Determination of the optical properties of La\(_{2-x}\)Ba\(_x\)CuO\(_4\) for several dopings, including the anomalous x=1/8 phase


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Determination of the optical properties of La$_{2-x}$Ba$_x$CuO$_4$ for several dopings, including the anomalous $x = 1/8$ phase


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The optical properties of single crystals of the high-temperature superconductor La$_{2-x}$Ba$_x$CuO$_4$ have been measured over a wide frequency and temperature range for light polarized in the $a$-$b$ planes and along the $c$ axis. Three different Ba concentrations have been examined, $x = 0.095$ with a critical temperature $T_c = 32$ K, $x = 0.125$ where the superconductivity is dramatically weakened with $T_c \approx 2.4$ K, and $x = 0.145$ with $T_c \approx 24$ K. The in-plane behavior of the optical conductivity for these materials at high temperature is described by a Drude-like response with a scattering rate that decreases with temperature. Below $T_c$ in the $x = 0.095$ and 0.145 materials there is a clear signature of the formation of a superconducting state in the optical properties allowing the superfluid density ($\rho_s$) and the penetration depth to be determined. In the anomalous 1/8 phase, some spectral weight shifts from lower to higher frequency ($\gtrsim 300$ cm$^{-1}$) on cooling below the spin-ordering temperature $T_{so} \approx 42$ K, associated with the onset of spin-stripe order; we discuss alternative interpretations in terms of a conventional density-wave gap versus the response to pair-density-wave superconductivity. The two dopings for which a superconducting response is observed both fall on the universal scaling line $\rho_s/\sigma_{dc} \approx 4.4\Delta_{c0}/T_c$, which is consistent with the observation of strong dissipation within the $a$-$b$ planes. The optical properties for light polarized along the $c$ axis reveal an insulating character dominated by lattice vibrations, superimposed on a weak electronic background. No Josephson plasma edge is observed in the low-frequency reflectance along the $c$ axis for $x = 1/8$; however, sharp plasma edges are observed for $x = 0.095$ and 0.145 below $T_c$.

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I. INTRODUCTION

The discovery of superconductivity at elevated temperatures in the copper-oxide materials a quarter of a century ago sparked an intense effort to understand the mechanism of the superconductivity in these compounds. Despite the wealth of information from the accumulation of hundreds of thousands of scientific papers, there is still no consensus on the pairing mechanism. While superconductivity was originally observed in the single-layer La$_{2-x}$Ba$_x$CuO$_4$ system, single crystals of this material proved difficult to grow and attention quickly shifted to the related La$_{2-x}$Sr$_x$CuO$_4$ materials where large single crystals were available. Further investigations into systems with two or three copper-oxygen layers in the unit cell led to rapid increases in the critical temperature ($T_c$), with the current maximum $T_c \approx 135$ K observed at ambient pressure in the HgBa$_2$Ca$_2$Cu$_3$O$_{8+\delta}$ system. The physical properties of this class of correlated electron materials and the nature of the superconductivity have been described in numerous review articles. While most high-temperature superconductors display a doping-dependent dome-shaped superconducting phase boundary, an anomalous weakening of the superconductivity is observed at a doping of $p = 1/8$ per copper atom in the related La$_{2-x}$Sr$_x$CuO$_4$ system, and more recently in the Bi$_2$Sr$_2$Cu$_2$O$_{8+\delta}$ and YBa$_2$Cu$_3$O$_{7-\delta}$ materials. This so-called “1/8 anomaly” increases dramatically in La$_{2-x}$Ba$_x$CuO$_4$ for $x = 1/8$ and results in the almost total destruction of the bulk three-dimensional (3D) superconductivity. There is also a structural transition over much of the phase diagram from a low-temperature orthomorphic (LTO) to a low-temperature tetragonal (LTT) symmetry that is not observed in pure La$_{2-x}$Sr$_x$CuO$_4$ crystals; the LTO→LTT transition is also accompanied by the formation of an antiferromagnetically (AFM) ordered state.

Recently, large single crystals of La$_{2-x}$Ba$_x$CuO$_4$ have become available that have allowed a much more detailed investigation of the electronic and structural properties of the 1/8 phase. With the onset of the LTT phase both long-range charge-stripe order, followed by spin-stripe order at a slightly lower temperature, are observed. The question of what role stripes play in the formation of a superconducting state in the cuprate materials is the subject of some debate, however, in this case it appears that the formation of static charge-stripe order has the effect of electronically decoupling the copper-oxygen planes, significantly increasing the resistance along the $c$ axis, and frustrating the formation of a bulk superconducting ground state. The 1/8 phase exhibits a pseudogap in the LTO phase, similar to what is observed in the underdoped cuprates. The remaining Fermi arc (or pocket) in the nodal region becomes gapped in the LTT phase close to the spin-ordering temperature. The gap in the nodal region is momentum-dependent with a $d$-wave form, $\Delta(\phi) = \Delta_0 \cos 2\phi$, where $\Delta_0$ is the gap maximum and $\phi$ is a Fermi surface angle, similar to the form of the superconducting gap observed in other cuprate superconductors. It has been proposed that this is a superconducting gap, but that the
pairs lack the necessary phase coherence required to form a condensate.\textsuperscript{28,29}

The optical properties of a material can provide important information about the nature of the lattice vibrations and the electronic transport, including the formation of charge and superconducting energy gaps. The role of charge-stripe and spin-stripe ordering in the 1/8 phase is of particular interest. Previous optical investigations of stripe correlations in an analogous phase of La\textsubscript{1−δ}Nd\textsubscript{δ}Sr\textsubscript{0.12}CuO\textsubscript{4},\textsuperscript{61–63} and in La\textsubscript{2−δ}Sr\textsubscript{δ}CuO\textsubscript{4}\textsuperscript{63–65} have resulted in some disagreement over the association of certain infrared spectral features with charge order.\textsuperscript{66,67}

In this work we determine the temperature dependence of the optical properties of single crystals of La\textsubscript{2−δ}Ba\textsubscript{δ}CuO\textsubscript{4} over a wide frequency range for three different dopings, \(x = 0.095, 0.125\) and 0.145 with \(T_c\)'s of approximately 32, 2.4 and 24 K, respectively, for light polarized in the conducting \(a\)-\(b\) planes, and along the poorly conducting \(c\) axis. The optical conductivity in the \(a\)-\(b\) planes for all three materials show a Drude-like metallic behavior for the free-carriers at room temperature with a scattering rate that decreases with temperature; for the superconducting \(x = 0.095\) and 0.145 samples there is a characteristic suppression of the low-frequency conductivity below \(T_c\), allowing the superfluid density \(\rho_s\) to be determined from this so-called missing spectral weight.\textsuperscript{68}

In contrast, in a refinement of earlier optical work,\textsuperscript{26,69} the 1/8 phase shows a dramatic narrowing of the free-carrier response below \(T_{so}\) accompanied by the transfer of some spectral weight to both high and low frequencies. This transfer of spectral weight is consistent with the formation of a momentum-dependent gap due to the formation of charge-stripe and spin-stripe order resulting in a nodal metal. The loss of low-frequency spectral weight also response mimics the missing spectral weight observed in the superconducting materials, and it is possible that the transfer of spectral weight is associated with the two-dimensional (2D) fluctuating superconducting state prior to the onset of bulk 3D superconductivity.\textsuperscript{29}

The two dopings for which superconductivity is observed fall on the universal scaling line, \(\rho_{so}/8 \simeq 4.4 \sigma_{dc} T_c\) (where \(\sigma_{dc}\) is measured just above \(T_c\)).\textsuperscript{70–73} which is consistent with the observation that there is substantial dissipation in the normal state of these materials.

The optical properties along the poorly-conducting \(c\) axis are dominated by the infrared-active lattice modes superimposed on a weak electronic background. In the superconducting \(x = 0.095\) and 0.145 samples, sharp plasma edges in the low-frequency reflectance develop below \(T_c\) due to Josephson coupling between the planes. No Josephson plasma edge, or any indication of metallic behavior, is observed for the 1/8 phase down to the lowest measured temperature, despite the dramatic reduction of the \(c\)-axis resistivity below \(T_{so}\).

