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Anisotropic electronic Raman scattering in untwinned $YBa_2Cu_3O_{7-\delta}$

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Raman scattering results from an untwinned domain of a pure-phase crystal of the $YBa_2Cu_3O_{7-\delta}$ superconductor are presented. The room-temperature spectrum in the polarization geometry parallel to the Cu(1)-O(1) chains shows significantly more spectral weight due to electronic scattering than does the spectrum in the perpendicular geometry. The anisotropy is further evidenced by the difference in antiresonance strength between the low-energy 116-cm^{-1} modes of these two geometries. Low-temperature spectra show similar anisotropy, suggesting that the Cu(1)-O(1) chains may contribute to this electronic scattering.

Recent light scattering results on single crystals of $YBa_2Cu_3O_{7-\delta}$ have shown that there is strong electronic scattering in these high-temperature superconductors.[1] This electronic scattering manifests itself in the Raman spectra through a substantial background continuum which exhibits strong interference effects with the 116- and 340-cm^{-1} vibrational modes of the crystal. Whereas previous reports of light scattering results on untwinned single crystals of $YBa_2Cu_3O_{7-\delta}$ have described the anisotropic nature of various phonon spectral intensities,[2] as yet unreported are light scattering results indicating any anisotropic nature of the electronic continuum. In this paper, Raman spectra are presented that provide evidence that the Cu(1)-O(1) chains contribute to the electronic scattering. This participation of the chains in the electronic excitations in $YBa_2Cu_3O_{7-\delta}$ supports our earlier suggestion that the electronic scattering is in part due to the chains.[1]

The $YBa_2Cu_3O_{7-\delta}$ crystals were grown using a copper-oxide-rich flux technique[3] with a modified oxygenation treatment. The as-grown crystals were heated in flowing oxygen at 600 °C for 1 h, cooled to 400 °C in roughly 1 h, held at 400 °C for four days, and then rapidly cooled to room temperature. The crystals were found to have sharp transitions (as shown in Ref. 4) with onsets at 90 K as determined by zero-field-cooled magnetic-susceptibility measurements.

While most of the crystals prepared in this way have extensive twinning along the (110) direction, a few exhibit sizable single-domain regions. The crystal used in this study (Fig. 1) was found to have two untwinned regions, the larger of which was approximately 0.5 mm^2, as identified by observation of the [001] face under a polarizing microscope. The **a** and **b** crystalline axes were identified optically with a polarizing microscope according to methods described elsewhere.[5]

The Raman measurements were taken in the pseudobackscattering geometry with the polarized 5145-Å line of an argon ion laser aligned perpendicular to the c axis of the crystal. In this orientation the incident light could be focused with its polarization vector either along the chain axis, producing (**y**,**y**) and (**y**,**x**) spectra, or perpendicular to the chain direction, producing (**x**,**x**) and (**x**,**y**) spectra. This allowed for coupling to excitations of orthorhombic A_g symmetry in the polarized geometries and to excitations of orthorhombic B_{1g} symmetry in the depolarized geometries.

The scattered light was detected with a Spex 1401 double monochrometer coupled to a home-built single dispersive stage. In conjunction with a nitrogen-cooled photomultiplier tube, these instruments provided a resolution of 2 cm^{-1} and a dark count of approximately 1 count/sec. The sample was placed in a liquid-helium cryostat which allowed for accurate temperature adjustment down to 3 K. To avoid laser heating of the sample during the low-

