Coherence, incoherence, and scaling along the c axis of YBa$_2$Cu$_3$O$_{6+x}$

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The optical properties of single crystals of YBa$_2$Cu$_3$O$_{6+x}$ have been examined along the c axis above and below the critical temperature ($T_c$) for a wide range of oxygen dopings. The temperature dependence of the optically determined value of the dc conductivity ($\sigma_{dc}$) in the normal state suggests a crossover from incoherent (hopping-type) transport at lower oxygen dopings ($x \approx 0.9$) to more coherent anisotropic three-dimensional behavior in the overdoped ($x \approx 0.99$) material at temperatures close to $T_c$. The assumption that superconductivity occurs along the c axis through the Josephson effect yields a scaling relation between the strength of the superconducting condensate ($\rho_{c,c}$, a measure of the number of superconducting carriers), the critical temperature, and the normal-state c-axis value for $\sigma_{dc}$, just above $T_c$: $\rho_{c,c} \approx \sigma_{dc} T_c$. This scaling relation is observed along the c axis for all oxygen dopings, as well as several other cuprate materials. However, the Josephson coupling model does not require incoherent transport, so that the observed agreement with this model does not conflict with the normal-state results which point to coherent behavior at high oxygen dopings.

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I. INTRODUCTION

The cuprate-based high-temperature superconductors all share the common feature that the superconductivity is thought to originate within the highly conducting copper-oxygen planes. The conductivity perpendicular to the planes along the c axis is much poorer and is in fact activated in many cuprates; the resulting transport is due to hopping along this direction and the large anisotropy in the resistivity results in the two-dimensional (2D) nature of these systems.\textsuperscript{3} The importance of the 2D character of these materials as a necessary prerequisite for superconductivity has been discussed,\textsuperscript{2} and it has recently been suggested that upon entering the superconducting state a dimensional crossover from two to three dimensions occurs.\textsuperscript{4,5} Transport measurements on a number of cuprate systems suggest that such a dimensional crossover may occur in the normal state in response to carrier doping in the copper-oxygen planes.\textsuperscript{5-10} A convenient system to examine is YBa$_2$Cu$_3$O$_{6+x}$ (YBCO), where the oxygen doping determines not only the in-plane carrier concentration, but also the nature of the c-axis transport. This material is one of the most thoroughly studied, and there have been numerous reports of the response of the physical properties to changes in oxygen doping, including transport\textsuperscript{5,11} and optical techniques,\textsuperscript{12-19} to name a few.

In YBCO oxygen dopings below the optimal value ($x \approx 0.95$) for the critical temperature $T_c$, the dc resistivity along the c axis is $\rho_c \approx T^\alpha$, where $\alpha \approx -1$, indicative of activated behavior. In these underdoped materials, the anisotropy between the a-b planes and the c axis, as gauged by the resistivity, is quite large: $\rho_c/\rho_a \approx 65$ at room temperature, and increases rapidly with decreasing temperature to $\rho_c/\rho_a \approx 3000$ ($x \approx 0.70$) just above $T_c$.\textsuperscript{11} For these reasons, transport along the c axis in the normal state in the underdoped materials is governed by hopping and is considered to be incoherent; below $T_c$ superconductivity normal to the planes is thought to involve Josephson coupling.\textsuperscript{20-22} However, as the material becomes nearly stoichiometric, or “overdoped” ($x \approx 0.99$), the c-axis properties change dramatically. Unlike in the underdoped systems, the resistivity rises linearly with $T$, although with a large, temperature-independent component. Furthermore, $\rho_c/\rho_a \approx 30$ at room temperature, and is nearly temperature independent in the normal state,\textsuperscript{11} characteristic of an anisotropic three-dimensional (3D) metal. This behavior is even more pronounced in the Ca-doped material.\textsuperscript{23} In addition, the anisotropic resistivity of the double-chain material YBa$_2$Cu$_4$O$_8$ also suggests an incoherent-to-coherent crossover in the out-of-plane behavior.\textsuperscript{8} This suggests that transport along the c axis of YBCO is becoming more coherent at high oxygen dopings, and that a crossover from 2D to 3D behavior may occur; this could have important consequences for the nature of the superconductivity.

