Mixed Lattice and Electronic States in High-Temperature Superconductors

R. J. McQueeney*, J. L. Sarrao, P. G. Pagliuso
Los Alamos National Laboratory, Los Alamos, New Mexico 87545

P. W. Stephens
Department of Physics and Astronomy, State University of New York at Stony Brook, Stony Brook, NY 11794 USA

R. Osborn
Argonne National Laboratory, Argonne, Illinois 60439

(Received on April 6, 2001)

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$_x$CuO$_4$. 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.

PACS numbers: 74.25.Kc, 63.20.Kr, 71.30.+h, 74.20.Mn

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. Much effort has been focused on the role of antiferromagnetic spin fluctuations in transport properties and superconductivity in the metallic state, mainly because it is believed that superconducting transition temperatures are too high to arise solely from electron-phonon coupling. Recently, this correlated metallic state has been discussed in terms of localized, atomic-scale charge density fluctuations. As hole doping affects mainly the hybridized Cu 3d$_{x^2-y^2}$ and O 2p(x,y) anti-bonding electronic states, the lattice should couple strongly to a localized matrix of slow charge fluctuations, perhaps decisively influencing charge transport and superconducting properties. Inelastic neutron scattering measurements of the lattice dynamics do show evidence of strong and unusual electron-lattice coupling in high-$T_c$ compounds with various crystal structures and methods of doping. However, many issues surrounding this anomalous coupling remain unclear. Of course, the ultimate issue concerns its role in superconductivity. Here, we report the systematic development of these anomalous modes on hole concentration and the abrupt formation of new oxygen lattice vibrations near the metal-insulator transition (MIT) in La$_{2-x}$Sr$_x$CuO$_4$. These new lattice modes may be associated with the bosons which interact with electronic states, causing a kink in the electronic dispersions observed by photoemission. These results are the best experimental evidence yet that the lattice is strongly mixed with charge dynamics in the high-temperature superconductors.

We performed inelastic neutron scattering measurements of phonon densities of states (DOS) in La$_{2-x}$Sr$_x$CuO$_4$ (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$_x$NiO$_4$. 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 in orthorhombic at $T = 10$ K. The new 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. Rather, the abrupt development of the 70 meV band must be related to the doping-induced MIT near $x = 0.05$, where trapped holes begin to become mobile. 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 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 $\vec{q} = (\pi/2, 0, 0)$ and $(\pi, 0, 0)$ soften anomalously from 80 meV ($x = 0$) to 70 meV ($x = 0.15$) with doping. Figure 3(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).

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 the effective interaction being mediated by the mobile holes. We are only interested in the highest energy modes propagating within CuO$_2$ plane. Also, for these vibrational energies (above 60 meV) the phonons 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 is used to reproduce the high-frequency DOS. The initial set of pairwise force constants are chosen to reproduce the measured in-plane phonon dispersion and DOS of undoped La$_2$CuO$_4$. Doping is introduced by varying the magnitude of the force constants while maintaining the full 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 figure 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 oxy-
gen 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 strongly softens oxygen breathing modes (near 85 meV) in addition to the half-breathing modes, inconsistent with experimental results. The calculated and measured differences in the $x = 0.06$ and 0.08 DOS are shown in figure 3(b) and results of the full DOS for $x = 0$, 0.06, and 0.08 are shown in figure 3(c). This simple model suggests that the electron-lattice coupling is one-dimensional in nature and the coupled hole states are oriented along the Cu-O-Cu bond direction.

![Image](image_url)

**FIG. 3.** Comparison to two-dimensional 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), and $x = 0.08$ (dotted).

However, differences between the metallic and insulating DOS cannot be explained fully with the simple model. The intensity of the 70 meV band is relatively large implying that many phonons (≈ 15% of all possible 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 where the half-breathing-like modes are observed to be dispersionless in a large region of the Brillouin zone between $\pi/2a < |q_z| < \pi/a$, $|q_y| < 0.3\pi/a$, and independent of $q_x$. 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 wavevectors $\mathbf{q} = 2\mathbf{k}_F$ (where $\mathbf{k}_F$ is the Fermi surface wavevector).

