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Author(s): R. J. McQueeney, LANSCE-12
J.L. Sarrao, MST-10

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Evidence for strong electron-lattice coupling in $\text{La}_{2-x}\text{Sr}_x\text{NiO}_4$

R. J. McQueeney* and J. L. Sarrao†

**Lujan Center and †Condensed Matter and Thermal Physics,
Los Alamos National Laboratory, Los Alamos, NM 87545*

Abstract. The inelastic neutron scattering spectra were measured for several Sr concentrations of polycrystalline $\text{La}_{2-x}\text{Sr}_x\text{NiO}_4$. We find that the generalized phonon density-of-states is identical for $x = 0$ and $x = 1/8$. For $x = 1/3$ and $x = 1/2$, the band of phonons corresponding to the in-plane oxygen vibrations (> 65 meV) splits into two subbands centered at 75 meV and 85 meV. The lower frequency band increases in amplitude for the $x = 1/2$ sample, indicating that it is directly related to the hole concentration. These changes are associated with the coupling of oxygen vibrations to doped holes which reside in the NiO_2 planes and are a signature of strong electron-lattice coupling. Comparison of $\text{La}_{1.9}\text{Sr}_{0.1}\text{CuO}_4$ and $\text{La}_{1.875}\text{Sr}_{0.125}\text{NiO}_4$ demonstrates that much stronger electron-lattice coupling occurs for particular modes in the cuprate for modest doping and is likely related to the metallic nature of the cuprate.

INTRODUCTION

It has been known for some time that strong phonon anomalies occur in the superconducting cuprates as a function of hole concentration. These anomalies were originally observed by inelastic neutron measurements of the phonon density-of-states of several superconducting cuprates and manifested themselves as strong softening of in-plane polarized oxygen modes as holes are added to the CuO_2 plane [1]. This softening behavior was confirmed by neutron scattering measurements of the phonon dispersion and was found to occur only for the longitudinal oxygen bond-stretching branch along the $(1,0,0)$ direction (along the Cu-O bond) and near the Brillouin zone boundary [2]. This effect appears to be universal, and has been observed in all cuprate systems for which the measurement has been made. More recently, careful phonon peakshape measurements in $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ have revealed anomalous splittings of this branch midway to the zone boundary at low temperatures [3]. This manifestation of strong and unique electron-lattice coupling for specific phonon polarizations and wavevectors in the cuprates brings several questions to mind: Are the phonon anomalies related to the inhomogeneous, or topological, distribution of doped holes, such as stripes? What is the origin of the

temperature effect? And finally, is the dynamic (or metallic) nature of the hole states important in the electron-lattice coupling?

To address these questions, we have undertaken measurements of the phonon density-of-states in the nickelate system, $\text{La}_{2-x}\text{Sr}_x\text{NiO}_4$, as a function of Sr (hole) concentration. The nickelates are isostructural to the 214-cuprates and are known to have similar phonon spectra in the undoped parent compounds [1]. For rather large Sr concentrations ($x > \sim 0.1$) [4,5], the nickelates form charge/spin ordered states similar to those proposed for the cuprates. However, in the nickelates the stripes are static and the system remains in insulator up to very large hole concentrations. Consequently, we can address several of the questions posed above. Additionally, the relevance of the electron-lattice coupling in the nickelate stripe ordering is an interesting question, and these measurements can help to address how the electron-lattice coupling enters in the stripe formation.

EXPERIMENTAL

In the present experiment, the inelastic neutron scattering spectra are measured for polycrystalline $\text{La}_{2-x}\text{Sr}_x\text{NiO}_4$ ($x = 0, 1/8, 1/4, 1/3, 1/2$) and also for $\text{La}_{1.9}\text{Sr}_{0.1}\text{CuO}_4$. The purpose is to obtain the dependence of the generalized phonon density of states (GDOS) on hole concentration, and also to determine the variation of the GDOS with temperature. This is the first systematic study of the doping and temperature dependence of the lattice dynamics of $\text{La}_{2-x}\text{Sr}_x\text{NiO}_4$ by neutron scattering. We pay particular attention to the contribution from in-plane polarized oxygen vibrations. This is made simple because the in-plane oxygen modes are well-separated in frequency from the other kinds of vibrations, as determined from model fits to phonon dispersion measurements. The features above 65 meV are associated entirely with planar oxygen vibrations.

$\text{La}_{2-x}\text{Sr}_x\text{NiO}_{4+\delta}$ powder samples were prepared by solid state reaction of stoichiometric ratios of La_2O_3 , SrCO_3 and NiO [6]. The $x = 0$ and $x = 1/8$ samples were subsequently annealed in flowing argon at 1100 °C for 20 hours to ensure that $\delta = 0$ [5]. X-ray powder diffraction measurements at room temperature yielded lattice constant values consistent with previous reports [7] and indicated predominantly single phase material, with only a small x -independent La_2O_3 impurity phase. Magnetic susceptibility measurements yielded data essentially equivalent to that of Cheong et al. [6], including the reported signal of stripe order for $x = 1/3$. The samples weighed about 40 grams each.

Time-of-flight inelastic neutron scattering measurements were performed on the Low Resolution Medium Energy Chopper Spectrometer at Argonne National Laboratory's Intense Pulsed Neutron Source. For all measurements, an incident neutron energy of 120 meV was chosen. Data were taken at $T = 10\text{K}$ and 300K for many different samples, some with the same Sr concentration but differing mass. Data were summed over all scattering angles from $2 - 120^\circ$ and corrected for background, absorption, sample mass, multiple scattering, and multiphonon processes. Excel-

lent agreement was achieved for different masses of the same stoichiometry. The data were then converted into the GDOS by accounting for the phonon amplitude factor.