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure1}
\caption{(a) Temperature vs hole doping phase diagram of La\textsubscript{2−δ}Ba\textsubscript{δ}CuO\textsubscript{4} single crystals (reproduced from Ref. 46). Onset temperatures: \(T_c\) for bulk superconductivity (SC), \(T_{co}\) for charge stripe order (CO), \(T_{so}\) for spin stripe order (SO), and \(T_{LT}\) for the low-temperature structural phases LTT and LTLO. The unit cell of La\textsubscript{2−δ}Ba\textsubscript{δ}CuO\textsubscript{4} is shown in the (b) high-temperature tetragonal (HTT), (c) low-temperature orthorhombic (LTO), and (d) low-temperature tetragonal (LTT) phases.
}
\end{figure}

\section{II. EXPERIMENT}

Single crystals of La\textsubscript{2−δ}Ba\textsubscript{δ}CuO\textsubscript{4} were grown using the traveling-solvent floating-zone method for a series of three nominal Ba concentrations, \(x_0 = 0.095, 0.125\) and 0.155. The \(x_0 = 0.095\) and 0.155 compositions are superconducting with transitions at \(T_c \simeq 32\) and 24 K; the superconductivity in the \(x = 0.125\) sample is strongly suppressed with \(T_c \lesssim 2.4\) K (all values for \(T_c\) were determined by magnetic susceptibility). The \(T_c\) for the nominal \(x_0 = 0.155\) crystal studied here (and in Ref. 69) is substantially lower than that of the piece (from the same growth) characterized in Ref. 46. Interpreting the discrepancy in \(T_c\) as a difference in Ba concentration, we estimate that the present sample corresponds to \(x = 0.145 \pm 0.01\), and we refer to it as \(x = 0.145\) throughout the rest of the paper.

The phase diagram of La\textsubscript{2−δ}Ba\textsubscript{δ}CuO\textsubscript{4} shown in Fig. 1(a) is considerably more sophisticated than that
spin order is likely very weak and is not observed; at low
charge order does not develop until \( T \rightarrow 0 \), phase at very low temperature. In the
\( x \) to a “low-temperature less orthorhombic” (LTLO)
dergoes a weakening of the orthorhombic strain, leading
of the charge-stripe and spin-stripe order.
\( \text{Fig. 1(d)} \); this phase is associated with the development
in a low-temperature tetragonal (LTT) phase
is accompanied by a weak in-plane distortion resulting
\( 46 \) resistivity for the
the LTO structure un-
ting the CuO
(Refs. 28 and 29). Weak
at \( T \rho \), below which it falls dramatically before
once again adopting a linear temperature dependence. Along
the \( c \) axis, \( \rho_c \) continues to increase below \( T_{co} \) until just below
\( T_{so} \) where it begins to decrease quickly until \( \approx 25 \) K, below
which it decreases less rapidly. Note different scales for \( \rho_{ab} \).

of the related \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \) material.\(^{46}\) Above room
temperature, this material is in the \( I4/mmm \) tetragonal
(HTT) phase [Fig. 1(b)]. With decreasing temperature
it undergoes an orthorhombic distortion into a \( \text{Bmnab} \)
(LTO) phase [Fig. 1(c)] which may be described as a tilting
of the \( \text{CuO}_6 \) octahedra towards the La ions leading
to a roughly \( \sqrt{2} \times \sqrt{2} \) larger basal plane rotated by \( 45^\circ \).
The HTT→LTO transition occurs at \( T_{LTO} \approx 305, 235 \) and
190 K for the \( x = 0.095, 0.125 \) and 0.145 materials,
respectively.\(^{46}\) At dopings close to \( x = 0.125 \) the
tilt of the octahedra moves between the La ions and is
accompanied by a weak in-plane distortion resulting in
a low-temperature tetragonal (LTT) phase \( P4_2/ncm \)
[Fig. 1(d)]; this phase is associated with the development
of charge-stripe and spin-stripe order.\(^{74}\) In the \( x = 0.095 \)
crystal close to \( T_{co} \approx T_{so} \approx T_c \) the LTO structure
undergoes a weakening of the orthorhombic strain, leading
to a “low-temperature less orthorhombic” (LTLO) \( Pccn \)
phase at very low temperature. In the \( x = 0.145 \) material
the LTO→LTT transition occurs at \( \approx 62 \) K, but
charge order does not develop until \( T_{co} \approx 35 \) K and the
spin order is likely very weak and is not observed; at low
temperature the structure is thought to be a mixture of
LTT+LTLO.

The \( ab \)-plane and \( c \)-axis resistivity of the \( x = 0.095 \)
material is shown close to \( T_c \approx 32 \) K in Fig. 2(a) (Ref. 34).
In the normal state \( \rho_{ab} \) displays a characteristic linear
decrease with temperature, until just above \( T_c \) where a
weak anomaly is observed that is associated with the on-
set of the structural transition previously described.\(^{46,75}\)
In contrast, in the normal state \( \rho_c \) increases with
decreasing temperature, leading to a resistivity anisotropy
\( \rho_c/\rho_{ab} \) that continues to increase until just above \( T_c \).
The \( ab \)-plane and \( c \) axis resistivity of the \( x = 0.125 \) material
is shown in Fig. 2(b) (Refs. 28 and 29). In the normal
state \( \rho_{ab} \) decreases with temperature in an approx-
imately linear fashion until a small jump is observed at
the charge-ordering temperature \( T_{co} \approx 54 \) K, followed
by a dramatic decrease at the spin-ordering temperature
\( T_{so} \approx 42 \) K; this latter transition is extremely sensitive
to the applied magnetic field.\(^{28}\) Below \( T_{so} \) a 2D fluctu-
ating superconducting state is thought to form and the
the resistivity continues to decrease exponentially; at the
same time a strong diamagnetic response is observed.\(^{29}\)

This material is conjectured to undergo a Berezinskii-
Kosterlitz-Thouless transition\(^{76,77}\) at \( T_{BKT} \approx 16 \) K,
below this temperature the resistivity that is effectively
zero.\(^{28}\) A true bulk (3D) superconducting transition is
finally achieved at \( T_c \approx 2.4 \) K. In the normal state \( \rho_c \)
shows little change with decreasing temperature until \( T_{co} \)
where, similar to \( \rho_{ab} \), a small jump is observed; however,
\( \rho_c \) continues to increase indicating a dramatic increase in
the resistivity anisotropy until slightly below \( T_{so} \), where
\( \rho_c \) begins to decrease quickly, reaching a negligible value
below \( \approx 6 \) K.\(^{29}\)

It is possible to cleave some of these materials, result-
ing in mirror-like \( ab \)-plane surfaces. However, in most
cases cleaving yields a “terraced” surface consisting of

FIG. 2. (a) The temperature dependence of the \( ab \)-plane
(solid line) and \( c \)-axis (dashed line) resistivity for \( x = 0.095 \)
(Ref. 34). In the normal state \( \rho_{ab} \) decreases linearly with
decreasing temperature, while \( \rho_c \) is observed to increase until
just above \( T_c \), below which both decrease dramatically and a
weak anomaly is observed in \( \rho_{ab} \). The \( ab \)-plane and \( c \)-axis
resistivity for the \( x = 0.125 \) material (Refs. 28 and 29).

Weak anomalies are observed in \( \rho_{ab} \) and \( \rho_c \) at \( T_{co} \); \( \rho_{ab} \) continues
to decrease until \( T_{so} \), below which it falls dramatically before
once again adopting a linear temperature dependence. Along
the \( c \) axis, \( \rho_c \) continues to increase below \( T_{co} \) until just below
\( T_{so} \) where it begins to decrease quickly until \( \approx 25 \) K, below
which it decreases less rapidly. Note different scales for \( \rho_{ab} \).

