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Long-wavelength dispersion of transverse acoustic phonons in untwinned $YBa_2Cu_3O_{7-\delta}$ single crystals

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In order to study the possibility of enhancing the electron-phonon coupling λ in cuprates by soft transverse acoustic (TA) modes, using high-resolution ($\Delta E = 1.3$ meV) inelastic x-ray scattering, we measured the in-plane TA mode at low $q \lesssim 0.1$ along the a^* axis on optimally doped untwinned $YBa_2Cu_3O_{7-\delta}$ single crystals. We find that the dispersion is linear with no softening down to $q = 0.02$ and unchanged upon cooling from 300 to 98 K. The sound velocity is found to be 2700 ± 70 m s^{-1}, in agreement with ultrasound data. Thus, no bending instability exists in the CuO_2 layers and we conclude that larger λ and T_c may be possible in cuprates by inducing this instability.

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The role of the conventional electron-phonon pairing mechanism in the high-temperature superconductivity of cuprates remains highly controversial. The interest in the subject has been renewed after the discovery of phonon-mediated superconductivity with T_c as high as ≈ 40 K in MgB_2.[1] This high T_c put forward the hypothesis that phonons significantly contribute to the pairing in cuprates, although additional, e.g., electronic, mechanisms may coexist and dominate. Early evidence of strong electron-phonon coupling and lattice instabilities in cuprates,[2] has been confirmed recently by various experiments, such as inelastic neutron scattering (INS)[3,4] and inelastic x-ray scattering (IXS).[5,6] However, the magnitude of the electron-phonon coupling parameter λ remains controversial[7,8] and the question arises whether λ could be enhanced in cuprates, thus further raising T_c.

In order to address the above issue, in this work we examine the possibility that soft transverse acoustic (TA) modes in the low q region, which has not been studied previously in cuprates, may significantly enhance λ and T_c. Our motivation arises from the experimental evidence that largest T_c values are concomitant to a TA softening in a wide range of superconducting systems. Dramatic T_c enhancements up to 10 K have been reported in A15 (Ref. 9) and transition metal carbides.[10] Sizeable effects have been known in a number of elements such as alkaline and transition metals.[11–13] In relatively simple systems, such as lithium[11] or tellurium,[14] first-principles calculations within the Eliashberg theory enable to quantitatively account for the correlation between T_c and softening-induced λ variations. In carbides, theoretical calculations indicate that soft TA modes may arise from resonances in the electronic polarization[15] that lead to large λ values in systems with reduced dimensionality and covalent d-electron bonds.[16] These two features are indeed common to A15 and cuprates.

Consistently with the above experimental and theoretical findings, as discussed elsewhere,[13] soft phonons may effectively enhance λ, considering that $\lambda_{\mathbf{q},\gamma}$ for a given mode of index γ and wave vector $\mathbf{q}$ scales as the inverse of the wave frequency $\nu_{\mathbf{q},\gamma}$:[17]

$$\lambda_{\mathbf{q},\gamma} = \frac{1}{\nu^2_{\mathbf{q},\gamma}} \sum_{m,n} \int |\langle u_{\mathbf{k+q},n} | \mathbf{e}_{\mathbf{q},\gamma} \nabla_{\mathbf{R}} V_{\mathbf{q}} | u_{\mathbf{k},m} \rangle|^2 \times \frac{\delta(E_{\mathbf{k},m} - E_F)\,\delta(E_{\mathbf{k+q},n} - E_F)}{(2\pi)^2 N(E_F)} \frac{d^3k}{\Omega_{BZ}}, \qquad (1)$$

where $N(E_F)$ is the electron density of states at the Fermi level, the integral is over the Brillouin zone Ω_{BZ}, the bracketed quantities indicate matrix elements of the gradient of the self-consistent potential V with respect to the atomic displacement $\mathbf{R}$, $\mathbf{e}_{\mathbf{q},\gamma}$ is the phonon polarization and m and n are band indices.

Equation (1) shows that a softening-induced enhancement of λ is potentially large for acoustic modes at $q \lesssim 0.1$ in reduced lattice units (r.l.u.), provided the corresponding matrix element is sizeable. The reason is twofold: (1) ν vanishes as $q \to 0$; (2) the $q \lesssim 0.1$ range has the largest spectral weight in the gap equation.[18] In layered systems, TA modes are interesting because a softening at $q \lesssim 0.1$ is favored by the bending instability of the layers. For isotropic layers, according to standard theory of elasticity, the corresponding mode dispersion is[19]

$$\omega^2_{\mathbf{q}} = U^2_{\parallel} q^2_{\parallel} + U^2_{\perp} q^2_{\perp} + \alpha q^4_{\parallel}, \qquad (2)$$

