Oxygen phonon branches in overdoped La$_{1.7}$Sr$_{0.3}$CuO$_4$

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The dispersion of the Cu-O bond-stretching vibrations in overdoped La$_{1.7}$Sr$_{0.3}$CuO$_4$ (not superconducting) has been studied by high resolution inelastic neutron scattering. It was found that the doping-induced renormalization of the so-called breathing and the half-breathing modes is larger than in optimally doped La$_{1.85}$Sr$_{0.15}$CuO$_4$. On the other hand, the phonon linewidths are generally smaller in the overdoped sample. Features observed in optimally doped La$_{1.85}$Sr$_{0.15}$CuO$_4$ which suggest a tendency towards charge stripe formation are absent in overdoped La$_{1.7}$Sr$_{0.3}$CuO$_4$.

I. INTRODUCTION

It has been known for quite some time that the frequencies of the Cu-O bond-stretching vibrations are strongly renormalized upon doping in all the cuprates investigated so far [1]. The frequency renormalization and a concurrent increase of the phonon linewidths are clear evidence of a strong coupling of these phonon modes to the charge carriers. Renewed interest in this phenomenon was generated by the observation of a kink in the quasi-particle dispersion by angle-resolved photoemission (ARPES) data [2] at an energy corresponding to the bond-stretching vibrations, except for the double peak at $q = 0.5$ below 60 meV which is related to Cu-O bond-bending vibrations.

![Energy scans taken along the line Q = (5-q,0,0) at T = 10 K.](image.png)

FIG. 1: (Color online) Energy scans taken along the line $Q = (5-q,0,0)$ at $T = 10$ K. Successive scans were off-set by 50 counts for the sake of clarity. Lines depict fit curves. The peaks are associated with longitudinal Cu-O bond-stretching vibrations, except for the double peak at $q = 0.5$ below 60 meV which is related to Cu-O bond-bending vibrations.

II. EXPERIMENTAL

The sample consisted of a single crystal of composition La$_{1.7}$Sr$_{0.3}$CuO$_4$ and a volume of about 1 cm$^3$. Its mosaic spread was about 1 degree. The experiments were carried out on the triple-axis spectrometer 1T located at the Orphée reactor using doubly focusing monochromator (Cu220) and analyzer (PG002) crystals. Cu220 was used as monochromator to achieve high resolution. The actual resolution - depending on the focusing conditions - is indicated in the lower panels of Fig. 2. Measurements were carried out both in the 100-001 and in the 100-010 scattering planes. All measurements were performed at $T = 10$ K.

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FIG. 2: (Color online) Upper panels: Dispersion of the longitudinal high-energy phonon branches in the (100)- and in the (110)-directions, respectively, for various doping levels in La$_{2-x}$Sr$_x$CuO$_4$. Lines are a guide to the eye. Data for $x = 0$ and 0.1 were taken from [1]. Data for $x = 0.15$ were taken from [7]. Lower panels: The data points show the line-widths (full width at half maximum) of the phonon peaks and the lines show the experimental resolution. The wavyness of the lines is due to focusing effects. Recent data [25] for $x=0.15$ measured with an improved $q$-resolution in the transverse direction are included as open symbols. The displacement patterns of the zone boundary modes are shown in Fig. 4.

III. EXPERIMENTAL RESULTS

A. Plane-polarized Cu-O bond-stretching modes

Fig. 1 demonstrates the high quality of the raw data. The dispersion and the line-widths of the high-energy longitudinal modes in the (100) and in the (110) directions are depicted in Fig. 2. Obviously, the strong doping-induced frequency changes between undoped and optimally doped LSCO are further enhanced when going to overdoped samples, in particular in the (110) direction. We emphasize that the doping-induced softening is observed only in the longitudinal branches: the transverse branch in the (100) direction was found to be completely flat as for all other LSCO samples investigated previously, and the dispersion of the transverse modes in the (110) direction is even slightly reduced with increasing doping levels (Fig. 3). As was stated previously [6], the pronounced dispersion of the transverse modes in the (110)-direction can be completely accounted for by Coulomb forces and is not indicative of a strong electron-phonon coupling. This view is further corroborated by the narrow line-widths of the transverse modes which are much smaller than those of the longitudinal ones. Examples are shown for zone boundary modes in Fig. 5.

FIG. 3: (Color online) Dispersion of the transverse high-energy phonons in the (110)-direction for various doping levels in La$_{2-x}$Sr$_x$CuO$_4$. Lines are a guide to the eye. Data for $x < 0.3$ were taken from [1]. The displacement pattern of the zone boundary mode is shown in Fig. 4 (middle).

FIG. 4: Displacement patterns of zone-boundary bond-stretching modes. Top: longitudinal mode in the (110)-direction (breathing mode); middle: transverse mode in the (110)-direction quadrupolar mode); bottom: longitudinal mode in the (100)-direction (half-breathing mode). Circle s and full points denote oxygen atoms and copper atoms, respectively. Only the displacements in the Cu-O planes are shown. All other displacements are small for these modes.
ing or quadrupolar character, respectively. The width of the Cu-O bond-stretching mode. The two modes are of breathing or quadrupolar character, respectively. The width of the quadrupolar mode is resolution limited.