FIG. 3. A photomicrograph of the cleaved \( ab \)-plane surface
of \( \text{La}_{1.875}\text{Ba}_{0.125}\text{CuO}_4 \) revealing a number of striations and
cleavage steps; the total area shown is roughly \( 2 \) mm \( \times \) \( 2 \) mm.
merous steps with exposed c-axis faces, shown in Fig. 3. In Fig. 4(a) the absolute reflectance of a freshly-cleaved ab-plane surface for the $x = 0.095$ material has been measured at room temperature at a near-normal angle of incidence over the infrared region using an in situ evaporation technique. In addition to the Hagen-Rubens $R \propto 1 - \sqrt{\omega}$ behavior expected for a metallic system and the two normally infrared-active vibrations at about 120 and 360 cm$^{-1}$, there are two strong antiresonances at $\simeq 460$ and 575 cm$^{-1}$ associated with the longitudinal optic modes of the infrared-active c-axis modes (Sec. IIIB). These features are absent from naturally-grown crystal faces, but are observed whenever a surface has been cut and polished, or as illustrated in the photomicrograph shown in Fig. 3, when a cleaved surface has numerous steps. This phenomenon has been documented extensively in the literature and is attributed to the misorientation of the ab-plane surface.

Clearly, it is desirable to minimize these extrinsic features while preserving the intrinsic behavior of the sample. Pursuing this goal, the opposite face was polished using a series of successively finer diamond grits using kerosene as a suspension agent, with a final lap using a 0.1 $\mu$m diamond paste. The infrared reflectance shown in Fig. 4(b) is quite similar to the reflectance of the cleaved surface, and shows that the antiresonance features are still present. It was subsequently determined from x-ray Laue diffraction that this surface was misoriented by $\simeq 4^\circ$. The sample was reoriented using the Laue camera to better than 1$^\circ$ and then repolished. The resulting reflectance in Fig. 4(c) indicates that the antiresonance features have been minimized. Thus, while cleaved surfaces are often desirable, in those cases where strong antiresonances are observed, determining the correct orientation and careful polishing may remove most of these unwanted spectral features.

The reflectance of single crystal La$_{2-x}$Ba$_x$CuO$_4$ for $x = 0.095$, 0.125 and 0.145 has been measured at a near-normal angle of incidence for light polarized in the ab planes, as well as along the c axis, over a wide frequency range from $\simeq 2$ meV to over 3 eV ($\simeq 20$ to over 24,000 cm$^{-1}$) at a variety of temperatures using the previously described overcoating technique. The ab-plane reflectance of the $x = 0.125$ sample was measured from a cleaved surface, while for the other dopings the surface was oriented to better than 1$^\circ$ and polished. The c-axis was revealed by cutting the ab face at 90$^\circ$, which was subsequently polished. The c-axis direction was determined by using an infrared polarizer to check the modulation of the infrared signal, with the minima corresponding to the c-axis direction.

The reflectance is a complex quantity consisting of an amplitude and a phase, $\tilde{R} = \sqrt{R} \exp (i\theta)$. Because only the amplitude $R = \tilde{R} \tilde{R}^*$ is measured in this experiment it is often difficult to decipher how the reflectance is related to the real and imaginary optical properties. Thus, the complex optical properties have been calculated from a Kramers-Kronig analysis of the reflectance.$^{86}$ The Kramers-Kronig transform requires that the reflectance be determined for all frequencies, thus extrapolations must be supplied in the $\omega \to 0$, $\infty$ limits. At low frequency, for light polarized in the ab planes, above $T_c$ in the normal state the metallic Hagen-Rubens form for the reflectance $R(\omega) \propto 1 - \sqrt{\omega}$ is employed. Below $T_c$ in the superconducting state $R(\omega) \propto 1 - \omega^2$; however, it should be noted that when the reflectance is close to unity the analysis is not sensitive upon the choice of low-frequency extrapolation. For light polarized along the c axis, at low frequency the Hagen-Rubens form is used in the normal state, while below $T_c$ we again use $R(\omega) \propto 1 - \omega^2$. For all polarizations the reflectance is assumed to be constant above the highest measured frequency point up to $\simeq 1 \times 10^5$ cm$^{-1}$, above which a free electron gas asymptotic reflectance extrapolation $R(\omega) \propto 1/\omega^4$ is employed.$^{87}$

FIG. 4. The reflectance of La$_{1.005}$Ba$_{0.095}$CuO$_4$ at 295 K in the far-infrared region of for light polarized in the a-b planes. (a) The reflectance from a cleaved surface. There are two prominent antiresonances at $\simeq 460$ and 570 cm$^{-1}$ which are associated with c-axis longitudinal optic modes that become infrared-active due to a surface misorientation. This effect can be seen in the c-axis loss function (dashed line). (b) The reflectance from the polished surface of the same sample with a measured surface misorientation of $4^\circ$; the antiresonances are still prominent. (c) The reflectance from the polished surface with a measured surface misorientation of $\lesssim 1^\circ$; the antiresonances are substantially weaker.
The temperature dependence of the reflectance in the far-infrared region.

III. RESULTS AND DISCUSSION

A. a-b plane

The temperature dependence of the reflectance of La$_{2-x}$Ba$_x$CuO$_4$ for light polarized in the a-b planes is shown over a wide frequency range for $x = 0.095$, 0.125 and 0.145 in Figs. 5(a), (b) and (c), respectively; the insets show the temperature dependence of the reflectance in the far-infrared region.

At room temperature the reflectance curves for all three dopings are similar in that the reflectance at low frequency is high and decreases with frequency until a plasma edge in the reflectance is encountered at about 1 eV. The reflectance in the infrared region is observed to increase with electronic doping. In the three samples the temperature dependence of the reflectance all show a similar increase in the low-frequency reflectance with decreasing temperature. In the superconducting state the $x = 0.095$ material displays an abrupt increase in the low-frequency reflectance, while the changes in the $x = 0.145$ material below $T_c$ are not as pronounced. At low temperature the reflectance of the $x = 0.125$ sample is dramatically different; below $\approx 200$ cm$^{-1}$ the reflectance increases down to the lowest measured temperature, but close to or below $T_{so}$, the reflectance in the 200 – 2000 cm$^{-1}$ region actually decreases with temperature, as shown in the inset of Fig. 5(b). While the reflectance contains a great deal of information about the electronic transport and structure in a material, the complicated behavior observed in the $x = 0.125$ material makes it difficult to interpret. The determination of the optical conductivity from a Kramers-Kronig analysis of the reflectance is more revealing.

1. $x = 0.095$

The real part of the optical conductivity for light polarized in the a-b planes of La$_{1.905}$Ba$_{0.095}$CuO$_4$ is shown in the infrared region at temperatures above $T_c$ in Fig. 6(a). At room temperature the conductivity appears rather broad in the far-infrared region, but as the inset in this panel shows it is in fact decreasing rather rapidly before giving way above $\approx 800$ cm$^{-1}$ to a broad plateau in the mid-infrared; there is a shoulder at about 4000 cm$^{-1}$ (0.5 eV) above which the conductivity continues to decrease until about 1 eV, at which point it begins to slowly rise again. Superimposed on the room-temperature conductivity curve are several sharp resonances at $\approx 126$, 358 and 682 cm$^{-1}$; these are the normally-active infrared vibrations which are observable due to the poor screening in this class of materials. The weak antiresonances observed in the reflectance in Fig. 4(c) at $\approx 460$ and 570 cm$^{-1}$ attributed to a surface misorientation manifest themselves as weak resonances or antiresonances in the conductivity. In the HTT phase, the irreducible vibrational representation for the infrared active modes is

$$\Gamma_{IR}^{HTT} = 4A_{2u} + 4E_u$$

where the singly-degenerate $A_{2u}$ modes are active along the c axis and the doubly-degenerate $E_u$ modes are active in the a-b planes. Below the structural phase transition in the LTO phase the unit cell is larger and more complicated. The lowered symmetry results in the removal of degeneracy and more infrared modes

$$\Gamma_{IR}^{LTO} = 4A_u + 6B_{1u} + 7B_{2u} + 4B_{3u}$$

where the $A_u$ modes are silent, and the singly-degenerate $B_{1u}$, $B_{2u}$ and $B_{3u}$ modes are active along the $c$, $b$ and $a$ axes, respectively. The additional low-temperature structural transition to either a LTLO or LTT+LTLO phase represents a further reduction of symmetry; for the LTT phase the irreducible vibrational representation becomes

$$\Gamma_{IR}^{LTT} = 3A_{1u} + 7A_{2u} + 8B_{1u} + 3B_{2u} + 12E_u$$
La$_{1.905}$Ba$_{0.095}$Cu$_{4}$O$_{4}$

