Mixed Lattice and Electronic States in High-Temperature Superconductors

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Inelastic neutron scattering measurements are presented which show the abrupt development of new oxygen lattice vibrations near the doping-induced metal-insulator transition in $La_{2-x}Sr_xCuO_4$. A direct correlation is established between these lattice modes and the electronic susceptibility (as measured by photoemission) inferring that such modes mix strongly with charge fluctuations. This electron-lattice coupling can be characterized as a localized one-dimensional response of the lattice to short-ranged metallic charge fluctuations.

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High-temperature superconductors are based on antiferromagnetic insulating materials caused by strong electronic correlations. Doping charge carriers (holes) into this system creates a two-dimensional correlated metallic state in the CuO_2 plane that becomes superconducting at low temperatures [1]. The development of the metallic state with hole doping and the metallic state itself still remain poorly understood. Much effort has been focused on the role of persistent antiferromagnetic spin fluctuations in transport properties and superconductivity of a Fermi liquid [2,3], mainly because it is believed that superconducting transition temperatures are too high to arise solely from electron-phonon coupling. At the other extreme, it has also been shown that the metallic phase has an instability towards charge and spin order and may consist of localized, atomic-scale charge and spin fluctuations, commonly referred to as the stripe model [4–6]. In any case, a study of the evolution of the lattice dynamics could shed light on the nature of the metal-insulator transition and the normal state properties. For example, because hole doping affects mainly the hybridized Cu $3d_{x^2-y^2}$ and O $2p_{(x,y)}$ antibonding electronic states, a localized matrix of slow charge fluctuations should interact with large dynamic lattice distortions, whereas very little doping dependence of the phonons is expected in a low-density Fermi liquid. Inelastic neutron scattering measurements of the lattice dynamics do show evidence of strong and unusual electron-lattice coupling in many high-T_c compounds [7–10]. However, several experimental and conceptual issues surrounding this anomalous coupling remain unclear. In particular, the doping dependence has not been studied in detail and the question of whether the phonon anomalies actually reflect the underlying charge distribution remains unresolved. Of course, the ultimate issue concerns the role of strong electron-lattice coupling in superconductivity. Here, we report the systematic development of anomalous phonon modes on hole concentration and the abrupt formation of new oxygen lattice vibrations near the metal-insulator transition (MIT) in $La_{2-x}Sr_xCuO_4$. It is plausible that these lattice modes are the bosons which interact with electronic states, causing a kink in the electronic dispersions observed by photoemission [11–14]. The results suggest that the lattice and charge dynamics are inexorably mixed in the high-temperature superconductors.

We performed inelastic neutron scattering measurements of phonon densities of states (DOS) in $La_{2-x}Sr_xCuO_4$ (LSCO) spanning hole concentrations from the undoped insulator to the optimally doped superconductor ($0 \leq x \leq 0.15$). The measurements were performed at $T = 10$ K on the LRMECS spectrometer at the Intense Pulsed Neutron Source at Argonne National Laboratory. Sample preparation details and extraction of the DOS from the raw data are similar to those previously reported for $La_{2-x}Sr_xNiO_4$ [15]. The results of the present measurements are shown in Fig. 1. Of primary importance is the abrupt development of new lattice modes near $\sim$70 meV for hole concentrations between 6%–8% ($x = 0.06$–0.08). Because the DOS is not an analytic function and cannot be reliably fit to a series of peaks, this development is characterized by calculating the curvature of the DOS at 72 meV (inset of Fig. 1) which changes sign in this concentration range.

The abrupt change in the DOS is not due to the tetragonal-orthorhombic structural phase transition, as all samples remain orthorhombic at $T = 10$ K (well below the tetragonal-orthorhombic phase transition temperature [16]). The new phonon band is also not caused by electrostatic impurity effects from Sr substitution because identical measurements on the isostructural nickelate compounds show no such band formation at these Sr concentrations [15]. Rather, the abrupt development of the 70 meV band must be related to the doping-induced MIT near $x \sim 0.05$, where trapped holes begin to become mobile [1,17]. The onset of superconductivity and dynamic incommensurate spin fluctuations (signatures of the stripe correlations) also occur in this critical concentration range. The nature of this MIT is unclear at present, however the transition does occur without a corresponding change in lattice symmetry. Thus, the 70 meV band signifies strong and unusual electron-lattice coupling in

