Evidence for Dynamic Charge Stripes in the Phonons of Optimally Doped YBCO

L. Pintschovius, Y. Endoh, D. Reznik, H. Hiraka, J. M. Tranquada

Forschungszentrum Karlsruhe, Institut für Festkörperphysik, P.O.B. 3640, D-76021 Karlsruhe, Germany
Institute for Material Research, Tohoku University, Katahira, Aoba-ku, Sendai, 980-8577, Japan
Laboratoire Léon Brillouin, C.E.A./C.N.R.S., F-91191 Gif-sur-Yvette, France
Physics Department, Brookhaven National Laboratory, Upton, NY 11973, USA

Superconductivity Research Laboratory, ISTEC, Shinonome, Koutu-ku, Tokyo 135-0062, Japan

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Inelastic neutron scattering investigations on optimally doped YBCO revealed a very pronounced temperature dependence in the Cu-O in-plane bond-stretching vibrations along the (010) direction: a shift of spectral weight by at least 10 meV has been observed in a narrow range of wave vectors halfway to the zone boundary. The temperature evolution starts at about 200 K, well above the superconducting transition temperature. The displacement pattern of the anomalous phonons indicates a dynamic one-dimensional charge-density modulation within the planes, but excludes a direct contribution from the Cu-O chains.

There is widespread belief that the electronic structure of high temperature superconductors is characterized by an inhomogeneous distribution of charge and spin in which the spins on the copper atoms form antiferromagnetic stripes, separated by domain walls carrying the charge. Although theory [1, 2] suggests that stripe formation should be a universal property of the cuprates, solid experimental evidence for stripes has been found so far only for a particular compound, i.e. La$_2$CuO$_4$Y$_{1-x}$Sr$_x$CuO$_4$ [3, 4], in which the stripes are static and apparently detrimental to superconductivity. In superconducting compounds, the stripes are expected to be dynamic in nature, but dynamic stripes are much more difficult to detect and so the experimental basis in favor of dynamic stripes is far from being solid. Inelastic neutron scattering can be used to observe the dynamic spin structure by the interaction between the neutron spin and the electron spin. Magnetic fluctuations consistent with the stripe phase picture have indeed been observed in the system La$_{2-x}$Nd$_x$Sr$_{0.15}$CuO$_4$ [4, 5], whereas the interpretation of the results for YBa$_2$Cu$_3$O$_{6.95}$-symmetry remains controversial [5, 6]. Inelastic neutron scattering can be used to look for fingerprints of dynamic charge stripes as well, but in this case, evidence for stripes is obtained only indirectly by searching for effects of the dynamic charge structure on the atomic vibrations. That is to say, phonons with a displacement pattern closely related to that resulting from the inhomogeneous charge distribution are expected to show an anomalous behavior somewhat reminiscent of the phonon anomalies observed in one-dimensional conductors at temperatures above the Peierls transition, i.e. as a precursor phenomenon to charge-density-wave formation [7].

In this Letter, we report observations by inelastic neutron scattering on optimally doped YBa$_2$Cu$_3$O$_{6.95}$ ($T_c = 93$ K) consisting of three twinned single crystals with a total volume of 1.5 cm$^3$. Details of the sample are given elsewhere [8]. The neutron experiments were carried out on the 1T triple-axis spectrometer at the ORPHEE reactor of the Laboratoire Léon Brillouin at Saclay, France. The (220) reflection of a Cu crystal was used to monochromatize the incident neutrons in order to achieve high resolution. Pyrolytic graphite (PG) (002) was used as analyzer crystal. Both crystals were horizontally and vertically focusing. A PG filter was placed into the final beam to suppress higher order contaminations. The measurements were performed at various temperatures between T=10 K and 300 K. The experiments focused on the Cu-O in-plane bond-stretching vibrations in the (100) and in the (010) directions because it was known from previous investiga-

We show that a pronounced downward shift of spectral weight occurs for Cu-O in-plane bond-stretching vibrations propagating along the b-direction within a narrow range of wave vectors on cooling below room temperature. This result appears to be difficult to explain other than with the development of dynamic charge stripe correlations. We note that first results on an unusual temperature dependence of phonons in optimally doped YBCO were already reported in [9]. An important step for achieving a better understanding of these effects [10] has been to study the phonon branches of $\Delta_4$-symmetry instead of those of $\Delta_1$-symmetry: these branches differ from each other in that the elongations in the Cu-O bi-layer are in-phase for $\Delta_1$-symmetry and out-of-phase for $\Delta_4$-symmetry. We found that the behavior of the two types of bond-stretching branches is in principle very similar. However, anticrossings of the in-plane-polarized bond-stretching branch with c-axis- polarized branches leads to a formidable complication in the case of the branches of $\Delta_1$-symmetry [11, 12].