RESULTS AND DISCUSSION

The GDOS of $\text{La}_{2-x}\text{Sr}_x\text{NiO}_4$ are shown for several Sr concentrations at $T = 10\text{K}$ in Figure 1. The GDOS for the $x = 0$ and $x = 1/8$ samples are identical, thus hole doping has little effect on the phonon spectrum at this level. This is in strong contrast to the cuprates. Figure 2 shows the GDOS obtained for $\text{La}_{1.875}\text{Sr}_{0.125}\text{NiO}_4$ and $\text{La}_{1.9}\text{Sr}_{0.1}\text{CuO}_4$ at $T = 10\text{K}$. While the GDOS of the undoped parent compounds are virtually identical, at $\sim 10\%$ doping the spectra differ greatly above 65 meV. In particular, the subband forming near 70 meV in the cuprate, which is associated with the anomalous bond-stretching modes, has not formed in the nickelate. Thus, simple mechanisms related to substitutional disorder or Madelung terms in the interatomic potential cannot possibly be responsible for the softening. Rather, the phonon softening in the cuprate must require hole mobility in addition to strong electron-lattice coupling. The formation of this subband may be related to the metal-insulator transition near the 5% doping level, although this has not been measured to date.

However, this does not preclude the presence of electron-lattice coupling in the

FIGURE 1. The generalized phonon density-of-states of $\text{La}_{2-x}\text{Sr}_x\text{NiO}_4$ for $x = 0, 1/8, 1/4, 1/3$ and $1/2$ at $T = 10\text{K}$. Data for each concentration is separated by 0.01 units for clarity.

FIGURE 2. The generalized phonon density-of-states $\text{La}_{1.875}\text{Sr}_{0.125}\text{NiO}_4$ and $\text{La}_{1.9}\text{Sr}_{0.1}\text{CuO}_4$ at $T = 10\text{K}$. The cuprate GDOS is offset by 0.01 for clarity.

nickelates themselves. For the higher doping compounds, strong deviations are seen in the in-plane polarized oxygen modes above 65 meV. This is especially apparent in the $x = 1/2$ compound. Here, a subband of phonon modes is formed near 75 meV, quite similar to the cuprates although at a much higher doping level. This subband can also be seen very weakly in the $x = 1/3$ compound. Other changes, such as the narrowing and slight hardening of the band near 85 meV can be observed for the $x = 1/4$, $1/3$ and $1/2$ systems. Additionally, smaller changes are observed at the higher Sr concentrations, such as the changes near 45 meV. Unfortunately, many kinds of phonon modes involving different atomic species are involved below 50 meV, and it is not possible to discuss these changes with the present data alone.

It is tempting to associate the renormalization of the oxygen phonon modes in the nickelates with stripe formation. With the localization of holes and their real-space ordered arrangement, one might expect the interatomic potentials for the strongly covalent Ni-O bonds to be affected. For $\text{La}_{3/2}\text{Sr}_{1/2}\text{NiO}_4$ below 350K, the holes form a checkerboard pattern on the NiO_2 plane. Thus, the 75 meV and 85 meV bands could be related to vibrations of NiO_4 squares with or without a hole, respectively. An extension [8] of the recent theoretical work by Yi, et al. [9] has calculated the expected phonon spectrum of the NiO_2 plane in the presence of strong electron-lattice coupling in addition to the strong 2-orbital electron-electron interactions and in the background of stripe order using an inhomogeneous Hartree-Fock numerical method. The results show a quantitative agreement with the measured doping dependence shown here and relate the 75 meV subband to local lattice vibrations in the vicinity of the stripes (or phonon modes localized on the stripe). The level of electron-lattice coupling required for agreement with the GDOS within Yi's model is $\alpha \approx 3$. According to Yi's results, the strength of the electron-lattice coupling can

FIGURE 3. Comparison of the generalized phonon density-of-states of $\text{La}_{2-x}\text{Sr}_x\text{NiO}_4$ ($x = 0, 1/3, 1/2$) at $T = 10\text{K}$ and 300K . Each GDOS is offset by 0.01 for clarity.

dictate stripe position and this value is close to the transition from Ni-centered to O-centered stripes for $x = 1/3$. It has been shown experimentally that the stability of Ni-centered or O-centered stripes is quite sensitive to temperature.

Further connection of the stripe order and phonon subband formation in the nickelates can be obtained from the temperature dependence. For $\text{La}_{5/3}\text{Sr}_{1/3}\text{NiO}_4$, the stripe order is lost above $\sim 240\text{K}$. Figure 3 shows a comparison of the GDOS for the nickelate compounds with $x = 0, 1/3$, and $1/2$ at $T = 10\text{K}$ and 300K . The $x = 0$ and $1/2$ GDOS are very similar for the two temperatures, including the 75 meV subband for $x = 1/2$ which still retains hole order. For $x = 1/3$, the GDOS appears more like $x = 0$ and may be related to the loss of hole order. For such complicated crystal structures, it is difficult to compare different temperatures since the contributions of each chemical species vary with the Debye-Waller factor and multiphonon corrections are more significant. While these data do give the inference of the phonon renormalizations being sensitive to hole order, it is only suggestive.

Other observations such as mode splitting through the stripe ordering transition by Raman [10,11] and Infra-red [12] absorption, and the fact that charge order occurs before spin order in these systems [13] point to the importance of the lattice degrees of freedom in the nickelates. Considering our results in conjunction with these others, electron-lattice coupling may be crucial in the stabilization of charge-ordered states. Additionally, in the cuprates, phonon induced charge transfer excitations and the large and anomalous electron-lattice coupling may be relevant to the electronic transport properties. In short, both the nickelates and cuprates show strong electron-lattice coupling effects for oxygen modes in the transition metal-oxide plane.

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