# Disorder and superconducting-state conductivity of single crystals of $YBa_2Cu_3O_{6.95}$

D. N. Basov, A. V. Puchkov, R. A. Hughes, T. Strach, J. Preston, and T. Timusk
*Department of Physics and Astronomy, McMaster University, Hamilton, Ontario, Canada L8S 4M1*

D. A. Bonn, R. Liang, and W. N. Hardy
*Department of Physics, University of British Columbia, Vancouver, British Columbia, Canada V6T 2A6*
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The *ab*-plane optical conductivity of $YBa_2Cu_3O_{6.95}$ has been studied by infrared reflectance spectroscopy in the presence of disorder introduced by ion irradiation. The frequency dependence of the conductivity in the superconducting state suggests that ion irradiation has caused carrier localization. Localization explains both the suppression of $T_c$ and the reduction of the superfluid fraction. The temperature dependence of the penetration depth in pure and disordered $YBa_2Cu_3O_{6.95}$ supports the hypothesis of non-*s*-wave superconductivity.

High-$T_c$ materials can undergo a metal-insulator transition, either through reduced carrier concentration by chemical doping or by the addition of scattering from defects introduced by ion irradiation.[1] In this paper we show evidence that low energy ions have an advantage, since they influence the normal-state transport and suppress the critical temperature without modifying the chemical composition of the sample. This opens up new possibilities for studying the relationship between normal- and superconducting-state properties in materials with identical chemical compositions. Moreover, following Anderson's theorem,[2] nonmagnetic impurities are expected to have negligible influence on superconductivity in the case of *s*-wave pairing whereas a superconductor with an unconventional pairing state (*p* or *d* wave) is strongly affected by impurities.[3] Thus, a study of disorder may provide an experimental test of the nature of the superconducting state in high-$T_c$ materials.

We focused on changes in the *ab*-plane optical conductivity of $YBa_2Cu_3O_{6.95}$ single crystals resulting from disorder introduced by ion irradiation. The conductivity of the damaged crystal is consistent with carrier localization. This allows us to explain a variety of effects resulting from the ion irradiation of $YBa_2Cu_3O_{6.95}$ (YBCO). The temperature dependence of the superfluid density changes from a linear law at low temperature in the pure crystal to $1-(T/T_c)^n$ with $n$ between 2 and 4 in the strongly damaged sample suggesting non-*s*-wave superconductivity in the YBCO system.

High quality single crystals of $YBa_2Cu_3O_{6.95}$ with mirror smooth *ab* faces were grown using a flux technique.[4] Inductively coupled plasma (ICP) mass spectroscopy shows impurities at the 0.1 level. The crystals have a 0.5 K wide transition at 93.5 K and demonstrate a sharp peak in heat capacity[4] and an abrupt drop in microwave losses at $T_c$.[5] These crystals are almost defect free since the scattering rate at $T = 4$ K is as small as 1 $cm^{-1}$.[6]

The *ab* face of the crystal was irradiated with 160 keV $He^+$ ions which provide homogeneous damage to a depth of 400 nm.[7] The penetration depth along the *c* axis in the superconducting state is enhanced in the irradiated samples, but remains less than 400 nm so that the reflectivity spectra measured from the *ab* face of the crystal are solely determined by the optical constants of the damaged layer. While the thickness of the damaged layer is sufficient for the infrared spectroscopy, measuring the transport properties is problematic. We irradiated *c* axis $YBa_2Cu_3O_{7-x}$ thin films to evaluate the correlation between the ion dose and the critical temperature of the irradiated YBCO system. Figure 1 shows that the temperature dependence of the resistivity of 40 nm thick $YBa_2Cu_3O_{7-x}$ films remains linear for dose levels of less than $10^{15}$ He/cm$^2$, but with suppressed $T_c$. In agreement with the results of Valles *et al.*,[1] the absolute resistivity is increased by a constant amount, proportional to the dose level. We observed similar effects induced by ion irradiation in 200 nm thick films. Based on the thin film data, we expect a reduction in the crystals' critical temperature from 93.5 K before irradiation down to 85, 80, and 70 K with dose levels of 2, 4, and 8 $\times 10^{14}$ He/cm$^2$, respectively.