In this work we examine the optical properties along the c axis of YBa$_2$Cu$_3$O$_{6+x}$ for a wide range of oxygen dopings in the normal and superconducting states. The normal-state transport suggests that a dimensional crossover may occur in the overdoped YBCO samples. The strength of the condensate along the c axis is consistent with the Josephson coupling between the planes. However, because the Josephson effect may be observed for both coherent and incoherent transport along the c axis (tunnel and nontunnel junctions), the condensate does not necessarily favor incoherent transport in the overdoped material, where the normal-state properties suggest the system tends toward coherent behavior.
II. EXPERIMENT

Large, twinned single crystals of YBCO were grown by a flux method in yttria-stabilized zirconia crucibles and subsequently reannealed to a variety of oxygen contents from $x=0.50 \rightarrow 0.99$. The $T_c$s are sharply defined and vary over a large range, from a low of $T_c=53$ K in the most underdoped material ($x=0.50$) to a maximum of $T_c=93.2$ K in the optimally doped material ($x=0.95$). In the most overdoped sample ($x \approx 0.99$), $T_c$ is suppressed somewhat to $T_c \approx 90$ K. The reflectance polarized along the $c$ axis was measured over a wide frequency range and at a variety of temperatures using an overfilling technique. The real part of the optical conductivity $\sigma_1(\omega)$ was determined from a Kramers-Kronig analysis of the reflectance.

III. RESULTS AND DISCUSSION

A. Normal state

The optical properties along the $c$ axis have been thoroughly investigated by us and others, and will be discussed only briefly. The optical conductivity of YBCO is shown along the $c$ axis in Fig. 1 for $T \approx T_c$ at a variety of oxygen dopings. In the overdoped regime the conductivity has a metallic temperature and frequency dependence. However, with decreasing doping a pseudogap develops, and a nonmetallic activated response is observed. A convenient connection between the optical properties and transport is that $\sigma_{dc} = \sigma_1(\omega \rightarrow 0)$ as indicated in the plot and listed in Table I (for $T \approx T_c$). The extrapolated values for the resistivity $\rho_{dc}$ along the $c$ axis are shown in Fig. 2 for a variety of oxygen dopings at room temperature and $T \approx T_c$. At room temperature the resistivity increases exponentially with decreasing doping, $\rho_c \approx \rho_0 e^{-x/a}$, throughout the entire doping range. At low temperature the resistivity is observed to increase with decreasing temperature for $x \approx 0.85$, in agreement with the activated response observed in transport; once again the resistivity varies exponentially with doping. However, for $x \approx 0.9$ the resistivity decreases dramatically with increased doping and no longer follows the simple exponential relation. In addition, the temperature response of the resistivity is now "metallic," particularly so for the overdoped sample. This change in behavior also occurs close to the Mott maximum value for metallic behavior (ML), estimated to be $\rho_c \approx 10$ m$\Omega$ cm in these materials. The overdoped material is clearly the most metallic, and displays a Drude-like conductivity, an important result that was noted in earlier studies. The Drude model for the dielectric function describes the properties of a simple metal quite well, $\varepsilon(\omega) = \varepsilon_{sc} - \omega_p^2/[(\omega(\omega+i\Gamma))]$, where $\omega_p$ is the classical plasma frequency for YBa$_2$Cu$_3$O$_{6+\delta}$.

<table>
<thead>
<tr>
<th>$x$</th>
<th>$T_c$ (K)</th>
<th>$\sigma_{dc}$ (\Omega$^{-1}$ cm$^{-1}$)</th>
<th>$\omega_{ph,c}$ (cm$^{-1}$)</th>
<th>$\lambda_c$ (\mu m)</th>
</tr>
</thead>
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<td>78</td>
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<td>465±35</td>
<td>3.42</td>
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<tr>
<td>0.9</td>
<td>89</td>
<td>47±7</td>
<td>790±50</td>
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<td>0.95</td>
<td>93.2</td>
<td>220±20</td>
<td>1580±70</td>
<td>1.01</td>
</tr>
<tr>
<td>0.99</td>
<td>90</td>
<td>450±30</td>
<td>2070±90</td>
<td>0.77</td>
</tr>
</tbody>
</table>

* Taken at $\omega \rightarrow 0$ limit for $T \approx T_c$. 