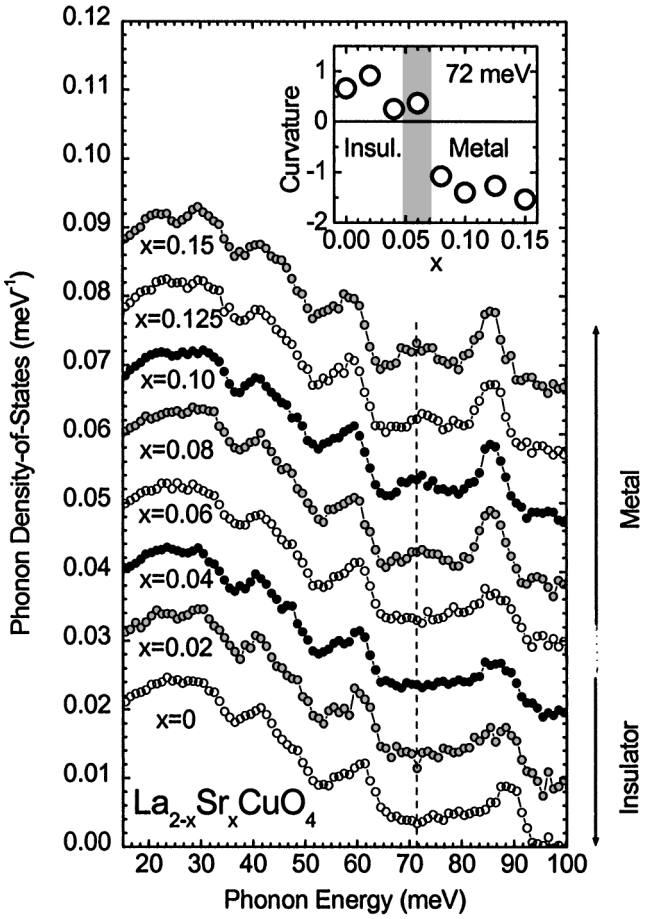

FIG. 1. The phonon densities of states of $La_{2-x}Sr_xCuO_4$ for several hole concentrations at $T = 10$ K. Each DOS is displaced along the y axis for clarity and the vertical dashed line indicates the new band formation. The scale on the right indicates schematically the metal-insulator transition as a function of doping. The inset shows the local curvature of the DOS at 72 meV as a function of Sr doping. The curvature was calculated after smoothing the DOS by convolution with a Gaussian of 2 meV standard deviation.

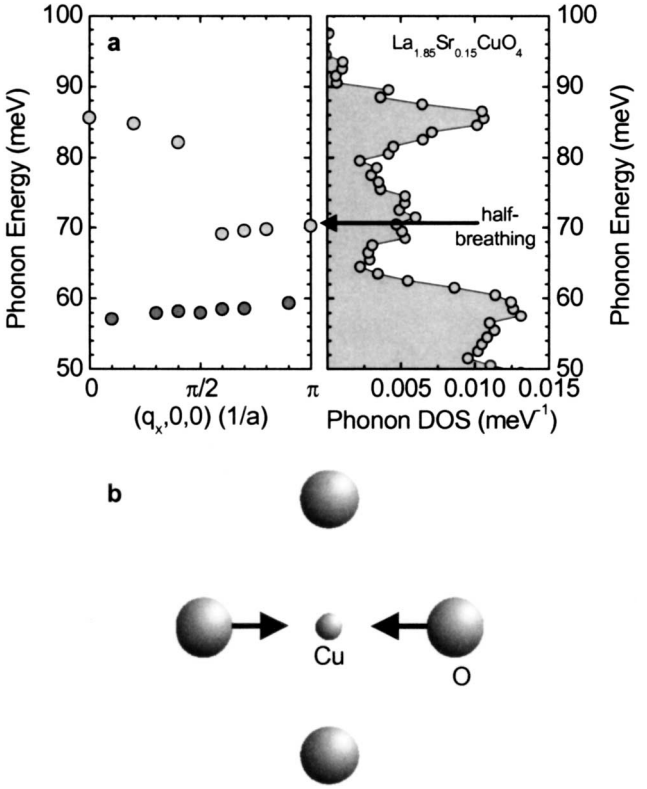

FIG. 2. Half-breathing mode in dispersion and DOS measurements. (a) The high-frequency phonon dispersion of superconducting $La_{1.85}Sr_{0.15}CuO_4$ along $(1, 0, 0)$ is shown in the left panel (from McQueeney *et al.* [10]). The flat mode originating at $(\pi, 0, 0)$ and ~70 meV is the half-breathing oxygen mode. The lower branch at 60 meV is the bond-bending branch. On the right, the high-frequency portion of the DOS of $La_{1.85}Sr_{0.15}CuO_4$ is shown. The ~70 meV band is comprised at least partly of the anomalous half-breathing modes. (b) Polarization of the half-breathing oxygen mode.

the metallic state of high-temperature superconductors that is not present in the insulating state. The MIT also affects lower-energy phonons, especially near 30 meV. Unfortunately, a plethora of phonon modes existing at this energy make analysis of the low-energy DOS features difficult, and they are not discussed further.

