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Optical Study of Localization in the *ab*-Plane Conductivity of Single Crystals of $YBa_2Cu_3O_{6.95}$ Induced by Ion Damage

D. N. Basov,[1,2] A. V. Puchkov,[1] R. A. Hughes,[1] T. Strach,[1] T. Timusk,[1] D. A. Bonn,[3] R. Liang,[3] and W. N. Hardy[3]

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We studied the changes in the dc resistivity, Raman scattering, and IR conductivity of single crystals of $YBa_2Cu_3O_{6.95}$ induced by damage from low-energy He^+ ion bombardment. It appears that T_c, transport properties, and optical conductivity are strongly affected by modest irradiation doses whereas the chemical composition of the sample is not modified. Carrier localization is evidenced by the frequency dependence of the optical conductivity in the strongly damaged crystal and is used to explain the suppression of both the superfluid density and T_c upon irradiation. The temperature dependence of the superfluid density is in agreement with the theoretical predictions for a *d*-wave superconductor. We also show that the intrinsic residual losses in the FIR are dramatically reduced in the disordered crystal.

KEY WORDS: Optical conductivity; localization; radiation damage; *d*-wave superconductivity.

We have studied the influence of defect scattering on the transport and optical properties of high-quality single crystals of $YBa_2Cu_3O_{6.95}$. We show that ion-beam irradiation does not change the oxygen content of the material but induces carrier localization which is responsible for the suppression of both T_c and the superfluid density.

High-quality single crystals of $YBa_2Cu_3O_{6.95}$ with perfectly smooth *ab*-faces were grown using a flux technique [2]. The crystals show a 0.5 K wide transition at 93.5 K, demonstrate a sharp peak in the heat capacity [2], and have an abrupt drop in microwave losses at T_c [3]. These crystals are almost defect-free since the value of the scattering rate at $T = 4$ K is as small as 1 cm^{-1} [3]. The *ab*-face of the crystal was irradiated with 160-keV He^+. Simulations using TRIM program show that ions of the above energy homogeneously damage the top 400 nm of the crystal's surface. The transport properties, however, cannot be evaluated from the data on the irradiated crystal since the bulk of the sample is undamaged. Laser-ablated *c*-axis thin films of $YBa_2Cu_3O_{7-x}$ on (100) $LaAlO_3$ substrates were used to obtain the correlation between ion dose and the reduction of T_c.

Figure 1a shows that the temperature dependence of the resistivity of 40-nm $YBa_2Cu_3O_{7-x}$ films remains linear for dose levels less than 10^{15} He/cm^2 but T_c is suppressed. In agreement with the results of Valles *et al.* [1], the absolute resistivity is increased by a constant amount, proportional to the dose level. Based on the thin-film data, we expect a reduction in the crystal's critical temperature from 93.5 K before irradiation down to 85, 80, and 70 K with dose levels of 2, 4, and 8×10^{14} He/cm^2, respectively.

Oxygen stoichiometry within the damaged layer of the irradiated crystal has been tested by means of Raman scattering. The position of the apical oxygen line at 500 cm^{-1}, which is sensitive to the oxygen content [4], shows no noticeable oxygen reduction due to the ion damage (Fig. 1b).

In Fig. 2 we show a series of conductivity spectra for different damage levels on the same crystal at a

[1]Department of Physics and Astronomy, McMaster University, Hamilton, Ontario, Canada L8S 4M1.
[2]On leave from P. N. Lebedev Physics Institute, Russian Academy of Sciences, Moscow, Russia.
[3]Department of Physics, University of British Columbia, Vancouver, B.C., Canada V6T 2A6.

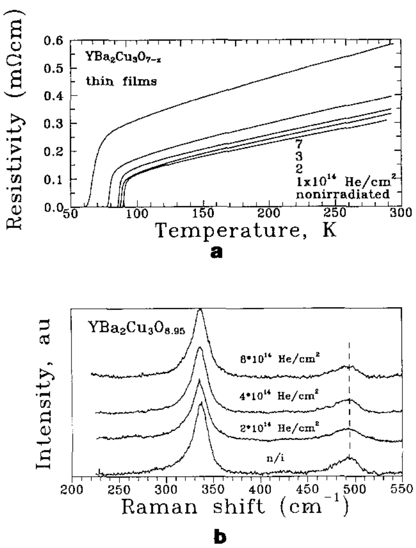

Fig. 1. (a) Temperature dependence of the dc resistivity of $YBa_2Cu_3O_{7-x}$ thin films irradiated with 160-keV He^+ ions. (b) Raman spectra from the *ab*-face of a He^+ irradiated single crystal of $YBa_2Cu_3O_{6.95}$.

temperature slightly above T_c and at 10 K. The conductivity was obtained through the Kramers–Kronig analysis of the reflectance which was measured in the frequency range between 30 and 14,000 cm^{-1}. The

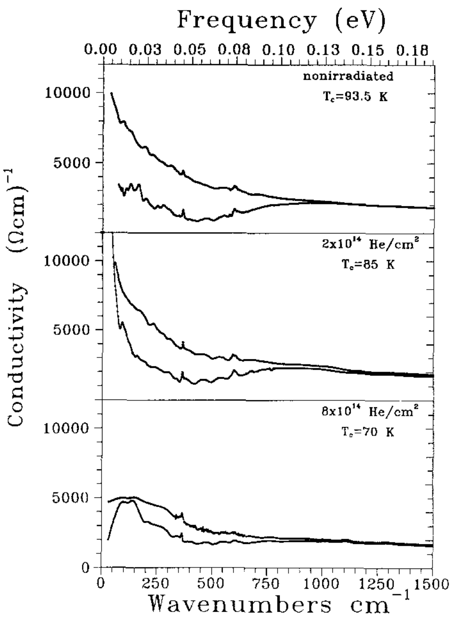

Fig. 2. Frequency dependence of the optical conductivity of the He^+-irradiated single crystal of $YBa_2Cu_3O_{6.95}$. Upper curves in all panels were obtained at a temperature slightly above T_c while bottom curves were obtained at 10 K.