FIG. 6. The real part of the optical conductivity of La$_{1.905}$Ba$_{0.095}$Cu$_{4}$O$_{4}$ for light polarized in the a-b planes. (a) The infrared region for several temperatures above $T_c$ = 32 K. The antiresonances observed in the reflectance in Fig. 4(c) just above and well below (long-dashed line) to reproduce the data (solid line) quite well. Inset: The Drude-Lorentz fit to the data at 295 K. The Drude component (dashed line) and Lorentz oscillators (dash-dot lines) combine (long-dashed line) to reproduce the data (solid line) quite well. (b) The optical conductivity just above and well below $T_c$ in the infrared region illustrating the transfer of spectral weight into the condensate. Inset: The Drude-Lorentz fit to the data at 35 K.

where the $A_{1u}$, $B_{1u}$ and $B_{2u}$ modes are silent; as in the case of the HTT phase the $A_{2g}$ and $E_u$ modes are active along the c axis and the a-b planes, respectively.

At room temperature La$_{1.905}$Ba$_{0.095}$Cu$_{4}$O$_{4}$ appears to be in the HTT phase (demonstrated by the c-axis properties in Sec. IIB); of four possible in-plane modes, only three are clearly identified (the remaining mode is likely too weak to be observed). At low temperature the vibrations typically narrow and increase in frequency; however, the low-frequency mode at $\sim 126$ cm$^{-1}$ in Fig. 6(a) appears to broaden anomalously at low temperature, suggesting it may be sensitive to the charge or spin order.

As the temperature decreases the low-frequency spectral weight associated with the free-carrier component increases rapidly. The spectral weight is defined here simply as the area under the conductivity curve over a given frequency interval:

$$N(\omega, T) = \int_0^\infty \sigma_1(\omega, T) d\omega.$$ 

For a metal, in the absence of other excitations, the area under the entire conductivity curve is the well-known sum rule,

$$\int_0^\infty \sigma_1(\omega) d\omega = \Omega/(2e^2),$$

where $\Omega$ is the classical plasma frequency. The value for the dc resistivity in the HTT phase (demonstrated by the long-dashed line) to reproduce the observed mid-infrared antiresonances.

The parameters for the Drude plasma frequency and scattering rate 1/$\tau_D$ determined in the normal state and effective penetration depth are determined for $T \ll T_c$.

<table>
<thead>
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<th>$x$</th>
<th>$T_c$ (K)</th>
<th>$\omega_p,D$ (cm$^{-1}$)</th>
<th>$1/\tau_D$ (cm$^{-1}$)</th>
<th>$\omega_p, S$ (cm$^{-1}$)</th>
<th>$\lambda_0$ (Å)</th>
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</thead>
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<td>32</td>
<td>4360</td>
<td>131</td>
<td>3550</td>
<td>4480</td>
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<tr>
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<td>$-$</td>
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<td>0.145</td>
<td>24</td>
<td>7360</td>
<td>261</td>
<td>$\sim$ 2970</td>
<td>$\sim$ 5360</td>
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</tbody>
</table>

$^a$ Fits at 100 K; $x = 0.125$ interpolated from 60 and 150 K data.
broad spectral range in the inset of Fig. 6(a). The fit to the data at in the normal state at 35 K is quite good and is shown in the inset in Fig. 6(b). The Drude plasma frequency $\omega_{p,D} \simeq 4360 \pm 300 \text{ cm}^{-1}$ does not vary with temperature, while the scattering rate decreases dramatically with temperature, $1/\tau_D \simeq 295$, 207, 131 and 75 cm$^{-1}$ at 295, 200, 100 and 35 K, respectively (Table I).

For a superconductor with an isotropic energy gap $\Delta$, the gap in the optical conductivity is $2\Delta$. In the superconducting state below $T_c$, shown in Fig. 6(b) for $x = 0.095$, the dramatic decrease in the low-frequency optical conductivity below $\sim 200 \text{ cm}^{-1}$ (25 meV) is associated with the formation of a superconducting energy gap. This result is in reasonable agreement with the ARPES estimate of $2\Delta_0 \simeq 20 \text{ meV}$ for the maximum gap value in this material. The “missing area” is referred to as the spectral weight of the condensate $N_c$ and may be calculated from the Ferrell-Glover-Tinkham sum rule.

$$N_c = N(\omega_c, T \simeq T_c) - N(\omega_c, T \ll T_c) = \omega_{p,S}^2 / 8. \quad (3)$$

Here $\omega_{p,S}^2 = 4\pi n_e e^2 / m^*$ is the square of the superconducting plasma frequency, and the superfluid density is simply $\rho_{sf} \equiv \omega_{p,S}^2$; the cut-off frequency $\omega_c$ is chosen so that the integral converges smoothly. The superconducting plasma frequency has also been determined from the real part of the dielectric function in the low frequency limit where $\epsilon_1(\omega) = \epsilon_\infty - \omega_{p,S}^2 / \omega^2$. Yet another method of extracting $\omega_{p,S}$ from $\epsilon_1(\omega)$ is to determine $[-\omega^2 \epsilon_1(\omega)]^{1/2}$ well below $T_c$ in the $\omega \rightarrow 0$ limit. All three techniques yield $\omega_{p,S} \simeq 3550 \pm 200 \text{ cm}^{-1}$, which corresponds to an effective penetration depth of $\lambda_0 \simeq 4480 \pm 270 \text{ Å}$. The fact that less than two-thirds of the free-carriers in the normal state have condensed ($\omega_{p,S}^2 / \omega_{p,D}^2 \lesssim 0.6$) is consistent with the observation of strong dissipation in the normal state.

2. $x = 0.145$

The temperature dependence of the real part of the optical conductivity for the $x = 0.145$ material is shown in Fig. 7 at a variety of temperatures above and below $T_c$. In the normal state the overall behavior is not unlike that of the $x = 0.095$ material; at low frequency a Drude-like component narrows rapidly with decreasing temperature but gives way to a broad, incoherent component above $\sim 60 \text{ meV}$. The conductivity in the normal state can be fit reasonably well using the Drude-Lorentz model yielding $\omega_{p,D} \simeq 7360 \text{ cm}^{-1}$ and $1/\tau_D \simeq 500 \text{ cm}^{-1}$ at room temperature. The larger value for $\omega_{p,D}$ denotes the increase in the density of free carriers. It is interesting to note that the normal-state scattering rate in the overdoped material is roughly twice that of the underdoped system; this may be due to electronic correlations or it may be that the increasing cation disorder in the blocking layer results in a larger value of $1/\tau_D$ (Table I). Upon entry into the superconducting state a decrease in the low-frequency conductivity is once again observed; however, the onset at which this decrease occurs is rather indistinct and there is considerably more residual conductivity at low frequency than was observed in the underdoped material. Using the techniques previously discussed, the superconducting plasma frequency is estimated to be $\omega_{p,S} \simeq 2970 \pm 180 \text{ cm}^{-1}$, corresponding to a penetration depth of $\lambda_0 \simeq 5360 \pm 350 \text{ Å}$. As a result of the large normal state scattering rate, less than a fifth of the free-carriers have condensed ($\omega_{p,S}^2 / \omega_{p,D}^2 \lesssim 0.2$).