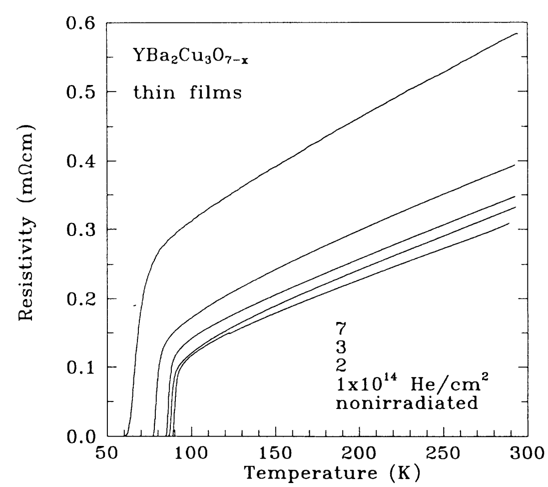


FIG. 1. Temperature dependence of the dc resistivity of the *c*-axis films of $YBa_2Cu_3O_{7-x}$ irradiated with $He^+$ ions. Ion irradiation causes the enhancement of the absolute value of the resistivity and a suppression of the critical temperature.

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Partial oxygen loss has been observed as the result of irradiation with ions in the 10 MeV energy range.[8] Optical measurements allow us to monitor the oxygen content in the irradiated layer. Independent of model, the oscillator strength sum rule states that the area under the conductivity spectrum is unambiguously related to the carrier density and thus to the doping level. The analysis of the total spectral weight in the irradiated crystal (top panel of Fig. 4) shows that the oxygen losses are too small to explain the observed suppression of $T_c$.

Another check of the oxygen stoichiometry in the damaged layer of the crystals is the position of the apical oxygen line at 500 $cm^{-1}$ in Raman spectra.[9] We measured spectra at room temperature for the nonirradiated crystal and for the same crystal after each irradiation dose (Fig. 2). The frequency of the apical oxygen line did not shift upon ion irradiation. Thus, both experimental methods show that 160 keV $He^+$ ions, at the doses used here, do not change the doping level of the $YBa_2Cu_3O_{6.95}$ crystals. Instead, it is the *disorder itself* that is the dominant factor determining the superconducting properties of the damaged material.

Reflectivity spectra of the unirradiated crystal and of the same crystal irradiated with doses of 2, 4, and $8\times10^{14}$ He/$cm^2$ were obtained from the *ab* face in the spectral range between 30 and 14 000 $cm^{-1}$. The spectra were measured at room temperature, at a temperature slightly above $T_c$ , at 10 K, and at various temperatures below $T_c$. The optical conductivity was derived by Kramers-Kronig analysis using data from Ref. 10 as a high frequency extension. In the superconducting state the two-fluid model was used for the extrapolation below 30 $cm^{-1}$.

In Fig. 3 we show a series of conductivities as a function of frequency at a temperature slightly above $T_c$ and at 10 K for the same crystal but for different damage levels. The experimental sensitivity is demonstrated by the presence of the weak structure due to phonons in the *ab* plane which is resolved well above the noise level.

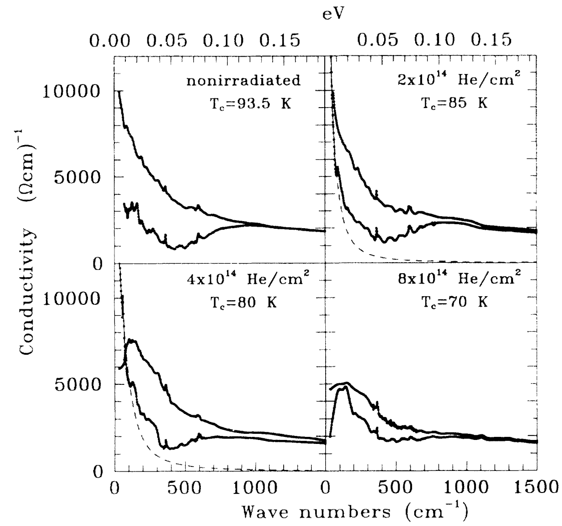


FIG. 3. Frequency dependence of the optical conductivity of the $He^+$ irradiated single crystal of $YBa_2Cu_3O_{6.95}$. Upper curves in all panels were measured at a temperature slightly above $T_c$, while the bottom curves were obtained at 10 K. The dashed lines show the fit of the narrow normal component in the superconducting conductivity to the Drude formula.

