q Dependence of Self-Energy Effects of the Plane Oxygen Vibration in $YBa_2Cu_3O_7$

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We have measured the temperature dependence of the peak position and linewidth of the 42.5 meV phonon branch in a twinned single crystal of $YBa_2Cu_3O_7$ as a function of wave vector $\mathbf{q}$. In the $(100)/(010)$ direction in the Brillouin zone, considerable softening and broadening occur below the superconducting transition temperature T_c at some values of $\mathbf{q}$. We observe an order of magnitude smaller softening and no linewidth broadening for $\mathbf{q}$ in the $(110)/(1\bar{1}0)$ direction. Possible implications of these findings for the symmetry of the superconducting order parameter are discussed.

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In superconductors, a redistribution of electronic states can result in changes of phonon peak positions and linewidths below the superconducting transition temperature T_c [1]. The magnitude of these effects is determined by the electron-phonon coupling strength and by the temperature dependence of $P(\mathbf{q}, \omega)$, the electronic polarizability at the phonon momentum $\mathbf{q}$ and energy ω. $\mathrm{Im}[P(\mathbf{q}, \omega)]$ contributes to the phonon linewidth, and is proportional to the joint density of occupied and unoccupied states separated by $\mathbf{q}$ and ω. $\mathrm{Re}[P(\mathbf{q}, \omega)]$ renormalizes the peak position. Since the opening of the superconducting gap $\Delta_{\mathbf{k}}$ at the Fermi surface has a direct effect on the density of states and on $P(\mathbf{q}, \omega)$, phonon self-energy effects can be used as a probe of $\Delta_{\mathbf{k}}$ [2–7].

The quasi-two-dimensional Fermi surface of the layered cuprate superconductors makes it possible to use phonon renormalization effects as a function of $\mathbf{q}$ to study the magnitude and phase of their superconducting order parameter over the entire Fermi surface [3,4]. This method is particularly promising for $YBa_2Cu_3O_{7-x}$ (YBCO), where Raman work has shown that the plane oxygen vibration of B_{1g} symmetry and energy 42.5 meV has a strong temperature dependence of both peak position and linewidth below T_c, presumably because its energy is close to 2Δ [8,9]. Unfortunately, Raman scattering is limited to the zone center, where $P(\mathbf{q}, \omega)$ is determined by the gap average over the Fermi surface [4,5]. Inelastic neutron scattering measurements away from the zone center are essential to probe $\Delta_{\mathbf{k}}$ at well defined values of $\mathbf{k}$. Pyka *et al.* measured the temperature dependence of the peak position of the 42.5 meV phonon in the $(100)/(010)$ direction in the Brillouin zone by inelastic neutron scattering in a twinned single crystal of $YBa_2Cu_3O_{7-x}$ at two different oxygen concentrations [10]. However, these results provided only limited information since the resolution was insufficient to measure the linewidth and because measurements along at least two directions in the Brillouin zone are required to distinguish s-wave and d-wave pairing states [4,5].

This Letter reports new results on the temperature dependence of the peak position and linewidth of the 42.5 meV phonon branch in a twinned single crystal of $YBa_2Cu_3O_7$ along the $(100)/(010)$ and $(110)/(1\bar{1}0)$ directions in the Brillouin zone. The linewidth results are particularly important since many calculations of phonon renormalization focused on the linewidth.

The experiments were performed on the H7 and H8 triple axis spectrometers at the High Flux Beam Reactor at Brookhaven National Laboratory. The final energy E_f was fixed in all our measurements, while the energy transfer was varied by scanning the initial energy E_i. The main experimental constraint was that second order Bragg scattering by the analyzer selects a neutron energy of $4E_f$, thus the scattering process $E_i \rightarrow 4E_f$ contributes to the scattered intensity. Since $E_i < 4E_f$ in our measurements, $E_i \rightarrow 4E_f$ is an energy gain process which is negligible when $k_BT \ll 4E_f - E_i$. However, at $k_BT \sim (4E_f - E_i)/2$, the spectra acquired a temperature dependent background due to energy gain multiphonon scattering. To minimize this contamination and optimize our resolution, w e used a Be002 monochromator and a PG002 analyzer with collimations of $10'/20'/40'/40'$ at a fixed final energy $E_f = 25$ meV to take data between 100 and 50 K. The calculated energy resolution was 2 meV full width at half maximum (FWHM). Data at 50 K and below were taken mostly with a PG002 monochromator and analyzer with collimations $10'/10'/40'/60'$ at a fixed final energy of 19 meV, and with a FWHM resolution of 1.9 meV. Our sample was a large fully oxygenated (75 g) single crystal of $YBa_2Cu_3O_7$ ($T_c \sim 93$ K). The mosaic spread of the