Inelastic neutron scattering measurements were performed on a composite sample of optimally doped YBa$_2$Cu$_3$O$_{6.95}$ ($T_c = 93$ K) consisting of three twinned single crystals with a total volume of 1.5 cm$^3$. Details of the sample are given elsewhere [13]. The neutron experiments were carried out on the 1T triple-axis spectrometer at the ORPHEE reactor of the Laboratoire Léon Brillouin at Saclay, France. The (220) reflection of a Cu crystal was used to monochromatize the incident neutrons in order to achieve high resolution. Pyrolytic graphite (PG) (002) was used as analyzer crystal. Both crystals were horizontally and vertically focusing. A PG filter was placed into the final beam to suppress higher order contaminations. The measurements were performed at various temperatures between T=10 K and 300 K. The experiments focused on the Cu-O in-plane bond-stretching vibrations in the (100) and in the (010) directions because it was known from previous investiga-

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FIG. 1: Dispersion relation of the out-of-phase Cu-O bond-stretching vibrations in optimally doped YBCO as seen on a twinned sample. The energies are plotted against the nominal wave vector, which is calculated from an average lattice constant $a_{av} = (a+b)/2$. The actual wavevectors for the scattering from each of the two twin domains differ by $2\delta = 2\pi/a - 2\pi/b$. The arrows show the zone center for phonons propagating along the a or the b direction, respectively. The ellipsoid depicts the instrumental resolution.

The twinning of our sample was, of course, a difficulty for the assignment of the neutron peaks to the a or the b direction. This problem was overcome by the technique described in refs. 9,15, i.e., by exploiting the fact that the dispersion relation for phonons propagating along (100) is shifted in reciprocal space against that of phonons propagating along (010) due to the difference in lattice constants of nearly 2% (see Fig. 1). A further strong support for our assignment came from previous measurements on a detwinned sample [12, 13]. Unfortunately, this sample cleaved after the first cooling below $T_c$. The question as to whether there is a subtle influence of $T_c$ on the temperature dependence of the phonon under discussion will have to be addressed in a future investigation using finer steps in temperature. Fig. 2b further suggests that the phonon softening proceeds by a shift of spectral weight and not a peak shift, i.e., a gradual depletion of the phonon intensity observed around 59 meV at $T=300$ K and a simultaneous buildup of intensity at much lower energies. This issue was further explored by scans at the nominal wave vector $Q=(2.7,0,2)$, corresponding to the same $q$ along $b^*$ as at $Q=(3.25,0,2)$ because focusing leads here to a better energy resolution. Unfortunately, the contributions from the $a^*$ and the $b^*$ directions show up at nearly the same energy at $T=200$ K. On cooling, a pronounced loss in intensity is observed which corresponds very well to that observed at $Q=(3.25,0,2)$. However, no peak broadening was seen at any temperature demonstrating that all the lost intensity is shifted out of the energy window of these scans except for a modest intensity gain at the lowest end of the window (50 meV) (Fig. 3b). We note that this intensity loss rapidly disappears when going to smaller $q$ (Fig. 3a). On the high $q$ side, the temperature effect becomes rapidly weaker as well and leads to a softening of the bond-stretching phonon peak of a few meV only. At the zone boundary, there was no discernible temperature dependence of the bond-stretching mode (Fig. 3c).

The phonon anomaly described above bears certainly some resemblance to the Kohn anomalies observed in one-dimensional conductors [17], i.e., precursor phenomena to a charge-density-wave transition occurring at low temperatures. Therefore, one might conjecture that it is related to flat parts of the Fermi surface related to the oxygen chains [13]. However, we emphasize that the phonon anomaly is observed in a branch of $\Delta_4$-symmetry for which the chain O elongations are zero. This fact excludes a trivial relationship of the anomaly with chain O related electronic states. Having ruled out a direct association with the Cu-O chains, we conclude that the observed anomaly is evidence for a significant electronic anisotropy, consistent with nematic behavior [24].

