Phonon Signature of Charge Inhomogeneity in High Temperature Superconductors

\[ \text{YBa}_2\text{Cu}_3\text{O}_{6+x} \]  

Y. Petrov\(^1\), T. Egami\(^2\), R. J. McQueeney\(^3\), M. Yethiraj\(^4\), H. A. Mook\(^4\), and F. Dogan\(^5\)  

\(^1\)Department of Physics and Astronomy, University of Pennsylvania, Philadelphia, PA 19104, \(^2\)Department of Materials Science and Engineering, University of Pennsylvania, Philadelphia, PA 19104, \(^3\)Los Alamos National Laboratory, Los Alamos, NM 87545, \(^4\)Oak Ridge National Laboratory, Oak Ridge, TN 37831, \(^5\)Department of Materials Science and Engineering, University of Washington, Seattle, WA 98195  

March 21, 2022

Abstract

Temperature and composition dependences of high-energy longitudinal optical (LO) phonons in YBa\(_2\)Cu\(_3\)O\(_{6+x}\), studied by inelastic neutron scattering measurements, provide clear evidence for temperature dependent spatial inhomogeneity of doped charges. The measurements indicate that charge doping increases the volume fraction of the charged regions, while the local charge density remains unchanged. For the optimally doped sample the charge distribution changes sharply near the superconducting transition temperature, with stronger charge inhomogeneity below T\(_C\). The remarkable magnitude of the phonon response to charge suggests strong involvement of phonons in charge dynamics.

PACS No. 74.25.Ke, 63.20.Kr, 74.20.Mn

For some time, high-temperature superconductivity in cuprates has been believed to occur in a homogeneous system through a magnetic mechanism. However, these assumptions are seriously challenged by recent experimental observations that suggest spatial charge inhomogeneity and lattice effects. In particular, the observation of the spin/charge stripe structure for non-superconducting La\(_{1.475}\)Nd\(_{0.4}\)Sr\(_{0.125}\)CuO\(_4\) has eloquently explained the observed incommensurate magnetic and lattice diffraction. In superconducting samples, the incommensurate periodicity is observed only in the dynamic spin structure. The dynamic periodicity changes almost linearly with composition and is consistent with the static superlattice periodicity for the non-superconducting composition. Thus, it was postulated that dynamic stripe correlations exist in superconducting cuprates, and various theories have been advanced assuming such correlations. However, direct evidence of a lattice signature of the charge inhomogeneity is incomplete. While our earlier work on the LO
phonons in La$_{1.85}$Sr$_{0.15}$CuO$_4$ (LSCO) implicated charge inhomogeneity \cite{7}, only one composition was studied. Phonon anomalies were recently observed for YBa$_2$Cu$_3$O$_{6+x}$ (YBCO, x = 0.2, 0.35, and 0.6) \cite{8}, but anomalies are rather weak, and temperature dependence was only cursorily characterized. In this report, we describe the results of neutron inelastic scattering measurements of high-energy LO phonons in superconducting YBCO (x = 0.2, 0.35, 0.6, and 0.93) that not only provide strong evidence for temperature dependent charge inhomogeneity, but also clarify important features of the charge inhomogeneity. The results indicate that the local charge density of the charged region remains constant with doping, even in the optimally doped sample, and doping only changes the volume fraction of the charged region. The size of the charged region is microscopic, and for the optimally doped sample the charge separation is significantly enhanced in the superconducting phase.

The studied YBCO samples were large single crystal disks with a height of about 1 cm and diameter ranging from 5 cm for the optimally doped sample (x = 0.93) to 4 cm for the strongly underdoped sample (x = 0.2). Measurements were carried out with the HB-3 triple axis spectrometer at the High Flux Isotope Reactor at Oak Ridge National Laboratory. To monochromatize incident neutrons a beryllium (101) reflection was used, while a pyrolitic graphite (002) reflection (Silicon (111) for x = 0.6 sample) was used for an analyzer, set to give a final neutron energy of 14.7 meV. The angular divergence of the beam was 48'-40'-80'-120' for large x = 0.93 crystal and 48'-60'-80'-240' for smaller x = 0.2, 0.35 and 0.6 crystals. As in Ref. 7 we focused on the LO mode along the in-plane Cu-O bond direction which is polar at the zone-center and half-breathing at the zone-edge. Inelastic neutron scattering measurements were made at energy transfers from 50 - 80 meV and momentum transfers, Q, along the (100) direction from (3, 0, 0) to (3.5, 0, 0) in the unit of the reciprocal lattice vector (\(a^* = 1.63 \text{ Å}^{-1}\)). Thus, the measurement only detects longitudinal optical (LO) phonons. Since the samples are twinned, a-axis and b-axis phonons are observed at the same time.