3. $x = 0.125$

The in-plane reflectance of La$_{2-x}$Ba$_x$CuO$_4$ for $x = 0.125$ is shown in Fig. 5(b). While some aspects of the reflectance have been previously discussed, there are several important points that bear repeating. As the temperature is lowered, the reflectance is observed to increase in the far- and mid-infrared regions. Close to or below the spin-ordering temperature $T_{co} = 42 \text{ K}$ the reflectance decreases in the $200 - 2000 \text{ cm}^{-1}$ interval; however, below about $200 \text{ cm}^{-1}$ the reflectance continues to increase. The consequences of this behavior may be seen more clearly in the conductivity shown for temperatures above $T_{co}$ in Fig. 8(a) and mainly below $T_{co}$ in Fig. 8(b). At high temperature the optical conductivity
FIG. 8. (a) The real part of the optical conductivity $\sigma(e^{-})$ for light polarized in the $a$-$b$ planes at several temperatures above $T_{co}$ and $T_{so}$. Inset: the infrared reflectance for the same temperatures. (b) The conductivity for several temperatures below $T_{co}$ and $T_{so}$ in the same region showing the transfer and loss of spectral weight occurring mainly below $T_{so}$. Inset: the infrared reflectance for the same temperatures contrasting the different responses at low and high frequency. The asterisks denote structure due to surface misorientation, while the arrows indicate the in-plane infrared-active lattice modes.

FIG. 9. The frequency dependence of the in-plane scattering rate $1/\tau(e^{-})$ for $\omega_p,D$ and $\epsilon_{c} = 4$ (although the choice of $\epsilon_{c}$ has little effect on the scattering rate or the effective mass in the far-infrared region). The infrared-active lattice modes observed in the conductivity have been fit using Lorentzian oscillators superimposed on a linear background and then removed from the complex dielectric function. The resulting temperature dependence of the in-plane scattering rate $1/\tau(e^{-})$ is shown in Fig. 9 for the three different dopings studied in this work. A simple Drude model would produce a frequency-independent scattering rate; however, despite the reasonable agreement with the Drude model, in Figs. 9(a), (b), and (c), $1/\tau(e^{-})$ displays an approximately linear frequency dependence. We note that in the $\omega = 0$ limit the two models should yield the same scattering rate, and indeed $1/\tau(e^{-})$ is the rapidly decreasing resistivity observed in Fig. 2(b).

4. Scattering rate

The Drude model is a reasonable description of a non-interacting Fermi liquid; however, the cuprates are either moderately or strongly correlated electron systems. The scattering rate and the effective mass are allowed to adopt a frequency dependence,

$$\frac{1}{\tau(e^{-})} = \frac{\omega_p^2}{4\pi} \text{Re} \left[ \frac{1}{\sigma(e^{-})} \right]$$

and

$$\frac{m^*(\omega)}{m_e} = \frac{\omega_p^2}{4\pi\omega} \text{Im} \left[ \frac{1}{\sigma(e^{-})} \right],$$

where $m_e$ is the bare mass, $m^*(\omega)/m_e = 1 + \lambda(\omega)$ and $\lambda(\omega)$ is a frequency-dependent electron-boson coupling constant. In this instance we set $\omega_p = \omega_{p,D}$ and $\epsilon_{c} = 4$ (although the choice of $\epsilon_{c}$ has little effect on the scattering rate or the effective mass in the far-infrared region).

The infrared-active lattice modes observed in the conductivity have been fit using Lorentzian oscillators superimposed on a linear background and then removed from the complex dielectric function. The resulting temperature dependence of the in-plane scattering rate $1/\tau(e^{-})$ is shown in Fig. 9 for the three different dopings studied in this work. A simple Drude model would produce a frequency-independent scattering rate; however, despite the reasonable agreement with the Drude model, in Figs. 9(a), (b), and (c), $1/\tau(e^{-})$ displays an approximately linear frequency dependence. We note that in the $\omega = 0$ limit the two models should yield the same scattering rate, and indeed $1/\tau(e^{-})$ is the rapidly decreasing resistivity observed in Fig. 2(b).

The temperature dependence $1/\tau_D$ (shown on the plots and for each doping) merits a brief discussion. For the three dopings examined, $x = 0.095, 0.125$, and $0.145$, $T_{co} = 54$ K, there is a characteristic narrowing of the Drude-like component and spectral weight is transferred from high to low frequency. The plasma frequency $\omega_{p,D} \simeq 6480 \pm 350$ cm$^{-1}$ is relatively constant, while the scattering rate decreases from $1/\tau_D = 439$ at room temperature to 81 cm$^{-1}$ at $T_{co}$ (Table 1).

In Fig. 8(b) for $T_{so} < T < T_{co}$ the scattering rate continues to decrease without any obvious loss of spectral weight or change in $\omega_{p,D}$. Below $T_{so}$ the Drude-like component in the conductivity continues to narrow; however, above $\sim 300$ cm$^{-1}$ there is now a noticeable increase in the conductivity, indicating a transfer of spectral weight from the low-frequency free-carrier component to localized or gapped excitations, the significance of which will be discussed in Sec. IIIIC. This is precisely the temperature at which the infrared reflectance, shown in the inset of Fig. 8(b), begins to decrease above $\sim 200$ cm$^{-1}$, while still increasing at low frequency. Below $\sim 30$ K little spectral weight is being transferred to high energy, instead the decrease in the low-frequency spectral weight is now associated almost entirely with the increasingly narrow free-carrier response, such as might be expected in a nodal metal. Attempts to fit the free-carrier component with a Drude-model yield a rapidly decreasing value for $\omega_{p,D}$ for $T < T_{so}$, but at the same time $1/\tau_D$ is also decreasing dramatically. We note that in the Drude model $\sigma_{dc} = \omega_{p,D}^2\tau_D/60$, so that the net effect is the rapidly decreasing resistivity observed in Fig. 2(b). At $\sim 6$ K the free-carrier response is now so narrow it may no longer be accurately observed, giving way to a broad, incoherent background conductivity. It is difficult to distinguish this behavior from the phenomenon of missing spectral weight in the superconducting state.
the values of the Drude scattering rate at 295 K increase steadily from $1/\tau_D = 295 \text{ cm}^{-1}$ to 439 cm$^{-1}$ and 500 cm$^{-1}$, respectively, indicating either stronger electronic correlations or that the increase in barium concentration is accompanied by increasing out-of-plane cation disorder, which may in turn lead to an increase in elastic scattering in the copper-oxygen planes. While $1/\tau_D$ decreases reasonably quickly with decreasing temperature for $x = 0.095$, this trend is not as pronounced for $x = 0.145$. As previously noted, in the 1/8 crystal $1/\tau_D$ decreases steadily with temperature, but for $T < T_{so}$ the reduction of the scattering rate is dramatic, an effect that is shown in more detail in Fig. 9(d).

The frequency-dependence of the scattering rate contains a wealth of information. The dashed line in Fig. 9 denotes $1/\tau(\omega) = \omega$: the region below this line is associated with a Landau-Fermi liquid regime where the quasiparticles are well defined. In the materials examined, at high temperature, $1/\tau(\omega) > \omega$ at low frequency, indicating that the conductivity in the a-b planes is strongly dissipative, and that the quasiparticles are not well defined. Across the far-infrared region $1/\tau(\omega)$ decreases with temperature and in the $x = 0.095$ material, just above $T_c$, at 35 K, and in the 1/8 phase at low temperature ($T < T_{so}$), $1/\tau(\omega) \approx \omega$, indicating well-defined quasiparticles and coherent transport; this condition is never satisfied in the $x = 0.145$ material. At low frequency, $1/\tau(\omega)$ decreases with decreasing temperature; however, in the 1/8 phase this trend is reversed for $T \lesssim T_{so}$ above about 150 cm$^{-1}$, shown in Fig. 9(d), where the scattering rate increases dramatically. This behavior is consistent with the formation of a momentum-dependent gap.