The new lattice modes at 70 meV consist, at least partially, of half-breathing-like oxygen phonon modes that propagate in the CuO_2 plane. Phonon dispersion measurements of LSCO by inelastic neutron scattering show that the half-breathing oxygen modes between $\mathbf{q} = (\pi/2, 0, 0)$ and $(\pi, 0, 0)$ (in units of $1/a$, where a is the Cu-Cu nearest-neighbor distance) soften anomalously from 80 meV ($x = 0$) to 70 meV ($x = 0.15$) with doping [5,8]. Figure 2(a) demonstrates clearly that this flat half-breathing branch contributes to the DOS as a van Hove singularity for $La_{1.85}Sr_{0.15}CuO_4$ near 70 meV. The half-breathing polarization is shown in Fig. 2(b) [7].

To learn more about the nature of the electron-lattice coupling of the half-breathing modes, we use a simple lattice dynamics model to isolate an effective interaction being mediated by the mobile holes. The modes we are concerned with propagate in the CuO_2 plane. Also, vibrations in the energy range of interest (above 60 meV) are comprised entirely of in-plane polarized oxygen modes. Therefore, a two-dimensional harmonic force constant (ball and spring) model of the CuO_2 plane can be used to reproduce the high-frequency DOS. The initial set of pairwise force constants is chosen to reproduce the measured in-plane phonon dispersion and DOS of undoped La_2CuO_4. Doping is introduced parametrically by varying the magnitude of the force constants while maintaining the periodicity of the CuO_2 plane. Models studied with spatially varying force constants (superlattice models) produced only small variations in the DOS. The model reproduces well the subtle changes observed in the high-frequency oxygen phonons within the insulating phase ($x < 0.06$), such as the gradual weak softening of the

~88 meV phonon band, by reducing the Cu-O nearest neighbor force constant. This is shown in Fig. 3(a).

The model has difficulty reproducing the large changes of the oxygen phonons in the metallic state at $x = 0.08$. One is required to introduce a repulsive force constant between next-nearest neighbor oxygens with a bridging copper to produce a band near 70 meV originating from the half-breathing mode. More importantly, this repulsion must be introduced anisotropically between only one oxygen pair in a given CuO_4 plaquette (not the orthogonal pair), thereby breaking the crystal symmetry (although perhaps only locally and over phonon time scales). Introducing the repulsion over both pairs will soften oxygen breathing modes (near 85 meV) in addition to the half-breathing modes, inconsistent with experimental results. The calculated and measured differences between the $x = 0.06$ and 0.08 DOS are shown in Fig. 3(b) and results of the full DOS for $x = 0$, 0.06, and 0.08 are shown in Fig. 3(c). This simple model suggests that the electron-lattice coupling is one dimensional in nature and the phonons couple to hole states that are oriented along the Cu-O-Cu bond direction.

However, differences between the metallic and insulating DOS cannot be explained fully with the simple model. In the data, the intensity of the 70 meV band is relatively large implying that many phonons (~15% of all possible planar oxygen modes) are affected. The metallic phonon bands are also narrower in energy than the insulating bands, which suggests phonon localization. The sharpness and intensity of the metallic bands are consistent with phonon dispersion measurements in LSCO [10] where the half-breathing-like modes are observed to be dispersionless in a large region of the Brillouin zone between $\pi/2a < |q_x| < \pi/a$, $|q_y| < 0.3\pi/a$, and independent of q_z [10]. This entire Brillouin zone pocket centered at $(\pi, 0, 0)$ forms the rather intense van Hove singularity in the DOS at 70 meV. These observations cannot arise from the usual electron-phonon coupling in a metal which causes Kohn anomalies in the phonon dispersion at specific wave vectors $\mathbf{q} = 2\mathbf{k}_F$ (where $\mathbf{k}_F$ is the Fermi surface wave vector).