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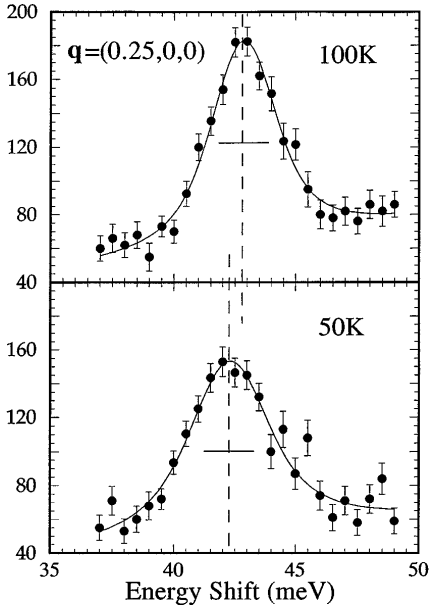

FIG. 1. 42.5 meV phonon at 100 and 50 K and $\mathbf{q} = (0.25, 0, 0)$. The solid lines are the results of fits as discussed in the text.

largest domain (~70% of the crystal) was 2° with some smaller domains as far as 5° from the main domain.

Previous Raman measurements have shown that at $\mathbf{q} = 0$ the 42.5 meV phonon involves an out-of-phase c-axis vibration of the plane oxygens [11]. We performed all our measurements in the Brillouin zone adjacent to wave vector $\mathbf{Q} = (1, 0, 14)$, where the structure factor for this phonon eigenvector is maximized. The integrated intensity of the phonon peak was $\mathbf{q}$ independent within ~10% in this Brillouin zone. This implies that the phonon eigenvectors away from $\mathbf{q} = 0$ are still dominated by out-of-phase c-axis vibrations of plane oxygens. Recent neutron measurements and model calculations are also consistent with this result [12]. (In our notation $\mathbf{q}$ represents momentum transfer reduced to the first Brillouin zone. The first two indices in our representation of $\mathbf{Q}$ and $\mathbf{q}$ correspond to the in-plane components in units of $2\pi/a \sim 1.63$ Å^{-1}. The third index corresponds to the component perpendicular to the layers in units of $2\pi/c \sim 0.54$ Å^{-1}.)

The measured spectra were fitted with a Lorentzian line shape convoluted with a Gaussian resolution function and a linear background (Fig. 1). The flat component of the background in our fits was adjusted in such a way as to keep the resulting integrated intensity temperature independent. The experimental uncertainty was ~0.1–~0.15 meV for the peak position and ~(10–20)% for the width (FWHM), depending on the measurement. The data were taken in three runs with the sample remounted and reoriented, and the monochromator realigned each time. Small differences in alignment resulted in different systematic errors of the peak position during each set of measurements. We corrected for these errors by offsetting each set of peak position values by constant amounts adjusted to achieve the best overlap at $\mathbf{q}$ points, where the same measurements were performed during different runs.

At $\mathbf{q} = 0$ [Figs. 2(a) and 2(b)], the peak position softens by ~0.85 meV from 100 to 50 K with no change below 50 K. Raman measurements of the peak position performed on a sample of oxygen content and T_c similar to ours are in excellent agreement with our data. We do not observe the linewidth changes seen in the Raman data presumably due to large statistical errors. It is also possible that our sample has a slightly different oxygen or impurity content from the one measured by Raman scattering, which has a strong effect on linewidth renormalization at $\mathbf{q} = 0$ [8,9]. However, the overall $\mathbf{q} = 0$ linewidth agrees well with the Raman results.