It is interesting to note that, aside from some artifacts due to the imperfect nature of the phonon subtraction, for the $x = 0.095$ material in Fig. 9(a) the frequency dependent scattering rate is essentially featureless except for a small kink at about 45 meV (also observed in the $x = 0.145$ material). In many high-temperature superconductors, $1/\tau(\omega)$ often has sharp inflection points in the normal state;\textsuperscript{101} when an inversion of the scattering rate is performed\textsuperscript{102,103} these structures correspond to peaks in the electron-boson spectral function.\textsuperscript{104-106} It has been proposed that the peak location is proportional to the position of the magnetic scattering resonance\textsuperscript{107,108} in polarized neutron scattering measurements $\Omega_r \approx 5.4 k_BT_r$, and more generally that it scales with the superconducting transition temperature, $\Omega_r \approx 6.3 k_BT_r$. In the underdoped cuprates the presence of a pseudogap on the Fermi surface is expected to produce kinks in the scattering rate at roughly $\Omega_r + \Delta_{pg}$ and $\Omega_r + 2\Delta_{pg}$.\textsuperscript{110} In the $x = 0.095$ ($T_c = 32$ K) material, taking the average, $\Omega_r \approx 16$ meV, while $\Delta_{pg} \approx 15$ meV (Ref. 56) yielding $\Omega_r \approx 31$ and 46 meV; the larger value is very close to where a kink is observed in $1/\tau(\omega)$ in Fig. 9(a). While it is tempting to associate this kink with a magnetic resonance, the absence of such a feature in the underdoped La$_{2-x}$Sr$_x$CuO$_4$ material\textsuperscript{111,112} suggests a different origin for this feature.

In the more heavily doped $x = 0.145$ ($T_c \approx 24$ K) material, $\Omega_r \approx 12$ meV and $\Delta_{pg} \approx 10$ meV suggesting features at $\approx 22$ and 32 meV; however, no obvious structure in the scattering rate is observed in Fig. 9(c) at either of these energies.

5. Parameter scaling

It has been noted that the high-temperature superconductors obey the scaling relation $\rho_s/8 \approx 4.4 \sigma_{dc} T_c$, where $\sigma_{dc} = \sigma_1(\omega \to 0)$ is the dc conductivity in the normal state measured at $T \gtrsim T_c$, and the superfluid density $\rho_s$ is determined for $T \ll T_c$; this scaling relation is valid for both the metallic a-b planes as well as along the poorly conducting c axis, in both the electron and hole-doped cuprates.\textsuperscript{70-73} In this representation, the scaling relation is valid over five orders of magnitude and as such is usually shown as a log-log plot. However, when only the a-b plane results are considered, the results span less than two orders of magnitude and it is convenient to rewrite this relation as

$$T_c \approx 0.23 (\rho_{dc} \rho_s)/8,$$

where $\rho_{dc} = 1/\sigma_{dc}$. The scaling relation now has a similar appearance to the well known Uemura relation,\textsuperscript{113} $T_c \propto \rho_s$, and may be shown using a linear scale. In Fig. 10

![Fig. 9. The temperature dependence of the in-plane frequency-dependent scattering rate in La$_{2-x}$Ba$_x$CuO$_4$ (with the phonons removed) in the far-infrared region for (a) $x = 0.095$, (b) the 1/8 phase, and (c) $x = 0.145$. (d) Detailed temperature dependence of the scattering rate for the 1/8 phase below 60 K. The values for $1/\tau_D$ determined from the Drude fits are indicated by the symbols at the origin. The dashed line indicates $1/\tau(\omega) = \omega$.](image-url)
the values for $T_c$ are plotted against $\rho_{dc} \rho_{so}/8$ determined in the $a$-$b$ planes for a variety of single-layer and double-layer cuprates. The values for La$_{2-x}$Ba$_x$CuO$_4$ for $x = 0.095$ ($T_c = 32$ K, $\sigma_{dc} = 4100 \pm 400 \Omega^{-1} \text{cm}^{-1}$ just above $T_c$, and $\rho_{so} = 8.8 \pm 0.8 \times 10^6 \text{cm}^{-2}$) are highlighted within the circles and fall on the general scaling line. These points also lie quite close to the related La$_{2-x}$Sr$_x$CuO$_4$ materials with comparable doping. This result is consistent with the observation that there is strong dissipation in the normal state.$^{114-117}$

\section*{B. $c$ axis}

\subsection*{1. Optical properties}

The temperature dependence of the reflectance of La$_{2-x}$Ba$_x$CuO$_4$ for light polarized along the poorly-conducting $c$ axis is shown in Figs. 11(a), (b) and (c) for the $x = 0.095, 0.125$ and $0.145$ dopings, respectively. The reflectance in this polarization is dramatically different than what is observed in the metallic $a$-$b$ planes (Fig. 5); as a result of the near total absence of a free-carrier contribution to the dielectric function, the reflectance is dominated by the normally infrared-active $c$ axis lattice modes. In the normal state as $\omega \to 0$ the reflectance displays a slight upturn, indicative of a weakly-conducting state; this is seen most clearly in the $x = 0.145$ material. Below $T_c$ a sharp plasma edge is observed in the reflectance at low frequency in Figs. 11(a) and (c) for the $x = 0.095$ and $0.145$ dopings, respectively. The onset of the plasma edge is extremely rapid in both materials. The $1/8$ phase material shows no hint of a plasma edge; however, this is not surprising as the bulk $T_c$ in this material is too low to be accessed in our current experimental configuration.

The $c$-axis plasma edge in the reflectance is observed in many of the cuprate superconductors,$^{118-126}$ and is attributed to the Josephson coupling of the copper-oxygen planes resulting in the formation of a supercurrent and the onset of a bulk 3D superconducting state.$^{127-129}$ It is interesting to note that the Josephson plasma edge is considerably broader in the more heavily-doped material; this observation is in agreement with the result for heavily-doped La$_{2-x}$Sr$_x$CuO$_4$ where it is suggested that the smearing of the plasma edge is due to spatial variations of the superconducting condensate.$^{125}$

The behavior along the $c$ axis is easier to understand when the conductivity is considered, shown in Figs. 12(a), (b) and (c) for the $x = 0.095, 0.125$ and $0.145$, respec-
TABLE II. The vibrational parameters for oscillator fits to the infrared-active phonon modes observed along the $c$ axis in La$_{1.905}$Ba$_{0.095}$CuO$_4$ at 295 K (HTT) and at 35 K (LTO), where $\omega_j$, $\gamma_j$, and $\Omega_j$ are the frequency, width and oscillator strength, respectively. The estimated errors are indicated in parenthesis. All units are in cm$^{-1}$.

<table>
<thead>
<tr>
<th>Mode $x$</th>
<th>$\omega_j$ (cm$^{-1}$)</th>
<th>$\gamma_j$ (cm$^{-1}$)</th>
<th>$\Omega_j$ (cm$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A_{2u}$</td>
<td>234.6 (0.2)</td>
<td>29 (0.6)</td>
<td>1085 (10)</td>
</tr>
<tr>
<td>$A_{2u}$</td>
<td>343.6 (0.1)</td>
<td>9.2 (0.5)</td>
<td>61 (4)</td>
</tr>
<tr>
<td>$A_{2u}$</td>
<td>381.0 (0.6)</td>
<td>34 (3)</td>
<td>71 (10)</td>
</tr>
<tr>
<td>$A_{2u}$</td>
<td>495.9 (0.1)</td>
<td>25 (0.6)</td>
<td>312 (5)</td>
</tr>
<tr>
<td>$B_{1u}$</td>
<td>132.7 (0.2)</td>
<td>5.7 (0.9)</td>
<td>55 (6)</td>
</tr>
<tr>
<td>$B_{1u}$</td>
<td>235.4 (0.2)</td>
<td>22 (0.6)</td>
<td>1110 (10)</td>
</tr>
<tr>
<td>$B_{1u}$</td>
<td>314.2 (0.1)</td>
<td>7.1 (0.2)</td>
<td>85 (2)</td>
</tr>
<tr>
<td>$B_{1u}$</td>
<td>348.0 (0.1)</td>
<td>4.6 (0.3)</td>
<td>113 (6)</td>
</tr>
<tr>
<td>$B_{1u}$</td>
<td>380.4 (0.9)</td>
<td>39 (6)</td>
<td>109 (13)</td>
</tr>
<tr>
<td>$B_{1u}$</td>
<td>495.8 (0.2)</td>
<td>19 (0.6)</td>
<td>338 (5)</td>
</tr>
</tbody>
</table>

The conductivity along the $c$ axis is attributed to thermally-activated hopping, which is consistent with the observation of an electronic background that decreases (weakly) with temperature. In addition, some of this decrease in the optical conductivity may also be due to a narrowing of vibrational features.