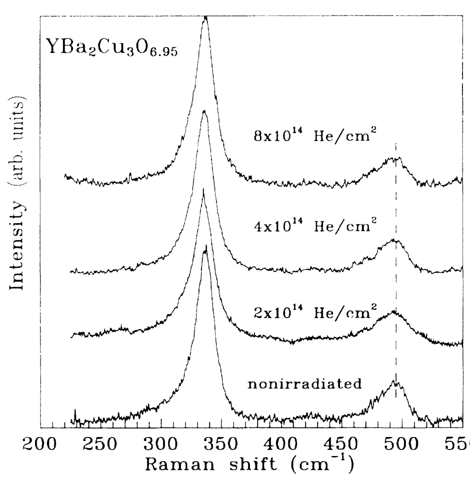


FIG. 2. Raman spectra measured from the *ab* face of the single crystal of $YBa_2Cu_3O_{6.95}$. Spectra were obtained for the nonirradiated sample and for the same sample after each irradiation dose. The position of the apical oxygen line at 500 $cm^{-1}$ shows no frequency shift suggesting that ion damage does not change the oxygen content in the damaged layer of the crystal.

In the superconducting state, the conductivity of the unirradiated crystal is in agreement with earlier reflectance[11,12] and direct absorption data.[13,14] Down to the low frequency limit of our measurements, the conductivity is finite showing residual losses and no energy gap. The high quality of the crystal suggests that losses are intrinsic to $YBa_2Cu_3O_{6.95}$ and are not due to sample imperfections.

We first discuss the conductivity in the superconducting state after light and intermediate damage levels (doses 2 and 4 $\times$ $10^{14}$ He/$cm^2$). Both conductivities are almost identical to that of the undamaged crystal except for the appearance of a new, quite narrow feature, centered at zero frequency, with the scattering rates of 50 $cm^{-1}$ and 75 $cm^{-1}$ for doses of 2 and 4 $\times$ $10^{14}$ He/$cm^2$, respectively. The dashed lines in Fig. 3 were calculated using the Drude formula [second term of the Eq. (1)] with the values of the plasma frequency $\omega_p$ and the scattering rate being equal to $\omega_p$=7400 $cm^{-1}$, $\hbar/\tau$=50 $cm^{-1}$ and $\omega_p$=8200 $cm^{-1}$, $\hbar/\tau$=75 $cm^{-1}$ for the crystals irradiated at dose levels of 2 and 4 $\times$ $10^{14}$ He/$cm^2$, respectively. A simple explanation of this behavior is that the narrow Drude-like quasiparticle component, seen in the microwave region,[6] has broadened, increasing from 1 $cm^{-1}$ in the pure crystal to 50 and then 75 $cm^{-1}$ in the irradiated crystal making the corresponding feature observable at far-infrared (FIR) frequencies. The increase in the plasma frequency of the normal component at $T$ = 10 K (and the apparent decrease of the superfluid weight that will be discussed below) indicates that defect scattering induced by ion damage is accompanied by a pair-breaking effect. Thus, it can be concluded that the single particle density of states is enhanced in the disordered crystal which is in general agreement with the recent study of impurity effects in a $d$-wave superconductor.[15]

Since, at modest damage levels, $\hbar/\tau$ is no longer negligibly small compared to $kT_c$ one may expect to observe gap structure in the FIR conductivity. But instead, the conductivity of the irradiated crystal shows the development of a low-lying Drude component indicating that the $YBa_2Cu_3O_{6.95}$ superconductor is gapless. Thus, it appears that the failure to observe a spectroscopic gap is not because of the lack of momentum conserving processes due to the clean limit,[16] but due to the unconventional nature of the response of $YBa_2Cu_3O_{6.95}$. A similar conclusion was obtained recently from the studies of YBCO and $Bi_2Sr_2CaCu_2O_8$ thin films transformed to the dirty limit through doping with Ni and Y ions respectively.[17,18] A picture involving an anisotropic gap with states extending down to the lowest energy such as a $d$-wave model[19–23] may be more appropriate.