The inelastic neutron scattering intensity pattern at T = 10 K for various doping levels is shown in Fig. 1. In this system the undoped (x = 0) sample has a dispersionless LO branch at 75 meV, while doping softens the zone-edge mode down to 55 meV \cite{9}. Thus one might expect continuous softening at the zone-edge as doping level is increased. Instead, Fig. 1 shows that the LO branch is always split into two, the high-energy branch around 75 meV and the low-energy branch around 55 meV. Instead of continuous softening the spectral weight is transferred from the high-energy branch to the low-energy one as doping is increased. This result is most naturally understood in the two-phase picture if we associate the high-energy and low-energy branches with the micro-phases possessing low and high charge densities, respectively. When the charges are segregated into microscopic domains, increasing the doping level does not change the local charge density in the domains, but simply increases their total volume fraction, causing the spectral weight transfer from the high-energy branch to the low-energy one. The characteristic Q-dependence of the two branches indicates that the size of the charged domain is microscopic, since otherwise two branches
with a similar $Q$-dependence in intensity will be observed.

For the optimally doped sample the inelastic magnetic neutron scattering is dominated by the resonance at 41 meV, so that the evidence of incommensurate magnetic modulation is weak. The present data, however, indicate very clear and strong charge inhomogeneity for the $x = 0.93$ sample, suggesting that the magnetic modulation may not be the dominant force to produce charge inhomogeneity. This is in accord with the observation that the charge signature sets in at a higher temperature than the spin signature does for the sample with the 1/8 charge density, implying that charge is driving the stripe ordering.

The dispersion shown in Fig. 1 agrees with the earlier result for the optimally doped YBCO, and has a striking resemblance to the one obtained earlier for LSCO.

Fig. 2 shows the intensity pattern for the $x = 0.93$ sample obtained at room temperature, $I_{300K}$, and the difference, $I_{300K} - I_{10K}$. At room temperature the two branches are more connected at the middle, in agreement with the result on LSCO. In order to study the temperature dependence in more detail the scattering intensity as a function of energy transfer was measured at $(3.25, 0, 0)$ at various temperatures. The average intensities over the energy range from 56 to 68 meV (Range 1), $I(1)$, and from 51 to 55 meV (Range 2), $I(2)$, are plotted in Fig. 3(a), and the difference, $I(2) - I(1)$, in Fig. 3(b). It is clear that a sharp change is observed near the superconducting transition temperature of $T_C = 95$ K. Above $T_C$, $I(2) - I(1)$ is practically constant. This indicates that the

Figure 1: Composition dependence of the inelastic neutron scattering intensity from YBCO single crystals with $x = 0.2, 0.35, 0.6$ and $0.93$, at $T = 10$ K.
spectral weight is shifted from Range 2 to Range 1 as temperature is raised up to $T_C$, with no further change above $T_C$. Since for the optimally doped YBCO $T_C$ coincides with the pseudo-gap temperature $T^*$, it is not clear whether the change is associated with $T_C$ or with $T^*$. Recent reports on the isotope effect on $T^*$ [12, 13] lead us to think the latter is more likely. Our preliminary study on $x = 0.35$ indicates gradual change and saturation around 200 K, which is close to $T^*$. An objection was raised recently regarding the temperature dependence in LSCO [14], this issue involves the problem of focusing of the spectrometer, and will be discussed elsewhere. For YBCO the presence of strong temperature dependence is unquestionable as presented here.