Despite the small value for the background conductivity, a suppression of the low-frequency conductivity due to the formation of a condensate is observed below $T_c$, in the $x = 0.095$ and 0.145 materials. The decrease in the low-frequency conductivity in the 1/8 phase below $T_{co}$ is likely due to the decoupling of the planes due to the formation of charge-stripe order; however, the dramatic decrease in the $c$-axis resistivity be-

![FIG. 12. The temperature dependence of the real part of the optical conductivity of La$_{2-x}$Ba$_x$CuO$_4$ for light polarized along the poorly-conducting $c$ axis for (a) $x = 0.095$, (b) $x = 0.125$, and (c) $x = 0.145$.](image-url)
can lead to significant uncertainties. Under these cir-

cumstances a more reliable approach is to examine the low-frequency response of the real part of the dielectric function, $\epsilon_1(\omega) = \epsilon_{\infty} - \omega^2 \epsilon_{p,S}/\omega^2$. For $T \ll T_c$ this method yields $\omega_{p,S} = 119 \pm 10$ cm$^{-1}$ for $x = 0.095$, and $\omega_{p,S} = 174 \pm 14$ cm$^{-1}$ for $x = 0.145$, which corresponds to effective penetration depths of approximately 13.4 and 9.1 $\mu$m, respectively. It is also possible to track the temperature dependence of the superfluid density using this method. The temperature dependence of the superfluid density for the $x = 0.095$ and 0.145 samples, expressed here as the ratio value of the square of the penetration depths $\lambda^2(0)/\lambda^2(T)$, is plotted against the reduced temperature $t = T/T_c$ and compared with optimally-doped La$_{1.85}$Sr$_{0.15}$CuO$_4$, shown in Fig. 13. In a comprehensive study of the evolution of the $c$-axis superfluid density in La$_{2-x}$Sr$_x$CuO$_4$ for a wide variety of dopings,$^{126}$ it may be noted that the superfluid density develops most rapidly for the optimally-doped material. It is therefore surprising that in underdoped La$_{1.905}$Ba$_0.095$CuO$_4$ the superfluid density develops extremely quickly just below $T_c$ before slowing below $T_c/2$; in this regard it is reminiscent of the behavior of a system with an isotropic $s$-wave energy gap,$^{131}$ while the more heavily doped $x = 0.145$ sample develops relatively slowly and is similar to what is observed in the copper-oxygen planes in a $d$-wave system.$^{132}$

We attribute this difference in behavior to the presence of a pseudogap in the underdoped materials where the antinodal region is gapped well above $T_c$. If the $c$-axis transport is dominated by the antinodal zone boundary region of the Fermi surface,$^{133-135}$ then when the Fermi pocket is gapped below $T_c$ the $c$-axis response will be dominated by the gap maximum $\Delta_0$ well away from the nodal region, and thus will not be sensitive to the $d$-wave nature of the system, instead resembling a more isotropic system.$^{72}$ The formation of charge order in the $x = 0.095$ material below $\approx 25$ K may frustrate the Josephson coupling between the planes,$^{34}$ leading to the observed slowing of the formation of the condensate below roughly $T_c/2$. On the other hand, the pseudogap is generally not present in materials close to optimal doping; below $T_c$ the entire Fermi surface is gapped. This suggests that in an optimally-doped material the $c$-axis response will be more sensitive to the $d$-wave nature of the energy-gap and therefore evolves more slowly, similar to the response observed in the copper-oxygen planes.$^{136}$

3. Parameter scaling

It was previously noted that the scaling relation $\rho_{s0}/8 \approx 4.4 \sigma_{dc} T_c$ is valid not only in the $a$-$b$ planes, but also along the $c$ axis.$^{70}$ While it is possible to plot the $c$ axis data in the form of $T_c$ versus $\rho_{dc} \rho_{s0}/8$, the highly anisotropic nature of these materials results in conductivities (resistivities) along the $c$ axis that are orders of magnitude smaller (larger) than those observed in the $a$-$b$ planes, resulting in a significant uncertainty in $\sigma_{dc} = \sigma_1(\omega \rightarrow 0)$ and the corresponding value for $\rho_{dc} = 1/\sigma_{dc}$. In addition, the scaling behavior of $\rho_{s0}$ emerges naturally if the superconductivity along the $c$ axis is considered to originate from Josephson coupling.$^{72}$

The $c$ axis values for the spectral weight of the condensate $\rho_{s0}/8$ are plotted as a function of $\sigma_{dc} T_c$ in Fig. 14 for a variety of single-layer and double-layer
FIG. 14. The log-log plot of the spectral weight of the superfluid density $N_c \equiv \rho_{s0}/8$ vs $\sigma_{dc} T_c$ along the $c$ axis for a variety of electron and hole-doped cuprates compared with La$_{2-x}$Ba$_x$CuO$_4$ for $x = 0.095$ and 0.145 (highlighted within the circles). The superscripts next to the symbols for La$_{2-x}$Sr$_x$CuO$_4$ and YBa$_2$Cu$_3$O$_{6+y}$ refer to different chemical dopings. The dashed line corresponds to the general result for the cuprates $\rho_{s0}/8 \approx 4.4 \sigma_{dc} T_c$.

cuprates. The dashed line is the best fit to the data, $\rho_{s0}/8 \approx 4.4 \sigma_{dc} T_c$. The $c$-axis values for La$_{2-x}$Ba$_x$CuO$_4$ for $x = 0.095$ ($T_c = 32$ K, $\sigma_{dc} = 3 \pm 1 \Omega^{-1}$ cm$^{-1}$ just above $T_c$, and $\rho_{s0} = 1.4 \pm 0.2 \times 10^4$ cm$^{-2}$) and 0.145 ($T_c = 24$ K, $\sigma_{dc} = 9 \pm 2 \Omega^{-1}$ cm$^{-1}$ just above $T_c$, and $\rho_{s0} = 3.0 \pm 0.3 \times 10^4$ cm$^{-2}$) are highlighted in the circles. These values and fall on the general scaling line and once again are quite close to the related La$_{2-x}$Sr$_x$CuO$_4$ materials with comparable doping.

C. The anomalous 1/8 phase

The behavior of the optical conductivity at low temperature in the $x = 1/8$ phase is fundamentally different than the other two dopings, where a response that is consistent with the opening of a superconducting $d$-wave energy gap is observed. The transfer of spectral weight from low to high frequency that develops close to or below $T_{so}$ is extremely unusual for a hole-doped cuprate and has the characteristics of an anisotropic transport gap on the Fermi surface due to the formation of charge or spin order. A partial gapping of the Fermi surface has been observed in other 2D materials with incommensurate charge-density waves (CDW’s). Unlike one-dimensional systems, where the modulation wave vector can perfectly span the Fermi surface and the CDW almost always leads to the complete gapping of the Fermi surface, in 2D perfect nesting is not possible and as a result the CDW gaps only a portion of the Fermi surface. Fortunately, ARPES measurements have been performed on this composition, so that we can analyze nesting of the Fermi surface by the measured modulation wave vectors. The charge-order wave vector is approximately equal to $4k_F$ in the antinodal region. There is certainly a gap on that portion of the Fermi surface, but it is already there as a pseudogap before the charge-stripe order appears. Therefore, the temperature dependence of the optical spectral weight transfer suggests a connection with the spin-stripe order for which the wave vector would nest parts of the Fermi arc. The ARPES measurements indicate that the gapping of the Fermi arc below $T_{so}$ is indistinguishable from the $d$-wave-like gap found in superconducting samples.