At the highest damage level, drastic changes occur in the real part of the complex conductivity $\sigma(\omega) = \sigma_1(\omega) + i\sigma_2(\omega)$ in the superconducting state at low frequency. Instead of $\sigma_1(\omega)$ monotonically decreasing with frequency at low frequencies (i.e., Drude behavior), a peak appears, centered at 100 $cm^{-1}$, which also can be 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.[24–26] We note that the position of the peak is far from the region where the superconducting state conductivity can be influenced by the choice of low frequency extrapolation and is therefore not an experimental artifact. It should also be emphasized that the observed qualitative change in the frequency dependence of $\sigma(\omega)$ can be unambiguously attributed to the localization of carriers in $CuO_2$ planes and not in the chains despite the twinned crystal structure of our sample. This follows from the recent studies on single domain samples of YBCO which show that the chain contribution to the conductivity is in the form a peak centered at 300 $cm^{-1}$, or even at midinfrared frequencies and has no Drude-like feature.[27,28] Finally, the localization behavior of the $ab$-plane conductivity cannot originate from disorder in the CuO chains, since even oxygen deficient samples show a Drude-like $\sigma(\omega)$ at low frequencies.[29]

Since the residual absorption in the strongly disordered crystal is determined by the localized carriers, ac losses at microwave frequencies are expected to be dramatically suppressed. This follows from the proportionality of microwave losses to the value of $\sigma_1(\omega)$ which is *decreasing* with lowering frequency.[6]

Figure 4 shows the changes to the total spectral weight and to the superfluid component as a result of irradiation. The top panel shows the correlation between the total spectral weight $N_{\rm norm}$ and the critical temperature of the crystal where the latter was altered by ion irradiation and by oxygen removal.[29] In both cases the spectral weight obtained from the integration of the normal state conductivity was normalized to that of the crystal with highest $T_c$ (undamaged or fully oxygenated, respectively). For the crystal with the highest damage, where the finite thickness of the irradiated layer may cause distortions to the normal state $\sigma(\omega)$, we have integrated the conductivity at $T = 10$ K and then added the weight of the superfluid found from the imaginary part of the conductivity as described below. It is clear from the plot that the carrier density remains almost unaffected in the damaged crystal contrary to that in the oxygen deficient samples where it is significantly reduced even for relatively small changes in $T_c$. The decrease by 10% of $N_{\rm eff}$ for the case of the irradiated material is most likely caused by the broadening of the midinfrared conductivity which will transfer spectral weight to the frequencies above the upper limit of the integration (2000 $cm^{-1}$). The comparison of the dependences for the irradiated and for the oxygen deficient samples suggests that possible oxygen losses in the damaged crystal are responsible for a suppression in $T_c$ of less than 5 K.