FIG. 1. The real part of the optical conductivity for light polarized along the $c$ axis of YBa$_2$Cu$_3$O$_{6+\delta}$ at $T \approx T_c$ for a variety of oxygen dopings. The extrapolated values of the dc conductivity $\sigma_{dc} = \sigma_1(\omega \rightarrow 0)$ are shown as symbols. The most heavily doped resistivity is now “metallic,” particularly so for the overdoped sample. The Drude-like conductivity, an important result that was noted in earlier studies. This change in behavior also occurs close to the Mott maximum value for metallic behavior (ML), estimated to be $\rho_c \approx 10$ m$\Omega$ cm in these materials. The overdoped material is clearly the most metallic, and displays a Drude-like conductivity, an important result that was noted in earlier studies. The Drude model for the dielectric function describes the properties of a simple metal quite well, $\varepsilon(\omega) = \varepsilon_{sc} - \omega_p^2/[(\omega(\omega+i\Gamma))]$, where $\omega_p$ is the classical plasma frequency for YBa$_2$Cu$_3$O$_{6+\delta}$. The overdoped material is clearly the most metallic, and displays a Drude-like conductivity, an important result that was noted in earlier studies. The Drude model for the dielectric function describes the properties of a simple metal quite well, $\varepsilon(\omega) = \varepsilon_{sc} - \omega_p^2/[(\omega(\omega+i\Gamma))]$, where $\omega_p$ is the classical plasma frequency for YBa$_2$Cu$_3$O$_{6+\delta}$.
quency, \( \Gamma = 1/\tau \) is the scattering rate, and \( \epsilon_s \) is a high-frequency contribution. The Drude conductivity is \( \sigma_s(\omega) = \sigma_{dc}(1 + \omega^2 \tau^2) \), which has the form of a Lorentzian centered at zero frequency with a width at half maximum of \( 1/\tau \). For \( T \approx T_c \), the optical conductivity of the overdoped system shown in Fig. 1 does have a Drude-like frequency response, \( \sigma_s(\omega) \approx \omega \Gamma \). In addition \( \sigma_{dc} = \sigma_s(\omega \rightarrow 0) \approx 450 \Omega^{-1} \text{cm}^{-1} \) for \( T \approx T_c \), is well above the Mott minimum value for metallic conductivity of \( \approx 100 \Omega^{-1} \text{cm}^{-1} \) in these materials.

Previous investigations of overdoped systems resulted in large values for both the plasma frequency \( \omega_{p,s} \approx 4000 \text{cm}^{-1} \) and the scattering rate \( 1/\tau \), \( \approx 1000 \text{cm}^{-1} \).\(^{12,17} \) A principal objection to coherent transport along the \( c \) axis has been the large values for \( 1/\tau \), which lead to mean free paths that are substantially less than a lattice spacing.\(^1 \) If the \( c \)-axis conductivity is modeled using a two-component approach (Drude component to model free carriers, plus Lorentzian oscillators to represent bound excitations), then it is possible to estimate the \( c \)-axis plasma frequency and scattering rate. This approach yields values of \( \omega_{p,c} \approx 3200 \text{cm}^{-1} \) and \( 1/\tau_c \approx 380 \text{cm}^{-1} \) at 100 K, smaller than previously observed values.\(^{27} \) These results may be compared to the values in the copper-oxygen planes along the \( a \) axis for the overdoped material\(^{28} \) of \( \omega_{p,a} \approx 10,000 \text{cm}^{-1} \) and \( 1/\tau_a \approx 120 \text{cm}^{-1} \). The resistivity anisotropy is then expected to be \( \rho_l/\rho_a = (\omega_{p,a}^2 \tau_a)/(\omega_{p,c}^2 \tau_c) \approx 30 \), which is in good agreement with bandstructure estimates.\(^{29} \) Furthermore, taking \( v_F = 7 \times 10^6 \text{cm/s} \) gives a mean free path along the \( c \) axis \( l_c = v_F \tau_a \approx 60 \text{Å} \), which is more than five times the size of the unit cell, suggesting the possibility of coherent (Bloch-Boltzmann) transport in the overdoped material for \( T \approx T_c \).\(^{17} \) The increasingly coherent transport along the \( c \) axis has also been discussed in relation to the strength of the inelastic scattering in the copper-oxygen planes.\(^{30} \)