While the conventional density-wave picture is a possible candidate to explain the observed redistribution of spectral weight at low temperature, another possibility is that this shift is associated with superconducting correlations in this material. We are not able to access the bulk superconducting state in the $x = 1/8$ material; however, the previously discussed scaling relation indicates that because of the dramatically reduced value for the bulk superconductivity in this material ($T_c \approx 2.4$ K) the Josephson plasma frequency along the $c$ axis is estimated to be $\omega_{p,S} \approx 17$ cm$^{-1}$. The Josephson plasma edge in the reflectance typically occurs in a region close to the renormalized superconducting plasma frequency $\omega_{p,S} = \omega_{p,S}/\sqrt{\text{FIR}}$. Given the experimentally-determined value of $\epsilon_{1,c} \approx 30$ at 50 cm$^{-1}$, this yields a value of $\omega_{p,S} \approx 3$ cm$^{-1}$, below our ability to measure using current optical techniques. The strongly reduced value for the $c$-axis Josephson plasma edge for $x = 1/8$ is consistent with the disappearance of this feature in La$_{1.85-y}$Nd$_y$Sr$_{0.15}$CuO$_4$, which occurs when the increasing Nd concentration causes the structure to change from LTO to LTT, resulting in stripe order. To explain the abrupt change in the Josephson coupling in the latter case, despite the much weaker reduction in $T_c$, Himeda et al. proposed a pair-density-wave (PDW) superconducting state involving local $d$-wave symmetry but modulated by a sinusoidal envelope function; the envelope function has the same period as the spin modulation, but its extrema are aligned with the maxima of the charge modulation. The rotation of the stripe (and PDW) orientation by 90$^\circ$ from layer to layer leads to a frustration of the interlayer Josephson coupling. This concept was rediscovered after the observation of quasi-two-dimensional superconductivity by transport measurements on La$_{2-x}$Ba$_x$CuO$_4$ with $x = 1/8$.

The PDW proposal also has implications for the in-plane conductivity. Given a sinusoidally-modulated pair wave function, the superfluid density can be written as

$$\rho_s(r) = 0.5\rho_s(1 + \cos(2q \cdot r + \phi)), \quad (7)$$

where $q = (\frac{1}{3}, 0, 0)$ or $(0, \frac{1}{3}, 0)$ is the ordering wave vector (in reciprocal lattice units based on the HTT phase),
and φ is the phase of the modulation with respect to the crystal lattice. Barabash et al.\textsuperscript{144} showed that inhomogeneity of the superfluid density will cause some of the conductivity that would normally be at ω = 0 to shift to finite ω. Orenstein\textsuperscript{145} went further, and showed that sinusoidal modulation of ρ(\boldsymbol{r}) shifts conductivity to a peak at the renormalized superconducting plasma frequency \(\omega_{p,s} \approx \omega_{p,s}/\sqrt{T_c}\). From Table I we extrapolate an in-plane value of \(\omega_{p,s} \simeq 3250 \text{ cm}^{-1}\) for \(x = 1/8\). Using the experimentally-determined value of \(\epsilon_{1,0} \simeq 4\) at 2 eV for \(\epsilon_{\infty}\) [there is little or no temperature dependence of \(\epsilon_{1,0}(\omega)\) in this frequency region], then we estimate the in-plane \(\omega_{p,s} \simeq 1600 \text{ cm}^{-1}\). In considering Fig. 8(b), we already noted that some spectral weight is shifted from low frequency to above 300 cm\(^{-1}\). To emphasize this behavior, Fig. 15 shows the ratio of \(\sigma_1(\omega, T)\) to that at \(T = 60 \text{ K}\). The enhancement of \(\sigma_1\) at \(\sim 80 \text{ meV}\) is clearly present at 40 K and has doubled by 6 K; however, there is no enhancement at 50 K, which is already below \(T_{co}\). The development of the 80-meV feature on cooling correlates with the drop in in-plane resistivity.

To compare with Orenstein’s analysis,\textsuperscript{145} we have to allow for the fact that we have superconducting correlations but not long-range order for 40 K \(\geq T > 16 \text{ K}\). As a consequence, there should be a finite scattering rate associated with the “superconducting” features. The small but finite width of the \(\omega = 0\) (“Drude”) peak for \(16 \text{ K} < T \lesssim 50 \text{ K}\) is consistent with this expectation. The development of the finite-frequency peak is qualitatively compatible with Orenstein’s prediction for a modulated superconductor, although it appears at a frequency somewhat lower than \(\omega_{p,s}\). The shifted spectral weight appears to be pushed above \(2\Delta_0\), where ARPES studies\textsuperscript{56,57} indicate \(2\Delta_0 \approx 40 \text{ meV}\) in the antinodal region.

There are some experimental results that complicate this picture. The ARPES studies\textsuperscript{56,57} indicate a d-wave-like gap along the near-nodal arc for \(T \lesssim 40 \text{ K}\), whereas the PDW state is predicted to have a gapless near-nodal arc.\textsuperscript{146} The d-wave-like gap suggests the presence of a more uniform superconducting component that develops below \(\sim 40 \text{ K}\). Such a uniform component might be necessary to get bulk superconductivity at low temperature, as the correlation length of the PDW order is limited by that of the stripe order, which is restricted to several hundred Å.\textsuperscript{46,47} Of course, if a uniform component is present, then one returns to the challenge of explaining why the interlayer Josephson coupling is frustrated.

A different complication is that a substantial narrowing of the zero-frequency peak is already apparent at 50 K, where we have no enhancement at \(\sim 80 \text{ meV}\). At that temperature, we have charge stripe order but not spin order, and the spin order may be necessary to pin the phase of \(\rho(\boldsymbol{r})\) with respect to the lattice. Perhaps at 50 K, in the absence of pinning, the PDW correlations start to show up at low frequency but not at higher frequency. In any case, the high-frequency peak remains at 6 K, below the point where 2D superconducting order develops,\textsuperscript{29} so that the modulated \(\rho_s\) should still be present there.

### IV. CONCLUSIONS

The temperature dependence of the optical properties of \(\text{La}_{2−x}\text{Ba}_x\text{CuO}_4\) for light polarized in the \(a\)-\(b\) planes and along the poorly-conducting \(c\) axis has been determined for \(x = 0.095, 0.125\) and 0.145. The \(x = 0.095\) and 0.145 samples are superconducting, with \(T_c = 32\) and \(\simeq 24 \text{ K}\), respectively. The in-plane optical conductivity can be characterized as a Drude-like response that narrows with decreasing temperature, superimposed on a flat, incoherent background; below \(T_c\) missing low-frequency spectral weight is the hallmark of the formation of a superconducting condensate. Interestingly, in both cases \(\omega_{p,s}^2/\omega_{p,b}^2 < 1\), indicating that only a fraction of the normal-state carriers condense; this is consistent with the observation of strong dissipation in the normal state.

The optical properties along the poorly-conducting \(c\) axis in the \(x = 0.095\) and 0.145 materials are insulating in character, dominated by the normally infrared-active phonons with only a weak electronic background; new lattice modes appear at the HTT\(\rightarrow\)LTO transition. Below \(T_c\) a sharp plasma edge is observed in the \(c\)-axis reflectance; this feature is attributed to the formation of a supercurrent along the \(c\) axis due to Josephson coupling.
between the copper-oxygen planes. Despite the dramatic reduction in the c-axis resistivity below $T_{\text{so}}$ in the 1/8 material, no Josephson plasma edge or hint of metallic behavior is observed. In the superconducting materials, the spectral weight of the condensate for the $a$-$b$ plane and the $c$ axis fall on the universal scaling line $\rho_{\text{so}}/s \simeq 4.4 \sigma_{\text{dc}} T_c$.

The behavior of the anomalous $x = 1/8$ phase in which the superconductivity is dramatically weakened ($T_c \simeq 2.4$ K) stands out in stark contrast. The in-plane optical properties are similar to the other two dopings at high temperature; however, close to or below $T_{\text{so}}$ there is an unusual suppression of the reflectance over much of the infrared region and a transfer of spectral weight from low frequency to energies above $\sim 20$ meV, indicating the formation of a momentum-dependent energy gap. Below $T_{\text{so}}$ the free-carrier response continues to narrow dramatically with decreasing temperature, to the extent that at the lowest measured temperature the spectral weight associated with the free carriers can no longer be observed. This response mimics the missing spectral weight associated with the formation of a condensate; however, we are currently unable to distinguish a momentum-dependent CDW gap from the similar behavior expected from a PDW state.

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