# Low Temperature Anomaly of LO Phonons in $La_{1.85}Sr_{0.15}CuO_4$ and $YBa_2Cu_3O_7$

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**Abstract.** Inelastic neutron scattering measurements of $La_{1.85}Sr_{0.15}CuO_4$ and $YBa_2Cu_3O_7$ show that the dispersion of the high energy LO phonon mode along the (1,0,0) direction is strongly temperature dependent, and at low temperatures develops an anomalous feature indicative of dynamic cell-doubling. The anomaly does not change through superconducting transition and gradually disappears between 50 and 250 K in LSCO. Possible implications are discussed.

## INTRODUCTION

It has long been assumed that the phonons play little or no role in the high temperature superconductivity of cuprates, since the isotope effect is small and the normal state properties show no evidence of strong electron-phonon coupling [1,2]. However, a large number of experimental observations suggest significant lattice involvement [3], including our recent inelastic neutron scattering measurements of the LO phonons in LSCO [4]. We found that the normal continuous dispersion observed at room temperature was replaced, at low temperatures, with a dispersion that was discontinuous at the half-way point of the Brillouin zone in the [1,0,0] direction. More recently we observed basically the same behavior in the optimally doped YBCO. In this paper we review these results and discuss their implications.

## EXPERIMENTAL RESULTS

Inelastic neutron scattering measurements were carried out with the triple-axis spectrometer at the High Flux Isotope Reactor at Oak Ridge National Laboratory

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on single crystals of $La_{1.85}Sr_{0.15}CuO_4$ ($T_C$ = 37 K) and $YBa_2Cu_3O_{6.93}$ ($T_C$ = 95 K). We used a beryllium crystal (002) as the monochromator and pyrolitic graphite (002) as the analyzer which was set to give a final energy of 14.87 meV. In order to achieve the energy transfer up to 90 meV we had to use the incident neutron spectrum far in the epithermal region. Thus the flux was low, resulting in low counting rates of only 1 - 5 counts/minute. Also we had to identify and stay away from numerous spurious peaks, for instance those involving Bragg scattering from the sample and inelastic scattering from the analyzer.

Figure 1 describes the phonon dispersion for the highest energy LO mode in the [$Q_x$, 0, 0] direction in the tetragonal index (parallel to the Cu-O bond in the CuO2 plane) for LSCO at T = 10 K and room temperature [4]. These LO modes involve almost exclusively the displacements of in-plane oxygen ions within the CuO2 plane. The dispersion at 300 K is largely in agreement with the previous room temperature measurements on $La_{1.9}Sr_{0.1}CuO_4$ [5]. The dispersion is strongly dependent upon temperature, and at 10 K it splits into two parts. The low-energy branch is well defined and nearly dispersionless down to (3.25, 0, 0), but then becomes very wide and diffuse. The high-energy portion is close to the dispersion in the undoped sample. Thus in the range from $Q_x$ = 3.1 - 3.4, two peaks are observed in the constant Q energy scan, one strong and the other weak. The temperature dependence apparently is not related to superconducting transition at $T_C$: No appreciable change was observed between 10 K and 50 K. Similar results were obtained for YBCO as shown in Fig. 2 as intensity contour plots. The temperature dependence in YBCO appears to be weaker than in LSCO, but is much stronger than anticipated for the normal thermal effect due to anharmonicity.

## DISCUSSION

The dispersion of the oxygen LO mode presented here shows two remarkable features. One is the apparent large discontinuity of dispersion around (0.25, 0, 0) at 10 K, and the other is the strong temperature dependence. The apparent discontinuity in dispersion was observed also for $Ba_{0.6}K_{0.4}BiO_3$ [6], nickelate [7] and more recently in doped CMR manganite [8]. Since the softening of the oxygen LO phonons at the zone edge is clearly related to the presence of the doped holes [5,9,10], it may be argued that the discontinuity at (0.25, 0, 0) must also be intimately connected with the presence of charges. Note that this mode, through the change in the Cu-O bond distance, induces charge transfer between Cu and O. We will first consider the possibility that this discontinuity is related to the spin/charge stripes, and discuss other possibilities.

### A. Relation with the spin/charge stripes

While initially the behavior of cuprates has been discussed assuming perfect periodicity in the lattice, a large number of observations point to the possibility of spatially inhomogeneous lattice and electronic structure including some sort of spin/charge phase separation [3,11–15] . More recently the observation of the spin/lattice stripes in the non-superconducting cuprates with the charge density of x = 1/8 [16] strengthened this view. While static stripes were observed for non-superconducting cuprates, it has been assumed that similar stripes exist even in the superconducting phase but they are dynamic, since similar incommensurate magnetic scattering has been observed only by inelastic neutron scattering [17].