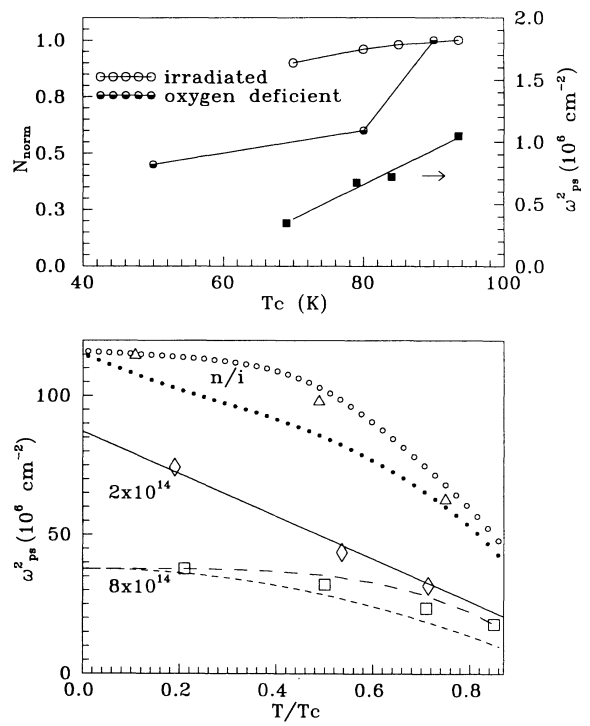


FIG. 4. Top panel: Correlation between the values of the critical temperature and the total spectral weight $N_{\rm norm}$ as obtained from the integration of the optical conductivity for the irradiated crystal (open circles) and for the oxygen deficient crystals (half-solid circles) (after Ref. 29). Also shown is the dependence of the squared value of the superfluid plasma frequency ($\omega_{ps}^2$) as a function of critical temperature for the single crystal of $YBa_2Cu_3O_{6.95}$ after different doses of irradiation (solid squares). Bottom panel: Temperature dependence of $\omega_{ps}^2$ for the crystal after different doses of irradiation: triangles, nonirradiated crystal; diamonds, crystal irradiated with dose level of $2 \times 10^{14}$ $He/cm^2$; squares, crystal irradiated with the dose level of $8 \times 10^{14}$ $He/cm^2$. The solid circles show the microwave result for the crystal from the same batch (Ref. 30) scaled to the value of $\lambda_L(0)$=144 nm. The open circles show the fit to the IR data for $\omega_{ps}^2$ using Eq. (1) as described in the text. The long dashed line and short dashed line illustrate the scaled $1-(T/T_c)^n$ dependences with $n$=4 and $n$=2, respectively.

The spectral weight of the superfluid is discussed in terms of the plasma frequency $\omega_{ps}$. For the case of London superconductor with penetration depth $\lambda_L$, one finds that $\omega_{ps} = 2\pi c/\lambda_L = [\omega\sigma_2(\omega, T)]^{1/2}$ and as a result the superfluid density may be obtained from the imaginary part of the complex conductivity. However, from Fig. 3 it is clear that the low frequency response of $YBa_2Cu_3O_{6.95}$ is influenced by the narrow normal component and therefore the complex conductivity, as a function of temperature and frequency, may be written as

$$\sigma(\omega, T) = \frac{i(2\pi c)^2}{\lambda_L^2(T)\omega} + \frac{\omega_p^2\tau/4\pi}{1+i\omega\tau} \quad \text{for} \quad \hbar\omega < 200 \text{ cm}^{-1} , \quad (1)$$

where $\omega_p$ is the plasma frequency of the Drude-like normal component, and $\hbar/\tau$ is its scattering rate. We note that at frequencies $\hbar\omega$ above $\hbar/\tau$ the imaginary part of the second term in Eq. (1) can no longer be neglected and the simple relationship between $\omega_{ps}$ and $\sigma_2(\omega, T)$ is no longer correct. The contribution of the second term in Eq. (1) to $\sigma_2$ is more obvious at intermediate temperatures between 10 K and $T_c$ when the value of $\omega_p$ is enhanced by thermal pair breaking. The condition of $\hbar\omega < \hbar/\tau(T)$ required to determine the *intrinsic* properties of the superfluid is satisfied for the pure crystal only at microwave frequencies[30] and for the lightly damaged crystal at our minimal reliable FIR frequencies (30 $\text{cm}^{-1}$). Both results show a *linear* dependence of the superfluid density below $T_c/2$. Meanwhile, the FIR measurements of the pure crystal revealed only a weak temperature dependence of $\omega_{ps}^2$ at low temperatures, a result often found with the IR method.[31,32] We argue that this weak dependence in the crystal under study *is not intrinsic* but instead is a consequence of the probing frequency $\hbar\omega$=30 $\text{cm}^{-1}$ being above the value of $\hbar/\tau$=1 $\text{cm}^{-1}$ before irradiation. The dependence observed in the pure crystal at $\hbar\omega$=30 $\text{cm}^{-1}$ is reproduced using Eq. (1) provided that the intrinsic dependence of $\hbar/\tau(T)$ and $\lambda_L(T)$ are taken from the microwave data.[6,30]