where $\omega = 2\pi\nu$, α is the bending stiffness, the suffixes $\parallel$ and $\perp$ indicate the in- and out-of-plane quantities, $U = c_{44}/\varrho$ is the square of the velocity of sound in the absence of the last term, c_{44} is the layer shear elastic constant, and ϱ is the volume density. Although the $q \lesssim 0.1$ range of acoustic modes has been little studied for the reason described below, the bending dynamics of Eq. (2) has been found in a number of systems, including A15,[9] graphite,[20] graphite intercalated compounds,[21] and layered transition metal dichalcogenides.[22] For the $[\xi, \xi, 0]$ TA mode of Nb_3Sn, a 50% decrease of ω is found at $q \approx 0.1$, which would potentially lead to a 300% increase of λ for this mode.

The above scenario may lead to a large enhancement of λ in cuprates as well, considering the magnitude of the in-plane TA phonon linewidth $\Gamma_{\mathbf{q}}=0.25$ meV at $\mathbf{q}=(1/4,1/4,6)$, found in $La_{2-x}Sr_xCuO_4$.[23] Although no estimation of λ is available for this mode, the above linewidth value is compatible with a strong coupling regime by taking into account the vanishing *d*-wave gap along the *xy* direction.[23] Thus, we studied the dispersion in the $q \lesssim 0.1$ range of interest on $YBa_2Cu_3O_{7-\delta}$ (YBCO) by means of IXS. This *q* range of acoustic modes is unexplored for cuprates as well as for most materials due to the experimental difficulty of combining a high resolution in energy $E=h\nu$ and in momentum transfer *q*, with a crystal having a very low degree of mosaicity.

We carried out the experiment at the undulator ID28 beamline of the ESRF, where the above experimental conditions are met. We employed beam spot sizes as small as $\approx 0.2\times 0.1$ mm^2 and Si (11 11 11) and (13 13 13) monochromator Bragg reflections.[24] Using the ID28 6-m-long spectrometer, these reflections provided an overall energy resolution $\Delta E=1.7$ and 1.3 meV, respectively.[25,26] Thanks to this very high resolution and suitable untwinned YBCO single crystals, we succeeded to measure the inplane TA dispersion $\omega_{\mathbf{q}}$ along Δ ($\mathbf{q}\parallel a^*$ or $[\xi,0,0]$) with out-of-plane polarization ($\mathbf{e}\parallel c^*$ or $[0,0,\zeta]$) from $q=0.3$ down to $q=0.02$. This is a challenging experiment, for the *q* resolution $\Delta Q_a$of this phonon is limited by two factors: (1) the mosaic spread of the crystal $\Delta\vartheta_c$ convoluted with the divergence of the incident beam, the convolution being measured experimentally by the rocking curve width $\Delta\omega$; (2) the transverse component $\Delta 2\theta_t$ of the width $\Delta 2\theta$ of the scattering solid angle. In a typical INS experiment, ΔQ is dominated by the former factor. In the IXS case, ΔQ_a can be greatly enhanced by virtue of the high brilliance of synchrotron undulator sources and optimized beamline optics, provided a high quality crystal with a low degree of mosaicity is used. This is the case of the present experiment.

We measured two YBCO single crystals, labeled as 810R and 811R, grown by the flux method, as described elsewhere.[27] The crystals are small platelets of $\sim 1\times 1\times 0.1$ mm^3 size from two batches prepared under the same conditions. Oxygen postannealing at 500 °C for two weeks enabled to achieve a full oxidation ($\delta\approx 0.1$). The degree of mosaicity was very low in both crystals, as indicated by the full widths at half maximum of the (006) rocking curves, $\Delta\omega=0.009$ and 0.018° for 810R and 811R, respectively, that are close to the horizontal beam divergence 0.007°. This gives a mosaic spread of 0.005°–0.016°. Using the Si (13 13 13) reflection ($\lambda_i=0.4824$ Å), the contribution of $\Delta\omega$ to ΔQ is 0.0003–0.0006 r.l.u. in the (0,0,6) Brillouin zone. We used two configurations of horizontal slits with horizontal openings of 0.190°and 0.095°, the latter for the measurements at *q* points near the zone center. To minimize the spurious contribution from the (0,1,0) direction near the zone center, the opening of the vertical slits of the analyzer was reduced to decrease the sagittal scattering angle from 0.57 to 0.19°. We found that, under the above conditions, ΔQ along the longitudinal $[\xi,0,0]$ and transverse $[0,0,\zeta]$ directions in the (0,0,6) Brillouin zone is 0.001–0.002 and 0.04–0.08 r.l.u., respectively. In conclusion, in the present experiment, ΔQ is dominated by the divergence of the scattered beam, i.e., by the analyzer slit opening. Using the Si (11 11 11) reflection ($\lambda_i=0.5701$ Å), the estimated ΔQ values are decreased by an amount of the order of the 16% increase of λ_i of this reflection with respect to the (13 13 13) one.

