Electron Raman scattering in $YBa_2Cu_3O_{6.5}$ crystals

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A detailed study has been made of electron scattering of light by $YBa_2Cu_3O_{6.5}$ single crystals with a sharp superconducting transition at $T_c \approx 53$ K. In the superconducting state, the spectral density of the electron continuum undergoes a restructuring at frequencies $\omega \lesssim 250$ cm^{-1} in the $(x'x'$ polarization), and a 2Δ peak arises at $\omega \approx 200$ cm^{-1}. In crossed polarizations $(x'y')$ the spectrum has a low intensity, and it does not undergo any significant changes at temperatures below T_c, in contrast with the spectrum of $YBa_2Cu_3O_7$.

Studies of electron Raman scattering in crystals of high-T_c superconductors in their superconducting and normal states can provide information on the nature of the electronic excitations in these materials and their symmetry.[1] When the material goes into the superconducting state, the low-energy part of the spectra of inelastic scattering of light undergoes a significant restructuring: The intensity of the electron continuum in the frequency region decreases, and a peak arises at a frequency close to the size of the superconducting gap, 2Δ (Ref. 2). The symmetry of this gap is manifested in measurements in different polarizations. Numerous detailed studies have been made of electron Raman scattering of $YBa_2Cu_3O_{6+x}$ samples (see, for example, the papers cited in Ref. 1). In the overwhelming majority of these studies, the test samples either had an oxygen content $x \approx 1$ and a transition temperature close to the maximum or were insulators. The spectra of the electron Raman scattering in high-quality crystals with $T_c \simeq 92$ K and a transition width $\Delta T_c \lesssim 1$ K, including single-domain crystals, clearly reveal a 2Δ peak at $T < T_c$. The position of this peak depends on the scattering geometry [$\omega_{max} = 350$ and 450 cm^{-1}, respectively, in the $(x'x')$ and $(x'y')$ polarizations, where the x' and y' axes are rotated 45° with respect to the crystallographic **a** and **b** axes].[3,4] This result is evidence that the superconducting gap in this compound is strongly anisotropic.

At the same time, the question of a correlation between T_c and the size of the superconducting gap, i.e., between T_c and the position of the 2Δ peak, is important for reaching an understanding of the nature of high-T_c superconductivity. In principle, experiments can answer this question in the case of $YBa_2Cu_3O_{6+x}$, since the transition temperature varies over a broad range, from zero up to a maximum $T_c \approx 92$ K, as a function of the oxygen content in the crystals. However, we can cite only a few studies of electron Raman scattering in crystals with an intermediate oxygen content and, correspondingly, a lower superconducting transition temperature.[5,6] Unfortunately, the results which have been found on samples with a lower value of T_c (Ref. 5) are not adequate for drawing an unambiguous conclusion about a correlation between the spectral position of the "gap" features are T_c, apparently because of pronounced variations in the oxygen concentration in the samples. This interpretation is supported, in particular, by the presence of a wide superconducting transition, with $\Delta T_c \approx 15$ K. Experiments on electron Raman scattering involving a variation in the oxygen concentration around the optimum value—that corresponding to the maximum T_c—have revealed that the spectral position of the 2Δ peak with A_{1g} symmetry[1)][in the $(x'x')$ scattering geometry] remains the same for all samples, while the 2Δ peak with B_{1g} symmetry [in the $(x'y')$ scattering geometry] undergoes a very large shift down the frequency scale even upon a very slight deviation from the optimum oxygen concentration.[6]

In this letter we are reporting measurements carried out on two $YBa_2Cu_3O_{6.5}$ single crystals with reduced transitions temperature $T_c \approx 53$ K (sample 1) and $T_c \approx 52$ K (sample 2), which have a fairly sharp transition, of width $\Delta T_c \lesssim 1$ K (see the insets in Figs. 2 and 3). The samples had a mirror-finish surface and typical dimensions of $1 \times 1 \times 0.2$ mm. Because of the sharp superconducting transition and some special measurements of Raman spectra at a high spatial resolution ($\lesssim 1.5$ μ) on various parts of the surface, we were able to conclude that the oxygen concentration in our samples is highly uniform.

Spectra of the electron Raman scattering were measured in an optical helium constant-temperature chamber with an adjustable temperature $T = 1.5-300$ K in the geometry of pseudoback-scattering from a fixed region on a surface of the crystal corresponding to the **ab** plane. The Raman scattering was excited by the line $\lambda = 4880$ Å of an Ar^+ laser. The power density in the excitation spot was $P \simeq 20$ W/cm^2 in the experiments in He vapor and $P < 4$ W/cm^2 in superfluid He. The intensity of the laser excitation was held constant within $\lesssim 1\%$ during the recording of the Raman spectra. The spectra were recorded on a Dilor XY triple monochromator with a multichannel optical detector.

