Temperature dependence of electron Raman scattering in superconducting and insulating $YBa_2Cu_3O_{6+x}$ single crystals

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(Submitted 3 November 1992)
Pis'ma Zh. Eksp. Teor. Fiz. **56**, No. 11, 587–591 (10 December 1992)

A detailed study has been made of electron Raman scattering in superconducting and insulating single crystals of $YBa_2Cu_3\,O_{6+x}$. The spectral changes which occur at frequencies $\omega < 600\ cm^{-1}$, on the one hand, and the different positions of the 2Δ peak in different polarizations, on the other, indicate that the superconducting gap is strongly anisotropic. In the normal phase the behavior of the imaginary part of the response function $R''(\omega)$ in the polarization $(x'x')$ corresponds to the model of a marginal Fermi liquid. In the polarization $(x'y')$, this behavior is independent of the temperature. In insulating crystals, $R''(\omega)$ is independent of the temperature up to $T \gtrsim 200$ K in both polarizations.

A significant thrust in recent research on the Raman spectra of the high-T_c superconductors has been to study the broad $(0 < \omega < 8000\ cm^{-1})$, structureless continuum of electron origin.[1] Research on the electron scattering of light in high-T_c superconducting crystals and in related high-T_c compounds can provide important information on the spectrum of electron excitations in the superconducting phase, in the normal (metallic) phase, and also in the insulating state. The topic of greatest interest is the temperature dependence of the electron scattering spectra at low frequencies. Here the spectral changes observed at $T < T_c$ make it possible to determine the size of the superconducting gap 2Δ and its symmetry properties,[2] while the temperature dependence at $T > T_c$ can provide data on the properties of the free carriers in the normal state. However, despite numerous studies of the continuum in the Raman spectra, there are still uncertainties in the results. For example, it was reported in Ref. 3 that the continuum in insulating $YBa_2Cu_3O_6$ crystals is of significant intensity—comparable to that of the continuum in superconducting samples. There is also uncertainty regarding the polarization dependence of the electron scattering spectra at low frequencies for superconducting $YBa_2Cu_3O_7$ crystals in the normal state at $T > T_c$ (Ref. 4). These difficulties may stem from (first) Raman scattering by impurities, surface effects, or other extrinsic effects. The quality of the samples, especially their surface quality, must therefore meet exacting standards. A second, and significant, source of difficulties may be the low level of the Raman scattering signal in the continuum. In this letter we are reporting a detailed study of Raman scattering in the insulating phase and also in the normal and superconducting phases of the compound $YBa_2Cu_3\,O_{6+x}$ over the frequency range $0 < \omega < 800\ cm^{-1}$. The high signal-to-noise ratio achieved in these experiments has made it possible to determine the temperature dependence of the electron scattering in various polarizations over the temperature range $T = 5$–300 K.

The experiments were carried out on a single-domain $YBa_2Cu_3O_7$ crystal with a sharp superconducting transition at $T_c = 92$ K and also on an insulating crystal of $YBa_2Cu_3O_6$. Each sample was a thin platelet ($d \simeq 30\ \mu m$) with a mirror-finish large **ab** plane with typical dimensions of 1×1 mm. The crystal was held in helium vapor in an optical helium constant-temperature chamber with an adjustable temperature. The Raman spectra were recorded in a geometry of backscattering of the light from a fixed region (a region of highest quality) on the basal plane of the crystal. The situation was monitored with a microscope. The light source was an Ar^+ laser with a wavelength $\lambda = 4880$ Å. The dimensions of the laser excitation spot were $\simeq 70 \times 280\ \mu m$; the power of the incident light was $P_0 \lesssim 10$ mW. The spectra were recorded with a Dilor *XY* triple monochromator with a multichannel optical detector.

According to the fluctuation dissipation theorem, the imaginary part of the response function of the electron system, $R''(\omega,T)$, can be found from the Raman spectra $S(\omega,T)$ by means of the relation

$$R''(\omega,T) = \left[1 - \exp\left(-\frac{\omega}{T}\right)\right] S(\omega,T). \tag{1}$$

To determine the temperature in the region in which the Raman scattering was excited, we used a measurement procedure like that of Ref. 5, carrying out measurements in the Stokes and anti-Stokes part of the Raman spectra. This approach made it possible to determine the temperature within $\Delta T \simeq \pm 5$ K. The heating which occurred in the excitation region at low temperatures was on the order of 30 K.

