity studies around the resonance frequency (≈11 THz) of the in-plane bending mode of a stripe-ordered nickelate. The experiments reveal signatures of both short and long-range nanoscale charge order dynamics.
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The dynamics of low-energy excitations and charge transport at the nanoscale is of critical importance for a wealth of fundamental physics and applications, ranging from quasi 1D conduction in nanostructured materials to charge ordering in complex compounds. The complex layered material $La_{1.75}Sr_{0.25}NiO_4$ (LSNO) represents an ideal model system to investigate such physics, since at low temperatures electrons organize in quasi 1D atomic-scale rivers of charge, called stripes [1]. Ultrafast studies are playing a pivotal role in highlighting the relevant cause-effect relations between real-space charge organization and the low-energy excitations [2,3]. However the ability to detect both short and long-range order via a single probe has remained challenging.

Here we report ultrafast optical-pump THz-probe spectroscopy of the stripe-ordered system LSNO. Ultrafast experiments in the multi-THz spectral range show strong THz reflectivity variations around the phonon bending mode frequency (≈11 THz). At low temperatures, this phonon mode exhibits a splitting directly related to the formation of the stripe order, while the background conductivity is reminiscent of the opening of the mid-IR pseudogap [3]. The transient THz probe therefore captures both the electronic and structural dynamics within the same spectrum. The results reveal the dynamical interplay between charge localization and the bending mode folding, providing insight into the symmetry breaking dynamics of nanoscale charge-order.

Figures 1(a) depicts the concept of the experiment, where an 800 nm pump photo-excites the LSNO sample, which is maintained in the stripe-ordered phase (T < 110 K). The pump pulse drives the ultrafast de-localization and re-localization dynamics as described in Ref. [3] and perturbs the amplitude and phase of the long-range charge order [2]. A non-resonant THz probe can readily detect the broad change in conductivity due to the closing of the mid-IR pseudogap associated with short-range correlations, whereas the detection of long-range order requires resonant probing of the IR-active lattice modes. We characterize the Ni-O bending mode resonance around 360 cm^{-1} via Fourier-transform spectroscopy of the reflectivity between ≈10 meV–1 eV. Reflectivity data are converted into the conductivity via Kramers-Kronig constrained variational analysis [3]. Figure 1(b) shows the in-plane spectra of

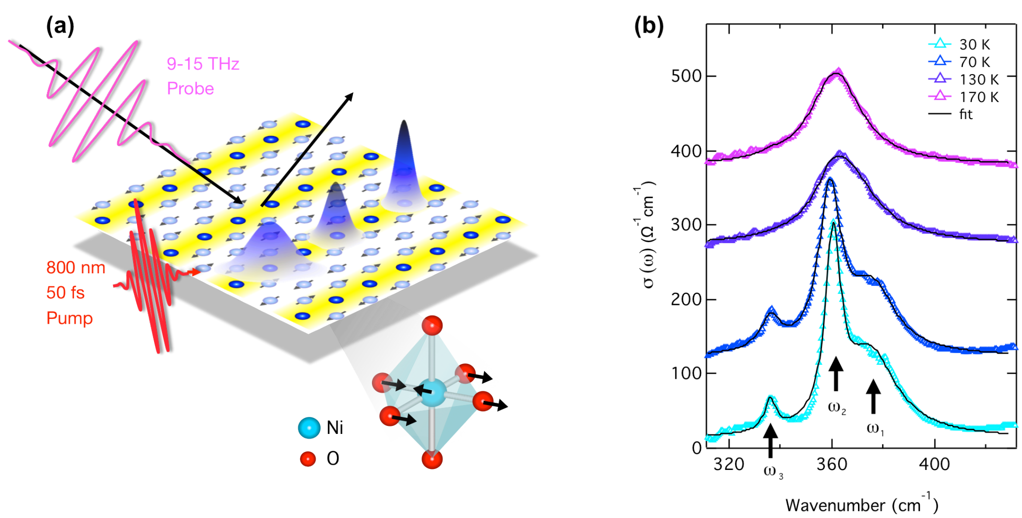

Fig. 1. **(a)** Representation of the pump-probe experiment on LSNO. The multi-THz probe detects both the localization dynamics of short-range charge correlations and the modifications of the Ni-O bending mode due to long-range charge order. **(b)** Equilibrium optical conductivity of the Ni-O bending mode as a function of sample temperature. Solid lines: multi-oscillator fits. Curves are offset vertically for clarity.