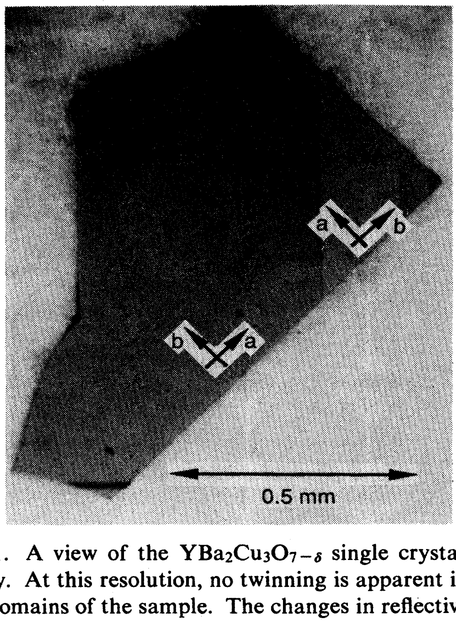

FIG. 1. A view of the $YBa_2Cu_3O_{7-\delta}$ single crystal used in this study. At this resolution, no twinning is apparent in the two labeled domains of the sample. The changes in reflectivity in the different regions serve to identify the untwinned domains and their orientations.

temperature runs, the power densities were maintained at 15 W/cm^2.

An untwinned single crystal of $YBa_2Cu_3O_{7-\delta}$ should display anisotropic phonon spectral intensities characteristic of its orthorhombic structure. The dissimilar bond polarizabilities along different crystalline directions of the 340- and 504-cm^{-1} phonons require that these modes should exhibit stronger intensities in the **(x,x)** and **(y,y)** geometries, respectively.[2] A stronger intensity is also expected in the **(y,y)** geometry for the 116-cm^{-1} mode because of its larger Raman polarizability along the chain direction. These features are all evident in the room-temperature data (Fig. 2). The 116- and 504-cm^{-1} modes exhibit stronger intensities in the **(y,y)** geometry by factors of approximately 2.3 and 1.6, respectively. The intensity of the 340-cm^{-1} mode in the **(x,x)** geometry is stronger by a factor of 1.5. The 150-cm^{-1} mode exhibits roughly the same intensity in both the polarized spectra. These features are consistent with previous results on untwinned samples,[2] and they serve to demonstrate the orthorhombic nature of this compound.

The background continuum constitutes real inelastic scattering as evidenced by the absence of scattering in the depolarized [**(y,x)** and **(x,y)**] geometries. This continuum may be due to interband electronic scattering for the reasons outlined in our earlier work.[1] Comparison of the room-temperature data on the continuum in the two geometries indicates that the spectral response of the electronic scattering is weakened when the exciting radiation is polarized perpendicular to the chains. The weakened electronic scattering also manifests itself in the "antiresonance" of the 116-cm^{-1} phonon. This mode's asymmetric line shape is described by an Auger-like process in which the phonon state couples strongly to the broad electronic continuum.[6] As a result, the strength of the continuum is reflected by the strength of the interference on the high-energy side of the 116-cm^{-1} mode. Therefore, the decreased asymmetry of this mode in the **(x,x)** spectrum indicates weaker interference demonstrating that this geometry provides a weaker electronic continuum.

The low-temperature spectra (Fig. 3) show a redistribution of the electronic continuum into a broad asymmetric peak, indicative of the formation of a superconducting-gap in these compounds.[1,7,8] As electronic states about the Fermi energy become disallowed due to the opening of the gap, the low-energy electronic continuum drops to a near-zero count rate in the Raman spectrum. Aside from a difference in intensity, the superconducting gap excitations show identical features in the two polarized spectra.

Examinations of the phonons in the superconducting state reveal that the 116-, 150-, 340-, and 504-cm^{-1} modes all continue to show roughly the same intensity ratios as in room-temperature data. The additional features at 440 and 580 cm^{-1} exhibit selection rules consistent with those derived by Thomsen *et al.*[2] The features detailing the anisotropy of the electronic scattering at room temperature are still apparent in the superconducting state. When the excited photon polarization is parallel to the chain direction, the scattering intensity due to the electronic excitations is stronger. Although the 116-cm^{-1} mode shows a weaker antiresonance character in the superconducting state in both of the polarized geometries, due to the shifting of the relevant electronic excitations to higher energies, it still exhibits a stronger antiresonance in