Figure 1 shows Raman spectra of $YBa_2Cu_3O_{6.5}$ (sample 1) at two temperatures: above and below T_c. In addition to the narrow lines corresponding to optical phonons, there is a broad electron continuum in this spectrum. Below T_c, there is a redistribution of the intensity in the continuum in the spectra, like that which has been observed[3,4] in $YBa_2Cu_3O_7$, but the region in which the spectral changes occur is at lower frequencies, and the magnitude of the effect is itself considerably smaller.

To give a detailed picture of the temperature-induced changes in the Raman spectra, we plot the ratio $r(\omega,T) = R''(\omega,T)/R''(\omega,100\text{ K})$ in Figs. 2 and 3, where $R''(\omega,T)$ is the imaginary part of the electron-response function, which is related to the experimental Raman spectrum $S(\omega,T)$ by

$$S(\omega,T) = [1 + n(\omega,T)]R''(\omega,T),$$

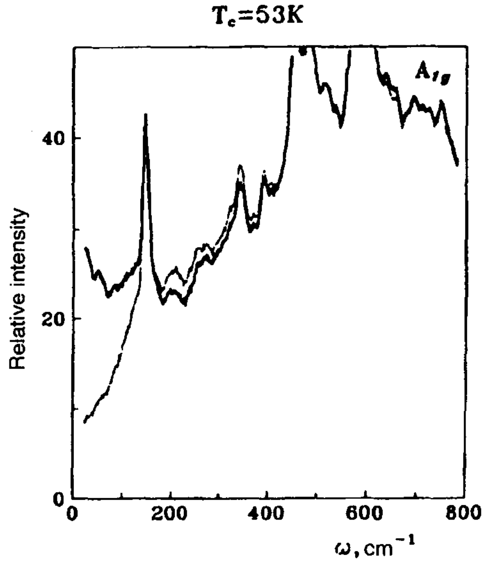

FIG. 1. Raman scattering spectra in a $YBa_2Cu_3O_{6.5}$ crystal (sample 1) in the $(x'x')$ polarization at two temperatures. Heavy curve—100 K; light curve—30 K.

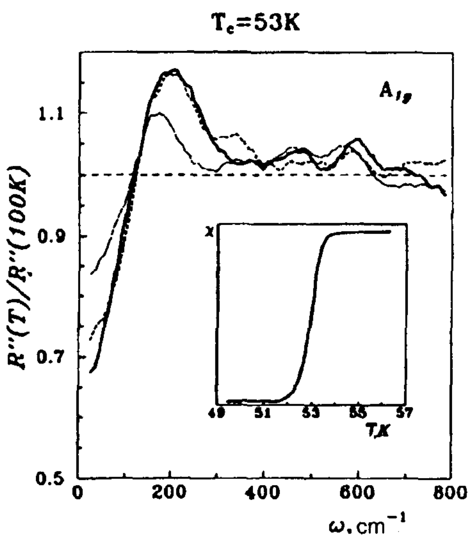

FIG. 2. The ratio $R''(\omega,T/R''(\omega,100\text{ K})$ for sample 1 in the $(x'x')$ polarization at various temperatures. Heavy curve—$\lesssim 4$ K; dashed curve—30 K; light curve—50 K. The inset shows the temperature dependence of the magnetic susceptibility, $\chi(T)$.

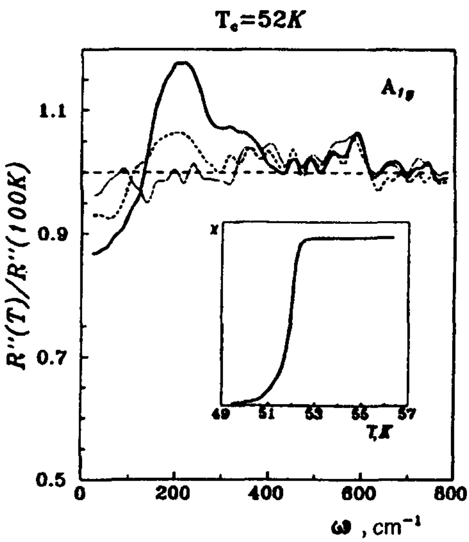

FIG. 3. The ratio $R''(\omega,T)/R''(\omega,$ 100 K) for sample 2 in the $(x'x')$ polarization for several temperatures. Heavy curve–30 K; dashed curve—50 K; light curve—70 K. The inset shows the temperature dependence of the susceptibility, $\chi(T)$.

where $n(\omega,T)=[\exp(\omega/T)-1]^{-1}$ is the Bose factor. Working from the Raman spectra in the Stokes and anti-Stokes regions, we estimated the heating of the sample in the excitation spot. In most cases the magnitude of the this heating in these experiments satisfied $\delta T \lesssim 30$ K within ± 5 K (Ref. 7). The temperatures shown in the figures incorporate this heating. Some special measurements were carried out in superfluid He at a reduced laser-excitation power, so that the temperature in the spot definitely did not exceed $T<4$ K. The results on $R''(\omega)$ at $T\approx 4$ K were multiplied by the corresponding ratio of laser pump intensities in order to refer the ratio $r(\omega,T)$ to the same scale.