### B. Superconducting state

In YBCO the onset of superconductivity is accompanied by the dramatic formation of a plasma edge in the reflectance along the \( c \) axis for all the oxygen dopings studied.\(^{13,14} \) In the copper-oxide superconductors, the order parameter is \( \Delta(T) \), the optical conductivity of the overdoped system shown in Fig. 1 does have a Drude-like frequency response, \( \sigma_s(\omega) \approx \omega \Gamma \). The optical conductivity along the \( c \) axis occurring from the Josephson effect.\(^{31} \) In such a case, the \( c \)-axis penetration depth \( \lambda_c \) is determined by the Josephson current density \( J_c \), and is \( \lambda_c^2 = \hbar c / 8 \pi^2 d \Delta(T) \), \( \approx 1/\Delta(T) \), where \( d \) is the separation between the planes.\(^{32,33} \) In the BCS theory, \( J_c \) is related to the energy gap \( \Delta(T) \) and the tunneling resistance per unit area in the normal state \( R_n \) by\(^{34} \)

\[
J_c = \frac{\pi \Delta(T)}{2e R_n} \tanh \left( \frac{\Delta(T)}{2k_B T} \right). \tag{1}
\]

Adopting the BCS isotropic \( s \)-wave gap weak-coupling value of \( \Delta(T \approx T_c) \approx 1.76 k_B T_c \) and assuming that \( R_n \approx \rho_{sc} d \), then \( J_c \approx T_c / R_n \) at low temperature, and \( 1/\lambda_c^2 \approx \sigma_{dc} \Delta(T_c) \), where \( \sigma_{dc} \) is the extrapolated dc conductivity along the \( c \) axis in the normal state \( (T \geq T_c) \).\(^{20,21} \) From \( 1/\lambda^2 = 2 \pi \omega_{p,c} \), the strength of the condensate is \( \rho_{sc} \approx \omega_{p,c}^2 \), yielding

\[
\rho_{sc} = 65 \sigma_{dc} T_c. \tag{2}
\]

where the right and left hand side of the expression have units of \( \text{cm}^2 \). We note that several other workers have arrived at a similar relationship based on different assumptions.\(^{35,36} \) If the clean limit is assumed (all the normal-state carriers collapse into the condensate), then for \( T \ll T_c \), the response of the dielectric function is purely real, \( \varepsilon(\omega) = \varepsilon_s(\omega) = \varepsilon_s - \omega^2 / \omega_s^2 \), so that the plasma frequency of the condensate is \( \omega_{p,c} = - \omega^2 \varepsilon_s / \omega_s^2 \) in the limit of \( \omega \rightarrow 0 \). The frequency dependence of \( \sqrt{-\omega^2 \varepsilon_s(\omega)} \) is shown in Fig. 3 for a variety of oxygen dopings for \( T \ll T_c \). The low-frequency extrapolations employed in the Kramers-Kronig analysis below \( 40 \text{cm}^{-1} \) is included as a guide to the eye. The strength of the plasma frequency is decreasing dramatically with decreasing oxygen doping (Table I).