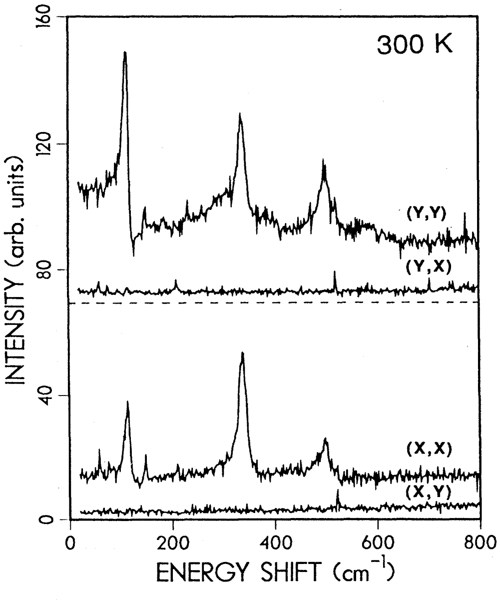

FIG. 2. The polarized [**(y,y)** and **(x,x)**] and depolarized [**(y,x)** and **(x,y)**] room-temperature spectra on an untwinned single crystal of $YBa_2Cu_3O_{7-\delta}$. Both polarization vectors lie on the *a*-*b* plane and the **y** direction is taken to be parallel to the chain axis of the crystal. The dashed line marks the zero-intensity level for the upper spectra.

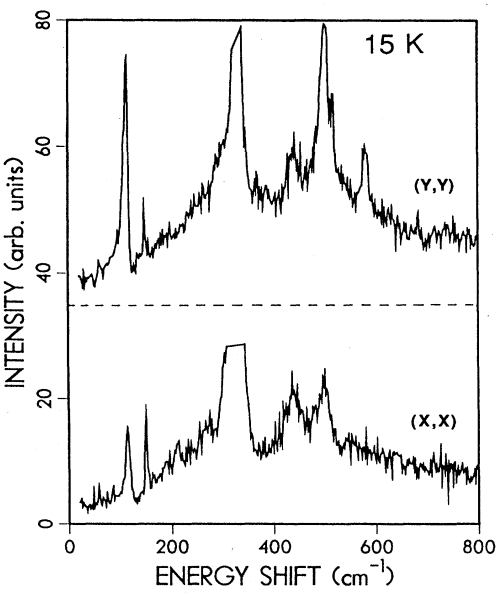

FIG. 3. Low-temperature polarized spectra of the untwinned sample. The 330-cm^{-1} mode has been truncated to make the features of the anisotropic electronic scattering more apparent. The dashed line marks the zero-intensity level for the upper spectra.

the geometry parallel to the chains.

In summary, from comparisons of the electronic continuum and the behavior of the 116-cm^{-1} phonon in the two polarized geometries, it is apparent that stronger electronic scattering occurs when the incident light is polarized along the direction of the chains. This provides evidence for the participation of the Cu(1)-O(1) chains in the electronic scattering process in both the normal and superconducting states.

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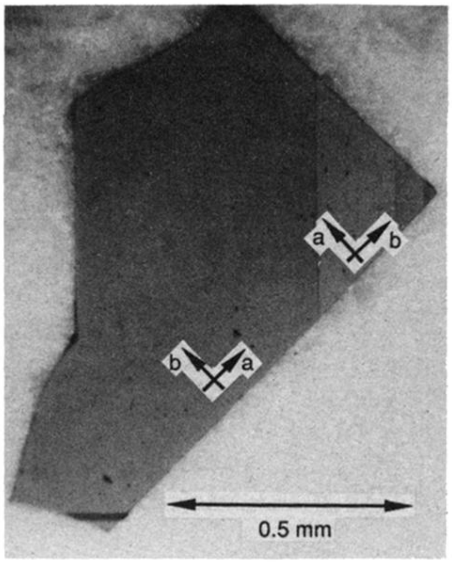

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