It can be seen from Figs. 2 and 3 that a change in temperature is accompanied by a redistribution of the intensity in the normalized $R''(\omega,T)$ spectra in the manner characteristic of the onset of a superconducting gap:

We find $r(\omega,T)<1$ at low frequencies and $r(\omega,T)\approx 1$ (i.e., very slight changes in the spectra) at higher frequencies.

A 2Δ peak forms $[r(\omega,T)>1]$ in the $(x'x')$ polarization. The spectral position of this peak at low temperatures is $\omega_{max}\approx 200$ cm^{-1}.

As the temperature approaches the superconducting transition temperature T_c, the 2 Δ peak shrinks, shifts down the frequency scale, and disappears completely at temperatures above T_c (Fig. 3).

Working from the value of ω_{max} in the YBa_2Cu_3 $O_{6.5}$ crystals in the A_{1g} symmetry, we find a ratio $2\Delta/T_c\approx 5.5$. This is essentially the same as the value of this ratio in $YBa_2Cu_3O_7$ crystals.[4]

We do not observe any significant spectral changes in $R''(\omega,T)$ in the polarization

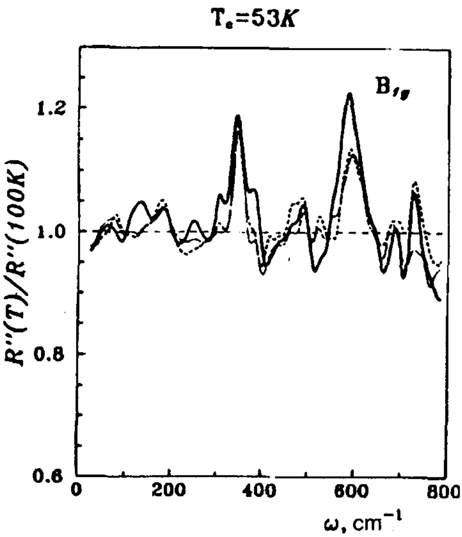

FIG. 4. The ratio $R''(\omega,T)/R''(\omega,\ 100\ \mathrm{K})$ for sample 1 in the $(x'y')$ polarization at several temperatures. Heavy curve—$\lesssim 4$ K; dashed curve—30 K; light curve—50 K.

$(x'y')$ in Fig. 4. We should point out that in this polarization the signal representing the electron-Raman scattering is considerably weaker (by a factor ~ 4) than the signal in the $(x'x')$ geometry. As a result, there is a larger normalization error. The peaks at frequencies $\omega \simeq 300$–350 and 550–600 cm^{-1} arise from the temperature dependence of the phonon of B_{1g} symmetry, with a frequency $\omega \approx 335$ cm^{-1}, which is allowed in the Raman spectra and from the temperature dependence of the phonon with $\omega \approx 600$ cm^{-1}, which is forbidden in these spectra.

The results thus show that the ratio $2\Delta/T_c \approx 5-6$ found from the spectra of the electron Raman scattering remains constant in $YBa_2Cu_3O_{6+x}$ as their T_c is varied from $\simeq 50$ to 92 K. In addition, the absence of any significant manifestations of a superconducting gap with a symmetry B_{1g} indicates that the superconducting gap for YBa_2Cu_3 $O_{6.5}$ has an A_{1g} symmetry. In a sense, these data agree with the results of Ref. 6, where it was demonstrated that even a very slight deviation from the optimum oxygen concentration leads to a substantial decrease in the frequency of the 2Δ peak in B_{1g} symmetry. The result of the present experiments, combined with the data of Ref. 6, suggest that the superconducting gap in YBa_2Cu_3 O_{6+x} becomes significantly anisotropic only near the optimum doping level.

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[1)]Since the orthorhombic distortions in $YBa_2Cu_3O_{6+x}$ are comparatively small, notation of the type A_{1g}, B_{1g} is often used in the literature for the case of orthorhombic high-T_c superconducting crystals, although this notation, strictly speaking, corresponds to the tetragonal phase.

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