The results described here represent the most convincing evidence obtained so far on the spatial inhomogeneity of the charged state in the superconducting cuprates, including the optimally doped sample. While the pattern of inhomogeneity is not directly discernible from the present result, a simulation indicates that a quasi-static stripe pattern is not compatible with the split dispersion, and the cell-doubling model exhibits a better fit [7]. But the shape of the domains is probably stripe-like, since the size of the domain speculated from the shape of the dispersion-less portion of the phonon dispersion is $8 \times 20 \, \text{Å}$ [7]. Thus it is possible to characterize the charged domains as fragments of stripes. The fact that greater charge inhomogeneity was observed below $T_C/T^*$ implies that charge inhomogeneity does not compete against superconductivity, but may
facilitate its occurrence. As shown in Fig. 4, the half-breathing mode at the zone-edge results in charge transfer between Cu and O, while the polar mode at the zone-center does not. Thus the zone-edge mode is expected to couple strongly to the doped holes in a charge transfer system such as the cuprates [15, 16, 17]. Indeed the two branches are remarkably different in energy (15 - 20 meV), suggesting strong involvement of the low-energy phonon branch in charge dynamics.

Such microscopic inhomogeneity was observed for manganites that show colossal magnetoresistance (CMR), even in the metallic phase [18, 19]. Also strong phonon softening similar to the one shown here was observed for manganites [20], suggesting intimate involvement of phonons in producing charge segregation. At low doping levels charges in manganites are localized as spin/lattice polarons or polaron aggregates, but as the doping level is increased the polaron aggregates percolate to produce an insulator-to-metal transition [18, 19]. In the metallic phase, charges in the percolating charged domains are not localized, but still spatially restricted. It is possible that a similar picture applies to
cuprates, probably with a smaller length scale. While the details of the charged domains and the pairing mechanism are still unclear, the present results strongly challenge conventional wisdom concerning the origin of high-temperature superconductivity.

The authors are grateful to A. R. Bishop, V. J. Emery, L. P. Gor’kov, A. Bussmann-Holder, and V. Kresin for useful discussions. The research at University of Pennsylvania was supported by the National Science Foundation through DMR96-28136. Measurements were made at Oak Ridge National Laboratory, which is managed by Lockheed Martin Energy Research under contract no. DE-AC05-96OR22464 for the Department of Energy.

References

[1] J. M. Tranquada et al., Nature (London) 375, 561 (1995).

[2] S. W. Cheong et al., Phys. Rev. Lett. 67, 1791 (1991).
[3] K. Yamada et al., Phys. Rev. Lett. 75, 1626 (1995).

[4] S. A. Kivelson, E. Fradkin and V. J. Emery, Nature 393, 550 (1998).

[5] J. Zaanen, Science 286, 251 (1999).

[6] J. Eroles, G. Ortiz, A. V. Balatsky and A. R. Bishop, Europhysics Lett., in press.

[7] R. J. McQueeney, Y. Petrov, T. Egami, M. Yethiraj, G. Shirane and Y. Endoh, Phys. Rev. Lett. 82, 628 (1999).

[8] H. A. Mook and F. Dogan, Nature 401, 145 (1999).

[9] L. Pintschovius, et al., Physica 185-189, 156 (1991).

[10] H. A. Mook, et al., J. Phys. Chem. Solids 59, 2140 (1998).

[11] J. M. Tranquada, et al., Phys. Rev. B 54, 7489 (1997).

[12] D. Rubio Temprano, J. Mesot, A. Furrer, K. Conder, H. Mutka and K. A. Müller, unpublished.

[13] A. Lanzara, G. Zhao, N. L. Saini, A. Bianconi, K. Conder, H. Keller and K. A. Müller, J. Phys.: Cond. Matter, in press.

[14] L. Pintschovius and M. Braden, Phys. Rev. B, 60, R15039 (1999).

[15] S. Ishihara, T. Egami and M. Tachiki, Phys. Rev. B 55, 3163 (1997).

[16] Y. Petrov and T. Egami, Phys. Rev. B 58, 9485 (1998).

[17] Y. Petrov and T. Egami, cond-mat/9912449.

[18] D. Louca, T. Egami, E. L. Brosha, H. Röder and A. R. Bishop, Phys. Rev. B 56, R8475 (1997).

[19] A. Moreo, S. Yunoki and E. Dagotto, Science 283, 2034 (1999).

[20] Reichardt W, Braden M, Physica B 263, 416 (1999).