At $\mathbf{q} = (0.25, 0, 0)$ [Fig. 2(c)], the peak position softens by ~0.9 meV from 100 to 30 K with no change below 30 K. At $\mathbf{q} = (0.5, 0.5, 0)$ [Fig. 2(c)], there is an order of magnitude smaller softening. The linewidth

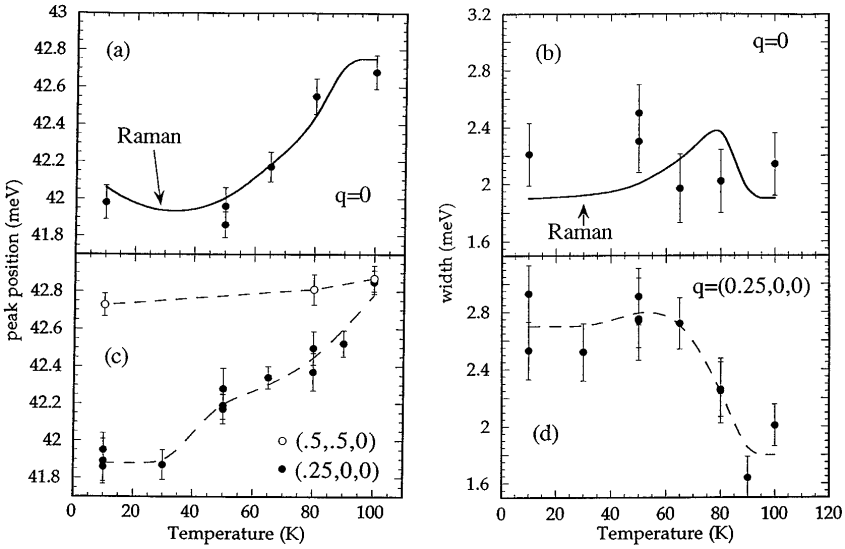

FIG. 2. Temperature dependence of peak position and linewidth at different values of $\mathbf{q}$. Dashed lines are guides to the eye and solid lines represent Raman data from Ref. [8].

of the $\mathbf{q} = (0.25, 0, 0)$ phonon clearly increases from ~ 2 meV at 100 K to ~ 2.8 meV at 50 K [Fig. 2(d)]. At $\mathbf{q} = (0.25, 0.25, 0)$ and $\mathbf{q} = (0.5, 0.5, 0)$, the linewidth showed evidence of narrowing, though this narrowing was within the experimental uncertainty. Our data do rule out any significant broadening below T_c at these $\mathbf{q}$ values.

Figure 3 shows that at 100 K both the linewidth and the peak position are $\mathbf{q}$ independent within the experimental uncertainty. At 50 K, the peak position increases almost linearly in the (100)/(010) direction from 41.8 meV at $\mathbf{q} = 0$, reaching 42.8 meV at $\mathbf{q} = (0.5, 0, 0)$. This result agrees with the data of Pyka *et al.* [10]. The linewidth is maximum at $\mathbf{q} = (0.25, 0, 0)$. At 10 K, the peak position is the same at $\mathbf{q} = 0$ and at $\mathbf{q} = (0.25, 0, 0)$. It then increases sharply from 41.8 meV at $\mathbf{q} = (0.25, 0, 0)$ to 42.85 meV at $\mathbf{q} = (0.5, 0, 0)$.

Raman results combined with our neutron data show that the peak position and width of the 42.5 meV phonon are $\mathbf{q}$ independent, which implies that Fermi liquid effects (such as Landau damping, Kohn anomalies, etc.) play a negligible role in determining the phonon dispersion.

A strong $\mathbf{q}$ dependence of the phonon renormalization below T_c can result from $\mathbf{q}$ dependences of the electron-phonon coupling and/or of the $P(\mathbf{q}, \omega)$. Two different microscopic mechanisms of coupling of the 42.5 meV phonon to electronic states have been suggested [3,7], but neither of them predicts order of magnitude variations of the coupling constant with $\mathbf{q}$ [7,13]. In interpreting our experimental data below T_c, we consider a simple BCS model with an anisotropic s-wave or d-wave gap and no electron-electron interactions otherwise. Within this approximation the coherent contribution to $\mathrm{Im}[P(\mathbf{q}, \omega)]$ is given by [5]

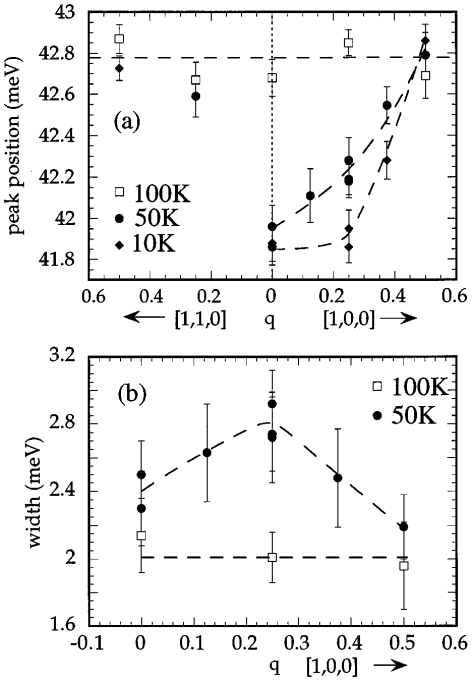

FIG. 3. $\mathbf{q}$ dependence of (a) peak position and (b) linewidth at different temperatures. Dashed lines are guides to the eye.