superconducting-state conductivity of the nonirradiated crystal shows residual absorption down to the lowest frequencies. After light damage, the quasiparticle scattering rate is increased from 1 cm^{-1} in the pure sample (as obtained from the microwave measurements [3]) up to 70 cm^{-1} (as shown by the narrow feature, centered at zero frequency in the FIR conductivity). Even though the value of the scattering rate is no longer negligible, energy-gap structure is not observed in the FIR conductivity, confirming recent results on other disordered high-T_c materials [5]. Therefore, we may attribute the failure to observe the spectroscopic gap, not to the lack of momentum conserving processes in the clean limit, but to the unconventional response of $YBa_2Cu_3O_{6.95}$. A picture involving an anisotropic gap with states extending down to the lowest energy (*d*-wave superconductivity [6,7]) may be more relevant to these results.

At the highest damage level the superconducting-state conductivity shows a peak centered at about 100 cm^{-1}, which is also seen in the normal-state data. This behavior is the signature of a system with localized carriers and has been observed in disordered doped semiconductors [9]. Since the carriers responsible for the residual absorption are localized in the strongly disordered $YBa_2Cu_3O_{6.95}$ crystal, we expect a substantial decrease in the microwave losses for the irradiated material. This follows from the proportionality of the superconducting state absorptivity to the value of $\sigma_1(\omega)$. The value of the conductivity is reduced by a factor of 2 in the strongly disordered crystal as compared to the pure one at our lowest reliable frequency of 30 cm^{-1}. The effect at the microwave frequencies should be even more obvious since the $\sigma_1(\omega)$ associated with the localized carriers decreases as frequency is lowered.

Figure 3 illustrates the changes seen in the superfluid spectral weight as a result of irradiation. We discuss this in terms of the plasma frequency of the superconducting carriers, ω_{ps}. The absolute value of ω_{ps} is suppressed upon irradiation, as shown in the inset. The corresponding values of the penetration depth are 144, 174, 180, and 277 nm for doses of 0, 2, 4, and 8×10^{14} He/cm^2, respectively. Thus, the irradiation significantly reduces the number of condensed carriers while the total carrier density is hardly affected. The correlation between T_c and ω_{ps}^2 does not follow the linear dependence starting at the origin established for a variety of high-T_c materials at various levels of doping [12]. This is in agreement with our Raman results which suggest that the mechanisms

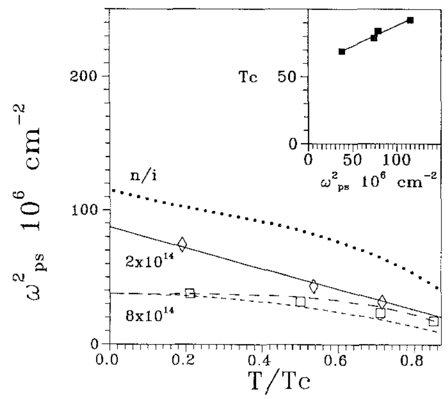

Fig. 3. Temperature dependence of ω_{ps}^2 for the crystal after different doses of irradiation. Dots—microwave result for the crystal from the same batch scaled to the value of $\lambda_L(0)=144$ nm obtained from FIR measurements of the nonirradiated crystal; rhombs and squares—FIR data for the irradiated crystal. Long dashed line and short dashed line: $1-(T/T_c)^4$ and $1-(T/T_c)^2$ dependences, respectively, scaled to the values of ω_{ps}^2 at $T=10$ K. Inset: correlation between the values of the critical temperature and ω_{ps}^2 for the single crystal of $YBa_2Cu_3O_{6.95}$ after different doses of irradiation.

of T_c suppression by the reduction of carriers and by ion irradiation are of different origin.

We believe that carrier localization, suggested by the frequency dependence of the conductivity at the highest dose level, may explain the suppression of the superfluid density and the lowering of T_c by ion damage. We emphasize that these effects are not due to oxygen deficiency, but may be attributed to the destroyed superconducting coherence when the electronic states near the Fermi level are localized [13]. The relatively low defect concentration, which induces localization in the single crystal of $YBa_2Cu_3O_{6.95}$, may be understood as a result of the quasi-2D character of the electronic properties of the high-T_c materials which in general favors localization.

The temperature dependence of ω_{ps} is shown in Fig. 3. As we have shown elsewhere, the correct determination of such a dependence for a superconductor with a narrow normal component in the superconducing conductivity is possible when the condensate is probed at frequencies below the value of the scattering rate $\hbar/\tau$ of this component [10]. This condition is satisfied only at microwave frequencies for the pure crystal or at the lowest FIR frequencies in the case of the lightly damaged sample. Under the above condition, one may write $\omega_{ps}=(\omega\sigma_2(\omega, T))^{1/2}$ so that ω_{ps} can be determined from the Kramers–Kronig analysis of the reflectivity for $\sigma_2(\omega)$. Both the microwave [11] and FIR measurements reveal the linear dependence at T below $T_c/2$, in agreement with the theoretical predictions for a d-wave superconductor [6–8]. In the strongly disordered crystal we found that the T dependence of ω_{ps}^2 has the form $1-(T/T_c)^n$ with n between 2 and 4 at low temperatures, which accords with what is expected within the d-wave model.

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