**FIG. 3.** The doping-dependent behavior of \( \sqrt{-\omega^2 \varepsilon_s(\omega)} \) for \( T \ll T_c \). In the \( \omega \rightarrow 0 \) limit this quantity is the plasma frequency of the condensate along the \( c \) axis, \( \omega_{p,c} \). The low-frequency extrapolation employed in the Kramers-Kronig analysis below \( 40 \text{cm}^{-1} \) is included as a guide to the eye. The strength of the plasma frequency is decreasing dramatically with decreasing oxygen doping (Table I).

The optically determined values for the superfluid density \( \rho_{sc} \) versus \( \sigma_{dc} T_c \) for YBCO are shown in the log-log plot in Fig. 4. In addition, the \( c \)-axis results for YBa\(_2\)Cu\(_3\)O\(_{y}\),\(^{21} \) Tl\(_2\)Ba\(_2\)CuO\(_{y}\),\(^{22} \) HgBa\(_2\)CuO\(_{y}\),\(^{39} \) and La\(_{2−x}\)Sr\(_x\)CuO\(_y\) (Ref. 39) are also shown. All the points fall on a line approximated by the scaling relation \( \rho_{sc} \approx 35 \sigma_{dc} T_c \), which is close to the result from Josephson coupling.\(^{39} \) In the log-log representation of Fig. 4, the numerical constant in the scaling relation is the offset of the line. The line may be shifted by assuming different ratios between \( 2\Delta / k_B T_c \); the initial value of \( 35 \) was based on the weak-coupling value of \( 2\Delta / k_B T_c \).
the assumption of an isotropic gap is qualitatively correct, yielding a reasonable agreement between theory and experiment. The Josephson result might have been expected for the underdoped materials where the transport along the c axis was activated and considered incoherent. However, it is less obvious for the optimally and overdoped systems; the overdoped material in particular gave indications of anisotropic 3D normal-state transport, and as such some deviation from this behavior might have been expected. Thus, it would be tempting to assume that the observed scaling $\rho_{c,c} \propto \sigma_{dc} T_c$ along the c axis justifies the view that the coupling between the planes is always incoherent. However, it is important to note that both tunnel junctions [superconductor-insulator-superconductor (SIS)] in the case considered here, as well as nontunnel junctions [superconductor-normal-superconductor (SNS)] can show the Josephson effect with a nearly identical Josephson current. 47–49 As a result, the qualitative agreement with the Josephson coupling model does not conflict with the normal-state results, which indicate that the transport is becoming more coherent at high oxygen dopings.

IV. CONCLUSIONS

The optical and transport properties in the normal state of YBCO suggest that the material shows signs of anisotropic 3D metallic transport at high oxygen dopings for $T \approx T_c$. A scaling relation $\rho_{c,c} \propto \sigma_{dc} T_c$ is observed along the c axis, in agreement with the result expected from Josephson coupling in the BCS weak limit case. We note that the Josephson coupling model does not require incoherent transport, suggesting that the transport may indeed tend towards more coherent behavior in YBCO at higher oxygen dopings.

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In the Drude-Lorentz or two-component fit, in addition to the effective plasma frequency \(\omega_p\), \(\gamma_i\), and \(\omega_{p,i}\) have also been used to fit the \(B_{1g}\) infrared active lattice modes (Ref. 14) as well as a number of other oscillators in the mid and near infrared: \(\omega_1 = 800, \gamma_1 = 2580, \omega_{p,1} = 5422, \omega_2 = 2400, \gamma_2 = 1540, \omega_{p,2} = 1840, \omega_3 = 1.2 \times 10^4, \gamma_3 = 1.4 \times 10^4, \omega_{p,3} = 1.9 \times 10^4\). All units are in cm\(^{-1}\).

T. Rõõm (private communication).


S. Chakravarty, H.-Y. Kee, and E. Abrahams, Phys. Rev. Lett. 82, 2366 (1999). The superfluid density was calculated to be \(\rho_{sc} = 4\pi \sigma_0(T)T^{\frac{3}{2}}[\Omega(p+1)][u - u_s]/p\). The expression in the curly brackets, described in detail in the reference, is assumed to be a universal constant. For \(T = T_c\), then \(\rho_{sc} \approx \sigma_0 T_c\), which is similar to the Josephson result.