$$\mathrm{Im}[P(\mathbf{q},\omega)] = \frac{\pi}{N}\sum_{\mathbf{k}}\left\{\left[1 + \frac{\varepsilon_{\mathbf{q}-\mathbf{k}}\varepsilon_{\mathbf{k}} - \mathrm{Re}(\Delta_{\mathbf{k}}\Delta^{*}_{\mathbf{q}-\mathbf{k}})}{E_{\mathbf{q}-\mathbf{k}}E_{\mathbf{k}}}\right][f(E_{\mathbf{q}-\mathbf{k}}) - f(E_{\mathbf{k}})]\delta(\omega - E_{\mathbf{q}-\mathbf{k}} + E_{\mathbf{k}})\right\}$$
$$+ \frac{1}{2}\frac{\pi}{N}\sum_{\mathbf{k}}\left\{\left[1 - \frac{\varepsilon_{\mathbf{q}-\mathbf{k}}\varepsilon_{\mathbf{k}} - \mathrm{Re}(\Delta_{\mathbf{k}}\Delta^{*}_{\mathbf{q}-\mathbf{k}})}{E_{\mathbf{q}-\mathbf{k}}E_{\mathbf{k}}}\right][f(E_{\mathbf{q}-\mathbf{k}}) - f(E_{\mathbf{k}}) - 1]\delta(\omega - E_{\mathbf{q}-\mathbf{k}} + E_{\mathbf{k}})\right\}, \quad (1)$$

where $\varepsilon_{\mathbf{k}}$ is the normal state dispersion relation for electrons, f is the Fermi function, $\Delta_{\mathbf{k}}$ is the momentum dependent gap, and $E^2_{\mathbf{k}} = \varepsilon^2_{\mathbf{k}} + \Delta^2_{\mathbf{k}}$. The first term arises from scattering by thermally excited quasiparticles, and the second arises from quasiparticle pair production, which dominates $\mathrm{Im}[P(\mathbf{q}, \omega)]$ when the phonon energy is greater than $|\Delta_{\mathbf{k}}| + |\Delta_{\mathbf{q}-\mathbf{k}}|$. The $\mathrm{Re}(\Delta_{\mathbf{k}}\Delta^{*}_{\mathbf{q}-\mathbf{k}})$ term is sensitive to the relative phase of $\Delta_{\mathbf{k}}$ and $\Delta_{\mathbf{q}-\mathbf{k}}$. If ω is slightly above $|\Delta_{\mathbf{k}}| + |\Delta_{\mathbf{q}-\mathbf{k}}|$, broadening of the phonon is expected below T_c when $\Delta_{\mathbf{k}}$ and $\Delta_{\mathbf{q}-\mathbf{k}}$ have the same phase. When $\Delta_{\mathbf{k}}$ and $\Delta_{\mathbf{q}-\mathbf{k}}$ have the opposite phase, narrowing is expected. The peak position renormalization is given by a Kramers-Kronig transformation of Eq. (1), thus it is also expected to be sensitive to the phase of the superconducting order parameter. The Fermi function and delta function terms limit the contribution to $\mathrm{Im}[P(\mathbf{q}, \omega)]$ to transitions between different regions of the Fermi surface that satisfy the Pauli exclusion principle as well as energy and momentum conservation. Thus the joint density of states contributing to the phonon self-energy depends on the shape of the Fermi surface, on the electronic dispersion curves, and on the magnitude of $\Delta_{\mathbf{k}}$.

It is now well known that $\Delta_{\mathbf{k}}$ is highly anisotropic in the cuprates, with maxima in the (100)/(010) directions and minima or nodes in the $(110)/(1\bar{1}0)$ directions. The sharp reduction in self-energy effects in the (100) direction is presumably simply a consequence of this gap anisotropy. (Zeyher [14] considered an isotropic s-wave superconductor with a cylindrical Fermi surface and found that phonon self-energy effects generally decrease with increasing $\mathbf{q}$. However, this reduction is modest as long as $q < 2K_f$, a condition that holds even at the zone boundary in YBCO, and is thus unlikely to account for our data.)