FIG. 5: (Color online) Energy scans taken at the zone boundary in the (110)-direction at T = 10 K. The peak in (a) corresponds to the longitudinal and in (b) to the transverse Cu-O bond-stretching mode. The two modes are of breathing or quadrupolar character, respectively. The width of the quadrupolar mode is resolution limited.

B. C-polarized modes

Since previous investigations [1] had shown that a particular c-polarized oxygen mode exhibits a very strong doping-induced softening and acquires simultaneously a massive broadening, we investigated this mode as well on our overdoped sample. This mode has been termed O$_z$-mode by Falter and co-workers [10] because it is a z(=c)-polarized zone boundary (Z-point) mode. These authors predicted a strong electron-phonon coupling of this mode prior to experiment based on the following argument: the displacement pattern is such that all apical oxygen atoms move simultaneously towards the Cu-O planes thereby inducing strong charge fluctuations. In order to see whether the anomalous character of this mode is enhanced or rather reduced on overdoping we performed an energy scan at Q=(0,0,15). This Q-point gives the maximum inelastic structure factor for the O$_z$-mode and the second mode, respectively (Fig. 6). We found that the energy of the O$_z$-mode in overdoped LSCO is practically the same as in optimally doped LSCO, i.e. very low compared to that in the undoped parent compound (48 meV vs. 70 meV). The line-width observed in overdoped LSCO is smaller than in optimally doped LSCO but only slightly so (by about 10%).

A comparison of the neutron results for x=0.30 with the recently published x-ray results [4] for the longitudinal bond-stretching modes in the (100)-direction in La$_{1.85}$Sr$_{0.15}$CuO$_4$ shows in general very good agreement. We note that the results for the q-dependent line-widths depicted in Fig. 3b of the x-ray paper suggest a weak maximum around q = 0.3 which is absent in the neutron data. This seems to be simply a consequence of the fact that the neutron data show a monotonic increase of the line-width towards the zone boundary whereas the x-ray signal was lost in this region of q-space. In a later inelastic x-ray study [8], data were published not only for the (100), but also for some longitudinal modes in the (110) direction. Again, good agreement is found between the x-ray data and the neutron data.

IV. DISCUSSION

The doping-induced frequency renormalization of bond-stretching modes in high-Tc materials has been known for many years. It bears clear resemblance to phonon anomalies found in many conventional superconductors like, e.g., Nb$_3$Sn [9]. Therefore, it is generally seen as evidence of a substantial coupling of the bond-stretching modes to the quasi-particles. Consequently, this issue has been addressed in several theoretical papers. Falter and co-workers start from the electronic band structure calculated within the local density approximation (LDA) [10, 11, 12, 13, 14]. The failure of LDA theory to reproduce the insulating ground-state of the undoped parent compounds is remedied by suitably imposing the long-wavelength limit of the electronic polarizability [14]. In this theory, the screening processes producing the softening are described in terms of charge fluctuations on the outer shells of the ions. The doping dependence of the bond-stretching modes is well accounted for in the range from undoped to optimally
doped LSCO. In particular, the theory correctly reproduces the much stronger softening in the (100)-direction as compared to that in the (110)-direction for underdoped and optimally doped samples. Unfortunately, no predictions were made for overdoped samples.

Other groups have chosen a very different approach to explain the phonon renormalization of the bond-stretching modes upon doping, i.e. using the t-J model and extending it to explicitly include electron-phonon couplings \[ \text{[15, 17]} \]. The correct anisotropy between the \([1,1]\) and the \([1,0]\) directions in optimally doped LSCO has been reported for the first time by Khaliullin and Horsch \[ \text{[15]} \]. As is explained in \[ \text{[15, 16]} \], the rapid drop of the frequency of the half-breathing mode when going from the undoped to the optimally doped compound results from a polaron peak in the electron density fluctuation spectrum \(N(q,\omega)\) being in the same energy range as the bond-stretching phonons. Similar results have been reported recently by Rösch and Gunnarsson \[ \text{[17]} \]. Horsch and Khaliullin have the merit of having used their theory to predict the doping dependence of the frequencies of the half-breathing mode and of the breathing mode up to high doping levels, and that several years prior to the experiments \[ \text{[18]} \]. The agreement between theory and experiment is impressive (Fig. 7).

In this context, we would like to mention that the downward dispersion of the longitudinal Cu-O bond-stretching branches has been successfully predicted by density functional theory for another member of the cuprate family, i.e. \(\text{YBa}_2\text{Cu}_3\text{O}_7\) (O7) \[ \text{[21]} \]. O7 is generally considered to be slightly overdoped. In the (100)-resp. (010)-directions, the calculated dispersion curves agree very well with the experimental ones observed on optimally doped O6.95 \[ \text{[23]} \]. In the (110)-direction, however, the calculated downward dispersion is somewhat stronger than observed in experiment on a highly doped sample \[ \text{[24]} \]. Unfortunately, the doping evolution of the downward dispersion cannot be studied by this theory in its present state because of the failure to describe the insulating state of the parent compounds.