The linear temperature dependence of $\omega_{ps}^2$ found in the pure and lightly damaged crystal is no longer observed after the highest dose of irradiation and the latter clearly follows a $1-(T/T_c)^n$ law with $n$ lying in between 2 and 4. The sensitivity of the $\omega_{ps}(T)$ dependence to relatively weak disorder demonstrates the non-$s$-wave nature of the pairing state in YBCO. In particular, the $d$-wave model of superconductivity predicts a linear law for the penetration depth with a crossover to a $1-(T/T_c)^2$ dependence in the presence of impurity scattering.[19–23,33] This is in reasonable agreement with the results plotted in Fig. 4.

Not only does the temperature dependence of $\omega_{ps}$ change radically upon irradiation, its absolute value is strongly diminished by the ion damage. The top panel of Fig. 4 shows the correlation between $T_c$ and the squared plasma frequency of the superfluid obtained from $[\omega\ \sigma_2(\omega)\ ]^{1/2}$ at $\omega$=30 $\text{cm}^{-1}$. The corresponding values of the penetration depth are 144, 174, 180, and 277 nm for doses of 0, 2, 4, and $8 \times 10^{14}$ $\text{He/cm}^2$, respectively. Thus, irradiation significantly suppresses the number of condensed carriers while total carrier density is hardly affected. The correlation between $T_c$ and $\omega_{ps}^2$ does not follow the dependence established for a variety of high-$T_c$ materials at various levels of doping.[34] This is in agreement with our results on the total carrier density, which suggests that the mechanisms for $T_c$ suppression by the reduction of the carriers and by ion irradiation are of different origin.

We believe that carrier localization may be the cause of the reduction of the critical temperature as the result of irradiation. In a strongly disordered metal the density of states at the Fermi level is suppressed[35] while Coulomb repulsion is enhanced.[36] Both effects decrease the critical temperature. In addition, the role of fluctuations in suppressing $T_c$ is enhanced since the coherence length is reduced in the disordered superconductor.[37]

We briefly summarize the experimental observations that justify the applicability of a localization approach to describe the influence of ion damage on the critical temperature of $YBa_2Cu_3O_{6.95}$: (i) No loss of carriers is found in the irradiated crystal suggesting that de-oxygenation is not responsible for $T_c$ suppression; (ii) the frequency dependence of the residual conductivity suggests carrier localization; (iii) the superfluid density is reduced while the total spectral weight is almost unaffected (it has been shown by Ma and Lee[38] that localization itself, even neglecting the effect of Coulomb repulsion, dramatically suppresses the superfluid density, though superconductivity may still be observed in a system with localized states); (iv) after higher irradiation doses, when $T_c$ is further diminished, the temperature dependence of the resistivity is no longer linear but shows activated behavior.[1] We also point out that the low dimensionality of the electronic transport in high-$T_c$ materials favors localization. This might explain the anomalous sensitivity of the critical temperature in copper oxide materials to relatively small ion doses as compared to the $A15$ materials.[39] It is quite obvious that the defect concentration required to induce localization and thus to reduce $T_c$ should be much lower for quasi-two-dimensional (quasi-2D) high-$T_c$ compounds than it is for the essentially 3D $A15$ superconductors.

Our observations of a dependence of $T_c$ on the density of the superconducting carriers (top panel of Fig. 4) and of dramatic changes in the residual conductivity (Fig. 3) may be explained by the presence of strong *inelastic* pair breaking. It would overwhelm the effect of impurities at high temperature, but would disappear at low temperature where defects would then be the dominant effect leading to a reduced superconducting density.

In conclusion, we found that the degradation of the critical temperature and of the superfluid density in irradiated single crystals of $YBa_2Cu_3O_{6.95}$ may be a consequence of carrier localization. In addition, we have discussed the importance of finite frequencies of both IR and microwave methods for the problem of the correct determination of the intrinsic temperature dependence of the superfluid density. Our results on the superfluid density are in qualitative agreement with theoretical predictions for a $d$-wave superconductor. Finally, disorder has been found to change dramatically the residual absorption in the superconducting state implying that low loss microwave materials can be obtained by appropriate ion irradiation.

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