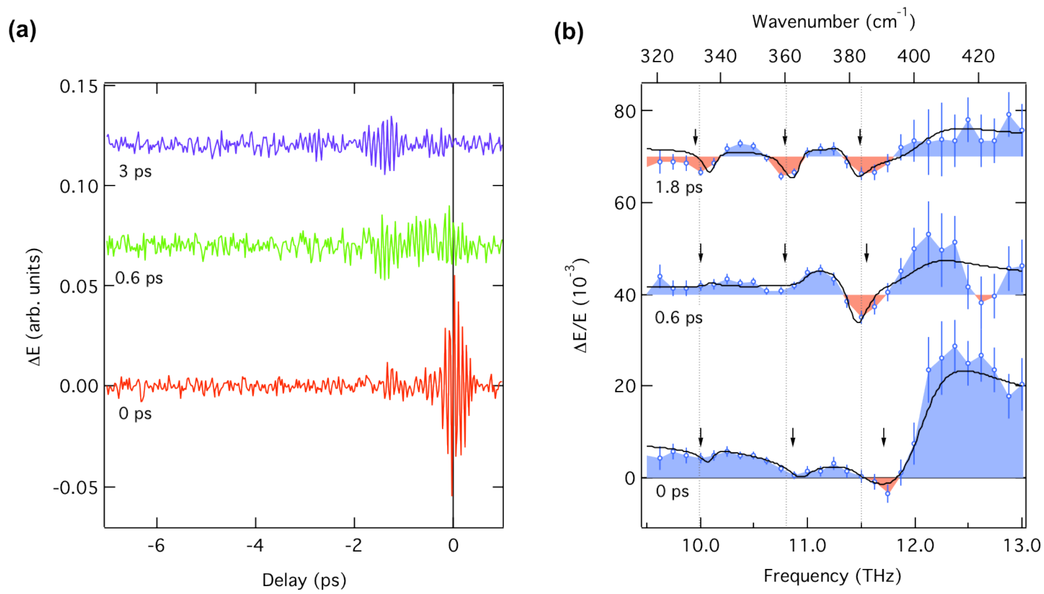

Fig. 2. **Fig. 2**. **(a)** THz temporal traces of the transient variations of the reflected electric field at different pump-probe delays ($\Delta t = 0$, 0.6, and 3 ps). **(b)** THz pump-probe spectral traces at indicated pump-probe 0, 0.6, and 1.8 ps delay times. Dotted vertical lines mark the positions of the three sharp phonon resonances at equilibrium from Fig. 1(b). The arrows mark the negative dips in the pump-probe data. Solid lines: multi-oscillator fits. Curves are offset vertically for clarity.

the bending phonon. Above the charge-ordering transition temperature the phonon behaves as a single Lorentz oscillator, while below 110 K three sharp resonances appear. This behavior is often observed in materials exhibiting charge order and it is well described by phonon zone folding at q_{CO} due to superlattice modulation [4].

To probe the dynamics of the short- and long-range order we perform ultrafast THz spectroscopy resonant on the Ni-O bending mode. We generate and detect THz radiation in thick GaSe crystals, which allow access to the multi-THz range with high spectral resolution. A resolution $\Delta\nu < 0.2$ THz is achieved by scanning up to 8 ps of the pump-probe THz field trace, as shown in Figure 2(a). The pump fluence is set to ≈ 0.5 mJ/cm^2 and the sample temperature is maintained at 20 K. The reflected THz electric field around $\tau_{THz} \approx 0$ ps promptly responds to photo-excitation, while a more complex structure appears in the time-domain THz trace at later pump-probe delay times. The corresponding spectral responses are shown in Figure 2(b). At a pump-probe delay of $\Delta t = 0$ ps, the broad increase of the reflected field indicates the modulation of the electronic background expected for the closing of the pseudogap [3]. At $\Delta t = 0.6$ and 1.8 ps we observe a fast decay of this broad component and the appearance of sharp negative features in proximity to the phonon resonances observed at equilibrium (see Fig. 1b). Simulations based on a multi-oscillator model and transfer matrix method well reproduce the data, with the pseudogap signal dominating at early pump-probe delays and the folded phonons remaining suppressed at later delay times.

We will discuss the multi-THz spectral results at several pump-probe delay times and temperatures. Our results indicate the suppression of the folded phonon signal after photo-excitation with a relaxation dynamics of ≈ 3 ps. This timescale is compatible with the amplitude dynamics of long-range order obtained from time-resolved X-Ray diffraction experiments and allows unequivocal recognition of the phase contribution [2]. The dynamics of the broad spectral component is in agreement with the localization timescale of short-range stripe correlations in LSNO [3]. The observation of clear signatures of both short-and long-range stripe correlations in the THz spectral response of LSNO provides unprecedented insight into the lattice zone-folding dynamics during the suppression of an electronic superlattice modulation.

References

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