The magnetic periodicity of our LSCO sample as determined by inelastic magnetic scattering is approximately $8a$ [18]. This would result in the charge periodicity of 4a in the stripe model, with the corresponding superlattice periodicity of (0.25, 0, 0). However, such a charge density modulation would create a new Brillouin zone boundary and a gap in the phonon dispersion at (0.125, 0, 0), and indeed a simple spring model suggests that should be the outcome, which does not agree with the observation.

A more plausible scenario is that the Kohn anomaly is responsible to the observed anomaly. For instance if the stripe periodicity of $4a$ is related to the Fermi momentum [19,20] by $2k_F = 0.25$, the screening function will have a singularity at q = 0.25. This could result in sharp decline in the dispersion. However, it does not explain why two modes are seen at the same Q at low temperatures.

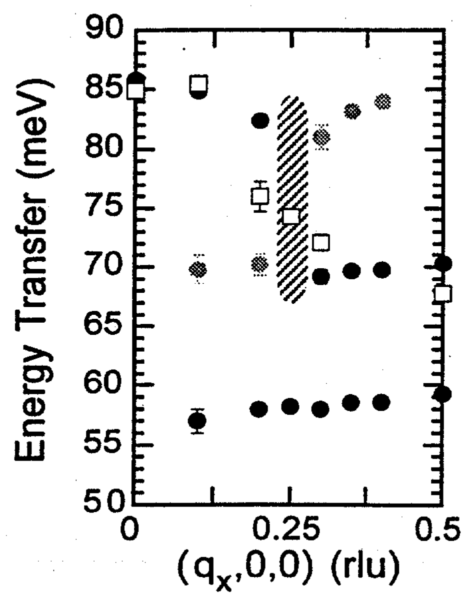


**FIGURE 1.** The dispersion relation of the high energy longitudinal optic branches of $La_{1.85}Sr_{0.15}CuO_4$ along ($Q_x$, 0, 0) at T = 10 K (circles) and room temperature (squares). The shadowed circles indicate the energy of the weak extra branches. The shade at (3.25, 0, 0) indicates a broad peak in the intensity [4]. The low energy branch (57 - 60 meV) is shown just as a reference, and is not related to the discussion in this paper.

## B. 2nd CDW scenario

The discontinuity of dispersion at (0.25, 0, 0) is indicative of cell-doubling that creates a new Brillouin zone boundary at (0.25, 0, 0). Indeed a simple spring model with the periodicity of $2a$ reproduces the observed dispersion remarkably well [4]. Such cell-doubling can result from charge ordering (CDW) on oxygen in the $CuO_2$ plane with the periodicity of $2a$ [4]. A similar model with the charges on Cu does not reproduce the observed dispersion at all. It should be noted, however, that this charge ordering, if it exists, is most likely dynamic, since a superlattice diffraction at (0.5, 0, 0) corresponding to such ordering has never been reported, including our own search, in the elastic or quasi-elastic scattering. The charge ordering also is most probably short range, since the absence of dispersion of the 70 meV mode suggest localization. The flat dispersion extends over the range, $0.25 < q_x < 0.75$ and $-0.1 < q_y < 0.1$, suggesting that the coherence lengths of the localized phonon to be $5a$ x $2a$, or 20 x 8 in the $a$-$b$ plane. It is interesting to note that the same coherence lengths were detected in $YBa_2Cu_4O_8$ by the pulsed neutron pair-density function (PDF) study [21]. The possibility of a charge density wave (CDW) being involved in high-temperature superconductivity has been suggested by several authors [22–24]. The conflict between this CDW and the stripe state