Could the phonon anomaly be the result of Fermi surface nesting within the Cu-O planes? A check of this possibility is provided by recent results of density functional theory [19]. These calculations indeed predict a local frequency minimum in a branch with bond-stretching character at $q=(0.025,0)$; however, this branch is of $\Delta_4$-symmetry and involves primarily chain O elongations. This feature is not unexpected in view of the one-dimensional nature of the Cu-O chains and may well be related to static charge modulations detected within the Cu-O chains by scanning tunneling microscopy (STM) [25, 26]. On the other hand, the calculations do not predict a similar anomaly in a branch of $\Delta_4$-symmetry. Rather the theoretical results are in surprisingly good

background for such wide energy intervals.
Q corresponds to a reduced wave vector \( q=0.22 \) along \( \mathbf{a}^* \), or \( q=0.28 \) along \( \mathbf{b}^* \), respectively. The 300 K data have been corrected for the increase in multi-phonon scattering on raising the temperature from \( T=10 \) K. Smoothed intensity differences, with respect to the intensity measured at \( T=300 \) K, are shown in Fig. 3(c). The 300 K data have been corrected for the increase in multi-phonon scattering on raising the temperature from \( T=10 \) K. (b) Smoothed intensity difference, with respect to the intensity measured at \( T=10 \) K. (c) Difference between the intensities measured at \( T=10 \) K and \( T=200 \) K, respectively, as measured in an independent run over a larger energy interval.

Agreement with the 200 K data (Fig. 4). The authors of that study were concerned that the high effective electron temperature (0.2 eV) used in their calculations to improve convergence might have suppressed the anomalous softening at \( T=0.25 \). Therefore, additional calculations were made using a much smaller effective electron temperature (0.02 eV). However, this had barely any influence on the calculated phonon frequencies. Apparently, the anomalous low-temperature dispersion of the in-plane bond-stretching phonons cannot be understood within the framework of such a theory. For this reason, it seems unlikely that the observed anomaly can be attributed to a Fermi surface nesting effect.

A better candidate to explain the observed anomalies is the coupling of the phonons to charge stripes, similar to what has been proposed in recent theoretical work. As indicated in Fig. 5, the displacement pattern of the anomalous phonons is such as to favor dynamic charge accumulation on (approximately) every fourth row of atoms along \( \mathbf{b} \). The idea of charge stripe formation motivated us to search for elastic superlattice peaks related to static charge density wave formation, but we found none. This means that if our interpretation of the phonon anomaly is correct, the charge stripes in optimally doped YBCO are really dynamic in nature.

The above reasoning assumes that the charge modulation is along \( \mathbf{b} \), which means that the stripes must run parallel to \( \mathbf{a} \), an orientation that would be orthogonal to that implied by studies of magnetic fluctuations in underdoped YBCO. On the other hand, a charge-density-wave instability (CDW) along charge stripes has been discussed theoretically in such a way that one-dimensional conductors and hence could lead to the same type of transition which is typically observed in such materials. Of course, this argument tacitly assumes that the onset temperature for stripe correlations is quite high which is an unresolved issue.

Comparing the data for optimally doped YBCO (O7) with those of underdoped O6.6 we find that the low \( T \) dispersion of O6.6 definitely shows an anomaly similar to that observed in O7 although not quite as clearly. Re-inspection of temperature-dependent data also revealed a significant temperature effect which was overlooked in because it does not show up as a peak shift but again...
as a shift of spectral weight. This indicates that the phonon anomaly reported in this paper is not restricted to optimally doped samples.

In summary, we observed a pronounced temperature effect in the Cu-O in-plane bond-stretching vibrations strongly indicative of dynamic charge stripe formation in optimally doped YBCO. This result certainly adds credence to long-held theoretical expectations that the cuprates should form a charge-inhomogeneous state. On the other hand, several questions remain to be answered: Why is the effect, if present at all, much less pronounced along the a direction? Why is the effect not seen in optimally doped LSCO? Last but not least, how does one reconcile the magnetic response of optimally doped YBCO with the idea of a dynamic stripe phase? As will be discussed in a separate paper, antiferromagnetic spin fluctuations have indeed been observed on the same sample, but their dispersive nature and their apparent in-plane isotropy do not fit to the naive expectations of the classical stripe picture.

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