At first glance, the monotonic decrease of the zone boundary frequencies with increasing doping suggests a monotonic increase of the electron-phonon coupling strength. We note, however, that the electron-phonon coupling strength is not directly related to the frequency renormalization but rather to the phonon linewidths. Therefore, the reduction of the phonon linewidths on overdoping is direct evidence that the coupling of the bond-stretching modes to the quasi-particles is indeed weaker in overdoped LSCO when compared to optimally doped LSCO. In particular, the massive line broadening of phonons propagating along (100) for \(q\) values around 0.3 observed for doping values 0.07 < \(x\) < 0.15 is absent in overdoped LSCO (Fig. 8). As has been explained in a recent paper \[ \text{[3]} \] based on results for compounds showing static stripe phase order, the extremely large linewidths observed in the (100) direction at halfway to the zone boundary are very probably related to dynamic charge stripe formation. Another signature of dynamic charge stripe formation discussed in \[ \text{[7]} \] is a very steep slope of the phonon dispersion around \(q = (0.25,0,0)\) leading to a strong deviation from a sinusoidal shape of the phonon dispersion curves (see also Fig. 25 in \[ \text{[24]} \]). In optimally doped LSCO, the maximum slope is not as large as found in compounds showing static stripe order but is still relatively large \[ \text{[24]} \]. In overdoped LSCO, however, the bond-stretching phonon frequencies along (100) follow a cosine behavior very well. Therefore, it can be said that the signatures of dynamic charge stripe formation observed in optimally and in slightly underdoped LSCO \((x = 1/8)\) are completely absent in a strongly overdoped (and non-superconducting) sample. In this context, we would like to point out that the signatures of dynamic charge stripe formation become weak as well when going to the strongly underdoped side of the phase diagram. In particular, the phonon dispersion curve along (100) becomes approximately sinusoidal (see the data for \(x = 0.1\) in Fig. 2 and further data for \(x = 0.07\) in \[ \text{[5]} \]). On the other hand, the linewidths observed in a sample with \(x=0.07\) \((T_c=20\text{ K})\) around \(q = (0.3,0,0)\) are still rather high \[ \text{[5]} \], which means that it is these linewidths which are most closely correlated with the superconducting transi-
FIG. 8: (Color online) Comparison of energy scans taken at $Q = (5.0, 0.3, 0.0)$ on optimally doped [25] and on overdoped LSCO at $T = 10$ K. The data were corrected for background scattering and normalized to yield the same integrated intensity. Both scans were taken with the same experimental set-up. Note that the width observed on the optimally doped sample is somewhat larger than that reported in [7] because of a better momentum resolution in the later experiment.

We note that none of the theories discussed above predicts any of the signatures of dynamic stripe formation observed in experiment. Theory did predict the tendency for charge stripe formation in the cuprates very early [22, 31] but failed to make detailed predictions for the doping dependence of this phenomenon. Qualitatively speaking, stripe formation is related to the correlated nature of the quasiparticles in the Cu-O2 planes.

V. CONCLUSIONS

In summary, we have found that the doping-induced frequency renormalization of bond-stretching modes is enhanced when going from an optimally doped to a strongly overdoped sample. In particular, the renormalization of the longitudinal modes in the (110)-direction becomes quite strong on overdoping, in very good agreement with theoretical predictions. However, the phonon linewidths decrease at the same time indicating that the electron-phonon coupling strength is not enhanced on overdoping. These results mean that superconductivity is not correlated with the general phonon renormalization but rather with the phonon linewidths. More specifically, the massive line broadenings observed around $q = (0.25, 0, 0)$ in LSCO at doping levels close to optimum doping indicative of dynamical charge stripe order are not found in strongly overdoped, non-superconducting LSCO. These linewidths are even smaller than the corresponding ones in strongly underdoped, but still superconducting LSCO.

VI. ACKNOWLEDGEMENTS

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Note that the frequency renormalization plotted in this figure cannot be compared directly to the quantities plotted in the x-ray papers (fig. 4 of [4] resp. fig. 11 of citeikeuchi). The so-called phonon softening shown in the x-ray papers is simply the difference between the zone center and the zone boundary frequencies, whereas the softening plotted in our fig. 7 is calculated with respect to the undoped case. Further, our fig. 7 is based on true zone boundary frequencies and not on a fit of the whole branch with a sine-function. It is basically this fit with a sine-function which produces the local maximum in the softening vs. doping curve around optimal doping in [4] because the dispersion of the LO branch along (100) deviates strongly from a sine-function for optimally doped LSCO but not for overdoped LSCO.

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