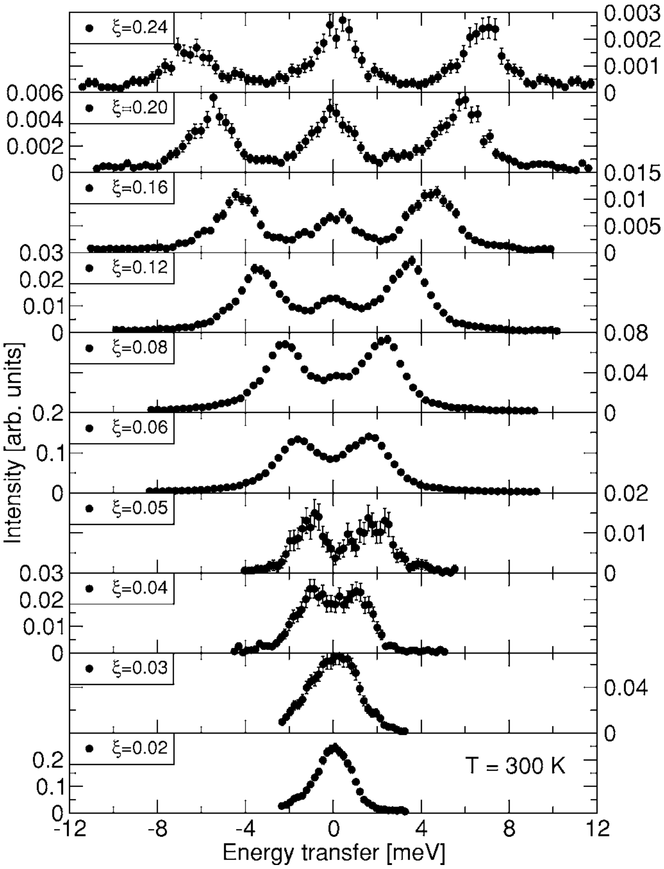

FIG. 1. Representative energy scans near (0,0,6) for various values of momentum transfer ξ along the a^* axis for the 810R YBCO single crystal at 300 K. Si (11 11 11) and (13 13 13) reflections were used for $\xi \geq 0.06$ and $\xi \leq 0.05$, respectively.

The phonon dispersion was measured in a fixed-*Q* configuration at various $(\xi,0,6)$ points of the Brillouin zone in the vicinity of the (006) Bragg reflection. The latter is the most intense (00*l*) reflection in fully oxygenated YBCO, thus providing the largest dynamical structure factor for low-*q* acoustic phonons. The same run of measurements was carried out on the two crystals. In Fig. 1, a series of room temperature energy scans at various values of phonon momentum ξ is reported for sample 810R. For all ξ values, one clearly distinguishes the triple peak structure consisting of the central peak, that corresponds to diffuse elastic scattering, and of the two lateral phonon Stokes and anti-Stokes peaks. The Si (11 11 11) data enabled to resolve the phonon peaks down to $\xi=0.04$. At lower ξ values, these peaks appear as small shoulders of the central peak. To resolve them, it was necessary to use the (13 13 13) reflection and closer analyzer slits. The relatively low intensity of the central peak

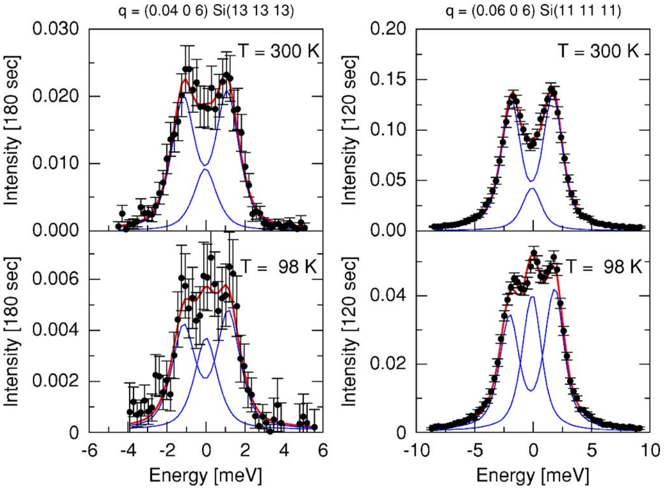

FIG. 2. (Color online) Comparison between the in-plane TA phonon peaks at 300 and 98 K measured on the 810R sample using the Si (11 11 11) and (13 13 13) reflections. Solid lines are a fit of the data (see text).