Figures 1 and 2 show Raman spectra of $YBa_2Cu_3O_6$ and $YBa_2Cu_3O_7$ crystals in the $z(x'y')\bar{z}$ geometry, corrected for the Bose factor [i.e., $R''(\omega)$], over the temperature range $T = 30$–300 K. The x' and y' axes are 45° away from the crystallographic **a** and **b** axes, respectively. It is easy to see that in the case of the insulating phase the continuum in $R''(\omega)$ is essentially independent of the temperature over the range $T = 30$–210 K. At $T = 300$ K, the intensity of the continuum increases over the entire spectral interval, without any significant redistribution in the spectrum itself. The temperature dependence of the spectra in the case of the superconducting crystal differs in important ways. In the first place, the scattering intensity in the continuum is nearly an order of magnitude higher than in the case of the insulating crystal. Second, at temperatures $T < T_c$ the intensity in the low-frequency part of the spectra ($\omega < 300\ cm^{-1}$) decreases, and a broad band peaking at $\omega \simeq 350$–$450\ cm^{-1}$ arises. The redistribution in the Raman spectra of the superconducting crystal can be followed more easily in terms of the ratios $r(\omega,T) = R''(\omega,T)/R''(\omega,110\ K)$ in the $(x'x')$ and $(x'y')$ polarizations, shown in Fig. 3. We see that in each polarization a spectral redistribution typical of the appearance of a superconducting gap[2] occurs at $T < T_c$: We have $r < 1$ at $\omega < 200\ cm^{-1}$, $r > 1$ at $300 < \omega < 700\ cm^{-1}$ (the so-called 2Δ peak), and[2)] $r \simeq 1$ at $\omega > 700\ cm^{-1}$. The crests of the 2Δ peak, ω_{max}, are in different spectral positions in the $(x'x')$ and $(x'y')$ polarizations. According to our estimates, in which we incorporate the contribution to $r(\omega)$ from the vibration at $335\ cm^{-1}$ because of its

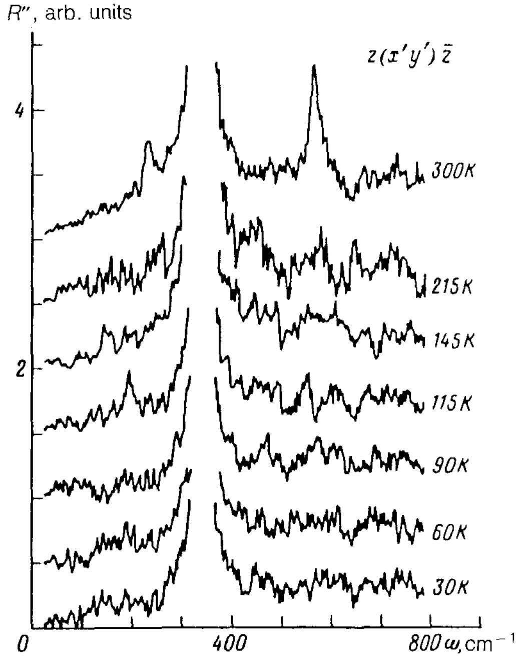

FIG. 1. Imaginary part of the response function, $R''(\omega)$, for a $YBa_2Cu_3O_6$ crystal in the $z(x'y')\bar{z}$ geometry at various temperatures. The spectra are shifted a distance of 0.5 successively along the vertical scale.

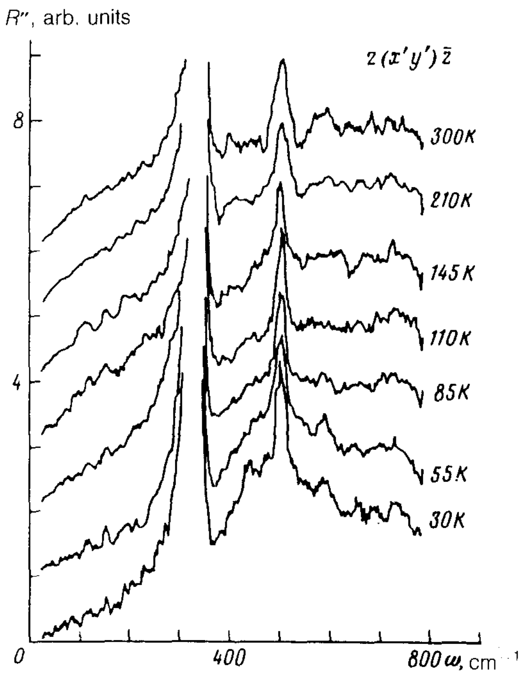

FIG. 2. Imaginary part of the response function, $R''(\omega)$, for a superconducting $YBa_2Cu_3O_7$ crystal in the $z(x'y')\bar{z}$ geometry at various temperatures. The spectra are shifted a distance of 1 successively along the vertical scale.