Strong electron-lattice coupling and the one-dimensionality of the charge states are corroborated by various angle-resolved photoemission results. Bogdanov et al. show that the electronic band dispersion has a kink due to mixing of holes with other bosonic excitations in the range of oxygen optical phonon energies. Lanzara et al. conclusively identify these excitations as phonons. For LSCO, Lanzara et al. also show that strong electron-lattice coupling occurs with phonons in the 70 meV energy range, i.e. the half-breathing modes. Ino et al. have studied the doping dependence of the photoemission in La$_{2-x}$Sr$_x$CuO$_4$. These results reveal that the $(\pi, 0, 0)$ electronic saddlepoint sits $\sim$500 meV below the Fermi level in the insulating phase. Doping moves the saddlepoint close to the Fermi level, eventually producing a MIT at $x \sim 0.05$ where charge dynamics are characterized as one-dimensional. The maximum spectral weight of the saddlepoint 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 behavior is unclear, but may arise from the disparity in the time scales of inelastic neutron scattering and transport measurements.

It is possible that the one-dimensional charge fluctuations originate from states in the extended electronic saddlepoint near $\mathbf{q} = (\pi, 0, 0)$. Then, half-breathing-like phonons and saddlepoint holes form localized states (or “wave packets” made up of many plane waves). The stripe scenario supports this conjecture by assuming an inhomogeneous and localized charge distribution in the metallic state. 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. Another source of charge inhomogeneity originates from large Peierls-type (phonon induced) charge transfer fluctuations. The introduction of charge fluctuations into electronic calculations within the local-density approximation is known to produce strong screening of the half-breathing phonon modes in La$_2$CuO$_4$. 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. These results suggest that the phonon softening in the metallic state is likely due to the 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.
ACKNOWLEDGMENTS

RJM would like to thank T. Egami, A. Castro Neto and A. Ramirez for helpful discussions. This work was supported by the U.S. Department of Energy under contract number W-7405-Eng-36 with the University of California. This work has benefited from the use of the Intense Pulsed Neutron Source at Argonne National Laboratory. This facility is funded by the U. S. Department of Energy, BES-Materials Science, under contract W-31-109-Eng-38.

1 M. A. Kastner, R. J. Birgeneau, G. Shirane, and Y. Endoh, Rev. Mod. Phys. 70, 897 (1998).
2 J. Zaanen and O. Gunnarson, Phys. Rev. B 40, 7391 (1989).
3 J. M. Tranquada, et al., Nature 375, 561 (1995).
4 K. Yamada et al., Phys. Rev. B 57, 6165 (1998).
5 L. Pintschovius et al, Physica(Amsterdam) 185C-189C, 156 (1991).
6 L. Pintschovius and M. Braden, J. Low Temp. Phys. 105, 813 (1996).
7 W. Reichardt et al., Physica(Amsterdam) 162C-164C, 464, (1989).
8 R. J. McQueeney et al, Phys. Rev. Lett. 82, 628 (1999).
9 P. V. Bogdanov et al., Phys. Rev. Lett. 85, 2581 (2000).
10 A. Lanzara et al., (unpublished).
11 R. J. McQueeney, J. L. Sarrao, and R. Osborn, Phys. Rev. B 60, 80 (1999).
12 P. G. Radaelli et al., Phys. Rev. B 49, 4163 (1994).
13 M. Imada, A. Fujimori, and Y. Tokura, Rev. Mod. Phys. 70, 1039 (1998).
14 A. Ino et al., Phys. Rev. B 62, 4137 (2000).
15 A. H. Castro Neto, J. Supercond. 13, 913 (2000).
16 C. Falter, M. Klenner, G. Hoffmann, Q. Chen, Phys. Rev. B 55, 3308 (1997).
17 S. Ishihara, T. Egami, M. Tachiki, Phys. Rev. B 55, 3163 (1997).