Our observation of very different behavior in the (110) direction further constrains models of $\mathrm{Im}[P(\mathbf{q}, \omega)]$. Figure 4 shows a diagram of the Fermi surface in $YBa_2Cu_3O_{7-x}$ [15] with some of the electronic transitions which contribute to the phonon self-energy at $\mathbf{q} \sim (0.25, 0, 0)$ and $\mathbf{q} \sim (0.5, 0.5, 0)$. Below T_c, all such transitions occur between regions of maximum gap

magnitude for both d-wave and anisotropic s-wave order parameters. Transitions contributing to the self-energy of the $\mathbf{q} \sim (0.25, 0, 0)$ phonon occur between regions of the Fermi surface where $\Delta_{\mathbf{k}} = \Delta_{\mathbf{q}-\mathbf{k}}$ for both order parameters; whereas for $\mathbf{q} \sim (0.5, 0.5, 0)$ and its umklapp counterparts, $\Delta_{\mathbf{k}} = \Delta_{\mathbf{q}-\mathbf{k}}$ for an anisotropic s-wave and $\Delta_{\mathbf{k}} = -\Delta_{\mathbf{q}-\mathbf{k}}$ for a d-wave order parameter. In the s-wave case, self-energy effects at the two $\mathbf{q}$ values should be of the same order of magnitude unless the joint density of states at $\mathbf{q} \sim (0.25, 0, 0)$ is much greater than at $\mathbf{q} \sim (0.5, 0.5, 0)$. Such an anisotropy in the joint density of states can arise from nesting effects [6]. In the d-wave scenario, the dramatic difference in the observed linewidth and peak position renormalizations at these $\mathbf{q}$ values could be a direct consequence of the superconducting order parameter having a different sign along the x and y directions, and is independent of the shape of the Fermi surface.

Detailed calculations for realistic YBCO Fermi surface and electronic dispersions are necessary to develop a quantitative understanding of our data and to assess the relative importance of coherence factors and density of states effects. Refined calculations should account for electronic correlations, impurity scattering, and electron-phonon coupling. No existing theoretical models predict the maximum in the linewidth broadening observed at $\mathbf{q} \sim (0.25, 0, 0)$ [Fig. 3(b)], since even in the case of nested Fermi surface the largest broadening is predicted at $\mathbf{q} = 0$ [6]. This result might be related to the maximum in the peak position shift at $\mathbf{q} \sim (0.25, 0, 0)$ in slightly oxygen-deficient $YBa_2Cu_3O_{6.92}$ reported in Ref. [10]. Finally, different dispersions of the 42.5 meV phonon branch at 50 and 10 K may imply that $\Delta_{\mathbf{k}}$ has a different temperature dependence at different $\mathbf{k}$, which needs to be understood as well.

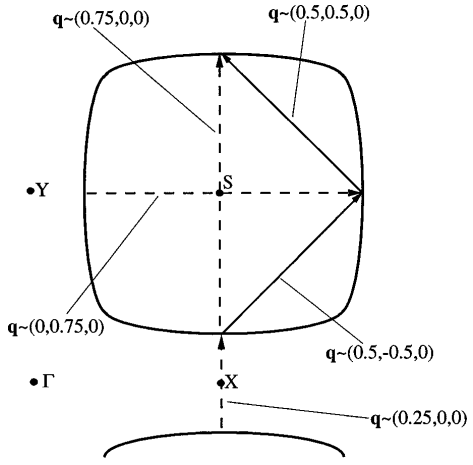

FIG. 4. Schematic of the Fermi surface of $YBa_2Cu_3O_7$ and the electronic transitions between regions of maximum gap assuming d-wave or anisotropic s-wave order parameters. Solid lines represent transitions contributing to the linewidth of the $\mathbf{q} = (0.5, 0.5, 0)$ phonon and dashed lines represent transitions contributing to the linewidth of the $\mathbf{q} = (0.25, 0, 0)$ phonon.

In conclusion, we developed a scattering geometry for performing linewidth-sensitive measurements of the out-of-phase c-axis vibration of plane oxygens in $YBa_2Cu_3O_7$, which is sensitive to the superconducting transition. We found strong anisotropy in the superconductivity induced phonon self-energy effects between the $(100)/(010)$ and $(110)/(1\bar{1}0)$ directions, which is indicative of a d-wave superconducting order parameter and/or a strong anisotropy of the electronic structure close to the Fermi surface.

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