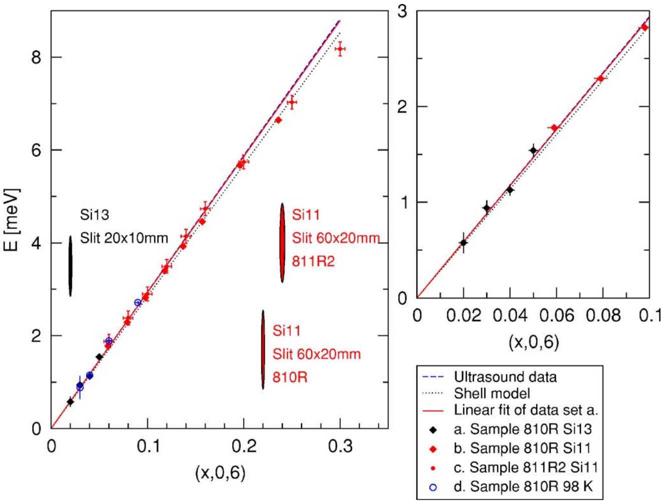

FIG. 3. (Color online) Dispersion of the in-plane TA phonon at 300 and 98 K for the two YBCO crystals 810R and 811R. The right panel shows in detail the low q data for 810R. Vertical lines indicate the energy and momentum resolutions that depend on the Si (11 11 11) or (13 13 13) reflection, slit openings, and crystal mosaicity (see text).

as compared to that of the phonon peaks indicates that the amount of disorder, responsible for diffuse scattering, is limited in this sample. For every scan, the above triple peak structure was fitted with the standard constraints regarding the relative position and intensity of the Stokes and anti-Stokes peaks set by the Bose-Einstein statistics. The phonons and diffuse elastic scattering peaks were modeled by harmonic oscillators and a delta function, respectively. The sum of these contributions was convoluted with the instrumental resolution function as described in Refs. 5 and 28. Four examples of fitted energy scans are shown in Fig. 2. The agreement within the error bars between the experimental data and the model is evident.

The phonon energies extracted from all of the above scans yield the dispersion of Fig. 3. The data of the two samples are reproducible within the experimental uncertainty. The accurate IXS measurements enable us to conclude that the dispersion is linear down to $q=0.02$. A linear regression in the $q \lesssim 0.1$ range for sample 810R, which gave the most accurate results thanks to the lower mosaicity, yields a transverse sound velocity in the a direction, $v_{s,a}=2700\pm70$ m s^{-1}. This value is in excellent agreement with the value of 2710 m s^{-1} derived from a previous ultrasound measurement of c_{55},[29] the relevant elastic constant for orthorhombic YBCO. Good agreement is also found with the value of 2620 m s^{-1} obtained from a shell model calculation.[30] The discrepancy in the latter case is compatible with the approximations of the model. To take into account the leveling off of the mode frequency at the zone boundary, we fitted the same data also using a sine function. This fit yields a slightly higher value, 2810 m s^{-1}, suggesting that the linear fit underestimates $v_{s,a}$ even at $q \lesssim 0.1$. In any case, it is apparent from Fig. 3 that the present data agree with the ultrasound ones within the error bars.

In order to unveil a softening-induced enhancement of λ, one has to consider that the softening may be absent at room temperature and appear only in the vicinity of T_c, as in the case of the A15 compound Nb_3Sn.[9] We then repeated the above measurements at 98 K, i.e., ≈ 6 K above T_c. From Figs. 2 and 3, it is noted that neither a hardening nor a softening is found. Thus, no change of the in-plane TA dynamics occurs in presence of the superconducting instability. No measurements at temperatures below T_c were performed, as this would concern the effects of phonon renormalization induced by the superconducting transition,[31] which goes beyond the scope of this work.

In conclusion, using IXS with unprecedented resolution in energy and momentum, we performed a measurement of in-plane TA phonon dispersion in the very low-q limit on cuprates. We succeeded to measure reproducibly the dispersion along the a^* axis down to $q=0.02$ on two untwinned YBCO single crystals. Upon cooling from 300 to 98 K, i.e., well inside the region of superconducting fluctuations, the dispersion remains linear within the experimental uncertainty and the sound velocity $v_{s,a}$ is unchanged. The measured $v_{s,a}$ value is in agreement with previous ultrasound measurements and shell model calculations. We conclude that no bending instability of the superconducting CuO_2 layers exists in YBCO. It would be interesting to induce this instability in cuprates, for a significant T_c enhancement would be expected, as in the case of A15 compounds. Further theoretical work would provide hints regarding those modifications of the electronic and crystal structures that may favor such instability. Finally, we demonstrated the suitability of IXS to study low-energy excitations in the $q \rightarrow 0$ limit, provided single crystal with a low degree of mosaicity are used.

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