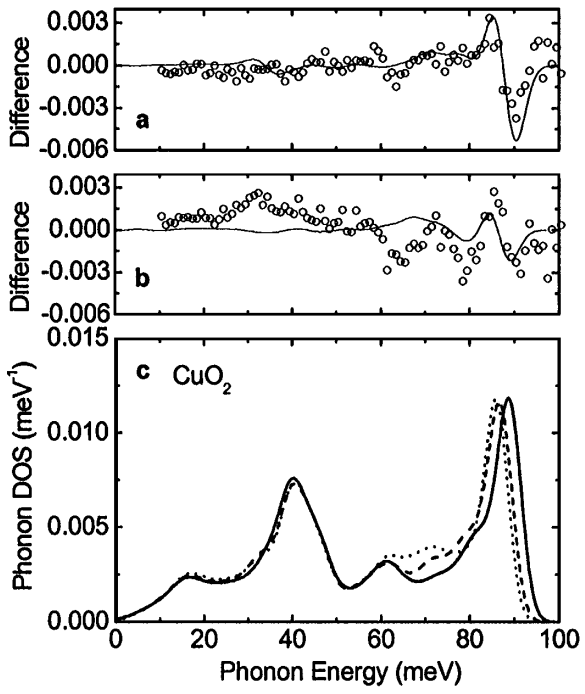

FIG. 3. Comparison of data to two-dimensional model DOS calculations. (a) The difference of the two model DOS calculations for $x = 0.06$ and $x = 0$ (line) and the difference in the data (circle). (b) The difference of the two model DOS calculations for $x = 0.08$ and $x = 0.06$ (line) and the difference in the data (circle). (c) The calculated CuO_2 phonon density of states for $x = 0$ (full line), $x = 0.06$ (dashed line), and $x = 0.08$ (dotted line).

The presence of strong electron-lattice coupling should be corroborated by angle-resolved photoemission spectroscopy (ARPES). Recent ARPES results in the $Bi_2Sr_2CaCu_2O_8$ superconductors show that the electronic band dispersion has a kink due to mixing of holes with bosonic excitations in the range of oxygen optical phonon energies (50–80 meV) [11–13]. Lanzara *et al.* have also observed similar kinks at ~70 meV in ARPES data from the LSCO system [14]. Many scenarios have been discussed with regard to the origin of the kink, such as the coupling of the electrons to phonons, to magnetic excitations (in particular, the magnetic resonance mode observed by neutron scattering [18]), or to collective charge fluctuations which appear below T_c. Lanzara *et al.* argue that strong electron-lattice coupling is the root cause mainly because the kinks are seen in both normal and superconducting states and the magnetic resonance mode has not been observed in LSCO. However, the existence of the kink above T_c, the full energy range of the renormalized electronic spectrum, and consequently the origin of the kink itself, are still subjects of debate [11–14,19,20]. The results in this Letter seem to lend tantalizing support to a phonon scenario, as the energy of the strongly coupled half-breathing phonon modes corresponds precisely to the kink energy for LSCO.

Ino *et al.* [21] have studied the doping dependence of the electronic structure in $La_{2-x}Sr_xCuO_4$ by the ARPES technique and a direct comparison can be made with the doping dependence of the phonons. The ARPES results reveal that the $(\pi, 0, 0)$ electronic saddle point sits ~500 meV below the Fermi level in the insulating phase. Hole doping transfers spectral weight close to the Fermi level, eventually producing a MIT at $x \sim 0.05$ where charge dynamics are characterized as one dimensional (the one dimensionality of the charge dynamics in LSCO observed by ARPES is also discussed by Zhou *et al.* [22]). The maximum spectral weight of the saddle point band crosses the half-breathing phonon energy (70–80 meV) near $x = 0.07$ consistent with new phonon modes being observed somewhat above the critical hole concentration of the MIT as measured by transport. This origin of this

behavior is unclear, but may arise from the disparity in the time scales of inelastic neutron scattering and transport measurements.

From ARPES, the low-energy one-dimensional charge fluctuations are shown to originate from states in the extended electronic saddle point near $\mathbf{q} = (\pi, 0, 0)$. This is the same region in reciprocal space where the anomalous phonon modes exist. Thus, both half-breathing-like phonons *and* saddle point holes can be thought to form localized states (or "wave packets" made up of many plane waves). This result lends some support to the stripe model where it is assumed that the metallic state is inhomogeneous with a localized charge distribution [4,5]. Castro Neto has shown that strong phonon and electronic responses at $(\pi, 0, 0)$ occur in the stripe model from the scattering of hole pairs at stripe domain walls [23]. Another source of charge inhomogeneity originates from large Peierls-type (phonon induced) charge transfer fluctuations. The introduction of such charge fluctuations into electronic structure calculations within the local-density approximation is known to produce strong screening of the half-breathing phonon modes in La_2CuO_4 [24]. Results obtained from exact diagonalization of the one-dimensional Peierls-Hubbard model also indicate that charge transfers caused by half-breathing modes are enhanced when strong correlations are included [25]. These results suggest that anomalous phonons in the metallic state may be due to a strong local interaction of phonons with charge fluctuations above the metal-insulator transition. This important interaction bears consideration in any theory of high-temperature superconductivity.

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