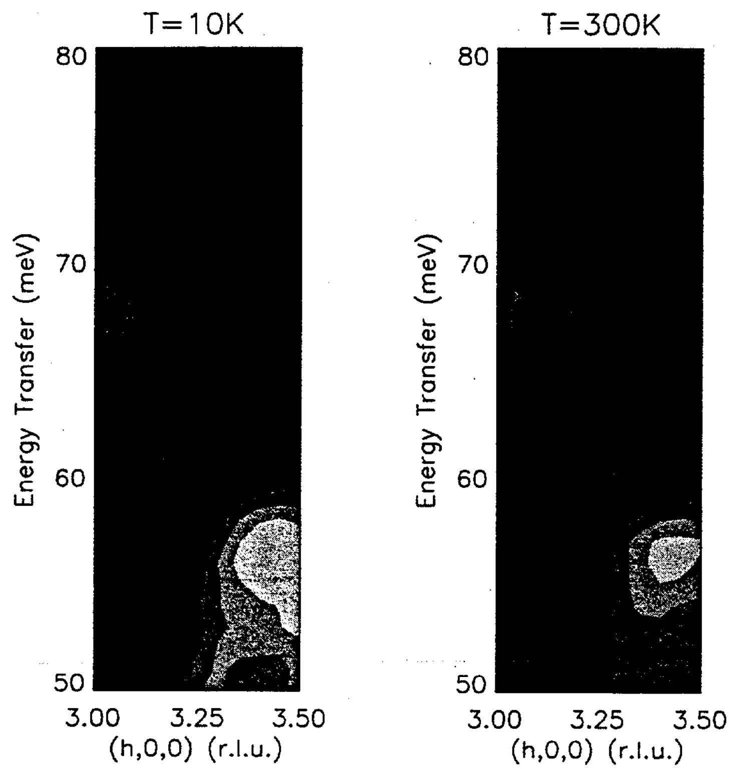


**FIGURE 2.** Intensity contour plot for the LO phonon branches of $YBa_2Cu_3O_{6.93}$ at T = 10 K (left) and at room temperature (right).

may be preventing either from developing into static long range order. It is possible that the LO phonons are resonantly coupling the two CDW states, producing the vibronic ground state [23].

A possible origin of the dynamic charge density modulation the Hubbard-Peierls instability [10,25]. Since the Cu band is nearly half-full, the Peierls instability can soften the zone-edge mode. Note that the q = 0 (zone-center) LO mode results in modulation of charges (CDW) on Cu, while the q = 0.5 (zone-edge) LO mode produces ferroelectric polarization. Charge transfer in the former case (Cu to Cu) is easier than in the latter (Cu to O), since the latter involves a large charge transfer gap (~2 eV). In this scenario the wavevector of the phonon anomaly remains nearly 0.5 regardless of the amount of doping, however, there are indications that it may change with doping [26]. It is important to study the composition dependence of the phonon anomaly. Within a few months we plan to study the phonon dispersion of an underdoped YBCO with $T_C$ of 60 K. It is equally important to confirm dynamic charge ordering more directly. In addition to the inelastic neutron scattering study at low energy transfer range, we are planning to carry out high-resolution electron-energy-loss-spectroscopy (EELS) measurements to study the charge dynamics in the system.

## C. Pair-scattering scenario

Another possibility is that the anomaly is caused by the phonon scattering involving electron pairs. While the backscattering of phonons by a single electron (hole) in the stripe state will result in momentum transfer of 0.25, the backscattering by a pair of electrons can create momentum transfer of 0.5 and a pseudo-Brillouin zone boundary at 0.25. In the BCS theory a Cooper pair is made of carriers with the opposite momenta, $k$ and $-k$, resulting in zero total momentum. However, the electrons that form the stripe state with the momentum of 0.125 are heavy carriers in the extended saddle point (M point) [19,20], and their ground state is the standing wave. Thus the pair can have 0 or $2k$ total momentum. Such a pair-scattering is obtained from the Frölich Hamiltonian through the usual canonical transformation by preserving the pair creation and annihilation terms, for instance, in the Hartree-Fock approximation. If this is the case our observation will provide a direct evidence of the strong phonon involvement in pairing.

The observation of similar discontinuity in nickelates and manganites may appear to weaken the argument on direct connection to superconductivity, but their cases involve samples with twice the charge density and half the stripe periodicity [7,8], and thus may be due to single carrier scattering. It should be noted that the energy of the carriers in the extended saddle point is of the order of 100 meV or less. Thus electrons and phonons are likely to be resonantly coupled also to form vibronic states [23,27]. For such a system the adiabatic approximation would not be valid, making the theoretical treatment difficult [28]. It may be argued that such a strong electron-phonon coupling is incompatible with the high normal state conductivity, but the conductivity is dominated by the light carriers in the vicinity of the X ([0.25, 0.25]) point, and the contribution from the M point may be totally masked. Regardless of the exact origin of the anomaly, the observed phonon dispersion strongly suggests that LO phonons may be intimately involved in the high-temperature superconductivity.

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