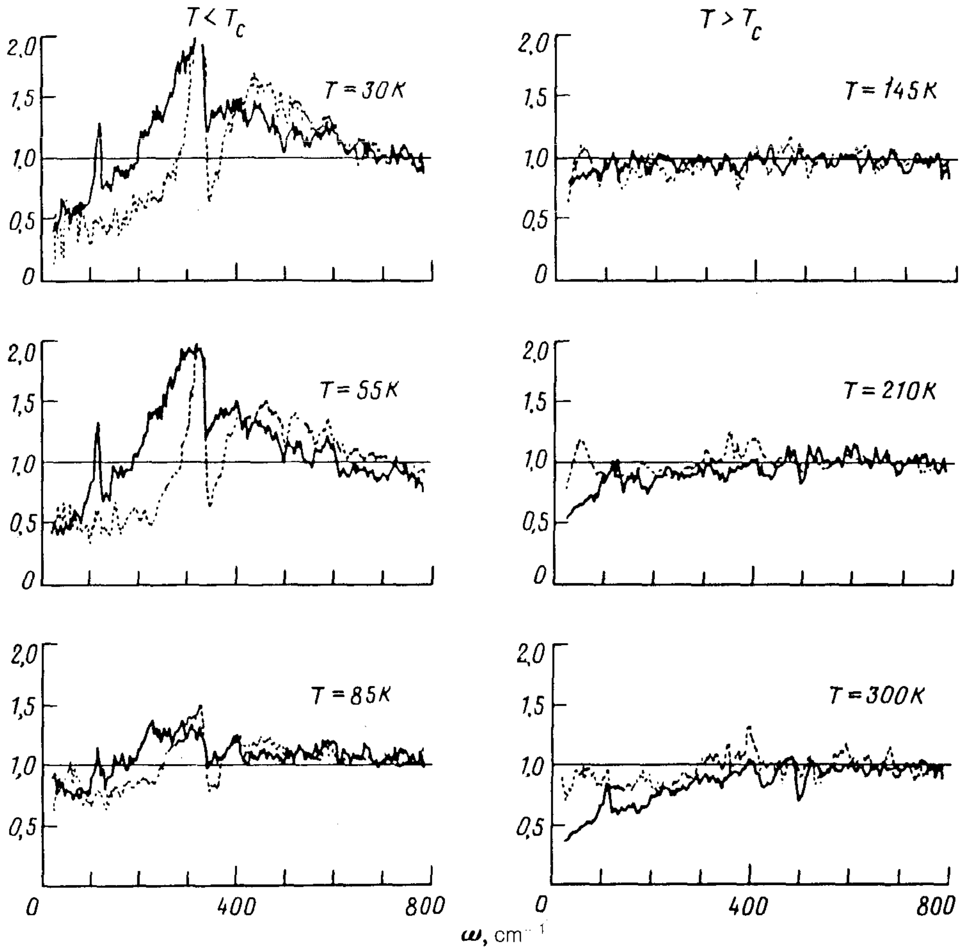

FIG. 3. The ratio $R''(\omega,T)/R''(\omega,110\text{ K})$ at various temperatures for a superconducting $YBa_2Cu_3O_7$ crystal. Solid lines—In the $(x'x')$ polarization; dashed lines—in the $(x'y')$ polarization.

softening in the course of the superconducting transition, we have $\omega_{max} \simeq 350\text{ cm}^{-1}$ in the $(x'x')$ geometry and $\omega_{max} \simeq 450\text{ cm}^{-1}$ in the $(x'y')$ geometry. These results indicate that the superconducting gap is anisotropic in **k** space. There are also several differences between the polarizations $(x'y')$ and $(x'x')$ in terms of the behavior $r(\omega)$ at $T > T_c$. In crossed polarizations, $(x'y')$, we have $r(\omega) \simeq 1$ over the entire spectral range, while in parallel polarizations, $(x'x')$, there is a temperature dependence in the ratio $r(\omega,T) \propto 1/T$ at frequencies $\omega < T$, while at $\omega > T$ we find $r(\omega) \simeq 1$. Note that the $r(\omega)$ behavior in the $(x'x')$ geometry has the temperature dependence characteristic of the model of a marginal Fermi liquid.[6] According to that model, $R''(\omega,T)$ can be described by

$$R''(\omega,T)=\begin{cases}\omega/T, & \omega<T,\\ \text{const}\cdot\text{sgn}(\omega), & \omega>T.\end{cases} \quad (2)$$

These results are thus consistent with the conclusion that there is an important difference between the Raman-scattering processes which shape the continuum in insulating and superconducting $YBa_2Cu_3\,O_{6+x}$ crystals. One might suggest that the continuum in the Raman spectra observed in the insulating phase is due to a scattering of the light by charge carriers localized at various trapping centers.

The features observed in the electron scattering in the superconducting crystals at $T<T_c$ agree qualitatively with the model of a two-component Fermi liquid which was analyzed in Ref. 7. According to that model, free charge carriers in the normal and superconducting phases contribute to the electron scattering. As a result, $r(\omega)$ would have a finite value at $\omega \lesssim 25$ cm^{-1}, as is seen experimentally. The differences between the polarizations $(x'x')$ and $(x'y')$ in terms of the position of the 2Δ peak and also in terms of the shape of the spectra at low frequencies are unambiguous evidence that the superconducting gap in the $YBa_2Cu_3O_7$ crystals is very anisotropic.

At low frequencies in the normal phase, on the other hand, there is a difference in the $R''(\omega,T)$ behavior for the different polarizations. The existing models used for a microscopic analysis of the mechanisms for electron scattering in high T_c crystals (the model of a strong electron–phonon coupling[8] and the model of a nested Fermi liquid[9]) are unfortunately incapable of explaining the entire set of experimental observations. Further research and the construction of a successful theoretical model are thus required for reaching an understanding of the properties of the free carriers in the highly correlated electron system in the normal state.

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[2)]Some special experiments established that the intensity of the electron scattering of the superconducting crystals at frequencies $\omega \gtrsim 700$ cm^{-1} is independent, within 10%, of the temperature over the entire temperature range studied.

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Translated by D. Parsons