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OPTICAL CONDUCTIVITY OF $Nd_{1.85}Ce_{0.15}CuO_4$: STRENGTH OF THE CONDENSATE

C. C. HOMES[a,]*, J. L. PENG[b], R. L. GREENE[b] and B. P. CLAYMAN[c]

[a]Department of Physics, Brookhaven National Laboratory, Upton, NY 11973-5000, USA
[b]Center for Superconductivity Research, Department of Physics and Astronomy, University of Maryland, College Park, MD 20742, USA
[c]Department of Physics, Simon Fraser University, Burnaby, B.C., Canada V5A 1S6

Abstract—The *ab*-plane reflectance of a $Nd_{1.85}Ce_{0.15}CuO_4$ single crystal ($T_c = 23$ K) has been measured from ≈ 30 to 9500 cm^{-1} above and below T_c, and the optical properties calculated from a Kramers–Kronig analysis. A rich phonon spectrum is observed with some new spectral features appearing below ≈ 100 K. The normal-state optical conductivity may be described by a Drude-like component and an overdamped mid infrared component. Below T_c the plasma frequency of the condensate is $\omega_{pS} \approx 10\,000$ cm^{-1}. The London penetration depth is determined to be $\lambda_{ab} = 1600 \pm 100$ Å, which places it well off the Uemura line. Estimates of the electron–phonon coupling from normal-state transport measurements yield $\lambda_{tr} < 0.5$. The small values for the penetration depth and the electron–phonon coupling constant suggest that the superconductivity in this material is not due to the electron–phonon mechanism, and is different to that in other hole-doped superconducting cuprates. © 1998 Elsevier Science Ltd. All rights reserved

In most high-temperature (high-T_c) superconducting cuprates, such as $YBa_2Cu_3O_{7-\delta}$ and $La_{2-x}Sr_xCuO_4$, the charge carriers are doped holes. However, in $Nd_{2-x}Ce_xCuO_4$, where superconductivity is induced by substituting Nd^{3+} with Ce^{4+}, the CuO_2 planes are believed to be doped with electrons [1, 2] as well as holes [3].

The $Nd_{2-x}Ce_xCuO_4$ system has attracted a great deal of interest because of its possible conventional BCS *s*-wave pairing in the superconducting state, as opposed to the unconventional *d*-wave behavior proposed for the hole-doped cuprates [4, 5]. The microwave surface impedance measurements on both thin films and single crystals have shown evidence for a conventional BCS *s*-wave behavior with a gap of $2\Delta \approx 4k_BT_c$ [6–8]. Tunneling measurements have also shown a resemblance to conventional superconductors [9]. However, the magnetic field dependence of the specific heat anomaly [10, 11] and thin film transmission [12] of $Nd_{2-x}Ce_xCuO_4$ both show a non-BCS-like behavior.

Whether or not this system can be considered as a conventional BCS-type superconductor is an important question, given the strong evidence that the other hole-doped cuprates are not. The optical properties of materials in the normal and superconducting states can provide a wealth of information about the electrodynamics of the system, and can yield important information about the nature of the superconductivity.

Large, single crystals of $Nd_{1.85}Ce_{0.15}CuO_4$ were grown from a CuO-based flux using a directional solidification technique [13]. During the course of the growth, the flux was allowed to flow out of the crucible at the end of the growth process, so that free-standing crystals are left in the bottom of the crucible that do not need to be mechanically separated from the flux. To induce superconductivity, the crystals were annealed in an inert gas atmosphere. Both resistivity and magnetization showed a sharp superconducting transition at 23 K, with a width of < 1 K [13].

The reflectance of a single crystal of $Nd_{1.85}Ce_{0.15}CuO_4$ has been measured for the radiation polarized parallel to the *a–b* plane from ≈ 30 to 9500 cm^{-1}, at temperatures above and below T_c on a Bruker IFS113v Fourier transform interferometer, using a sensitive overfilling technique [14]. The crystal examined in this case was $\approx 2 \times 2$ mm in the *a–b* plane, and had a flat, mirror-like surface that was free of flux. The optical properties (i.e. the complex conductivity $\tilde{\sigma} = \sigma_1 + i\sigma_2$) have been calculated from a Kramers–Kronig analysis of the reflectance. At low frequency, the reflectance was extrapolated to zero frequency by assuming a Hagen–Rubens $1 - R \propto \sqrt{\omega}$ dependence above T_c, and a superconducting $1 - R \propto \omega^2$ dependence below T_c. The reflectance has been extended to ≈ 35 eV using data from related materials [15, 16], above which a free-electron ($R \propto \omega^{-4}$) behavior was assumed.

The reflectance of $Nd_{1.85}Ce_{0.15}CuO_4$ is shown in Fig. 1(a) from ≈ 30 to 1000 cm^{-1} in the normal state and below T_c. The optical conductivity has been determined from a Kramers–Kronig analysis of the reflectance and is shown in Fig. 1(b) for the same temperatures over

*Corresponding author. Tel: +1-516-344-7579; fax: +1-516-344-2739; e-mail: homes@bnl.gov

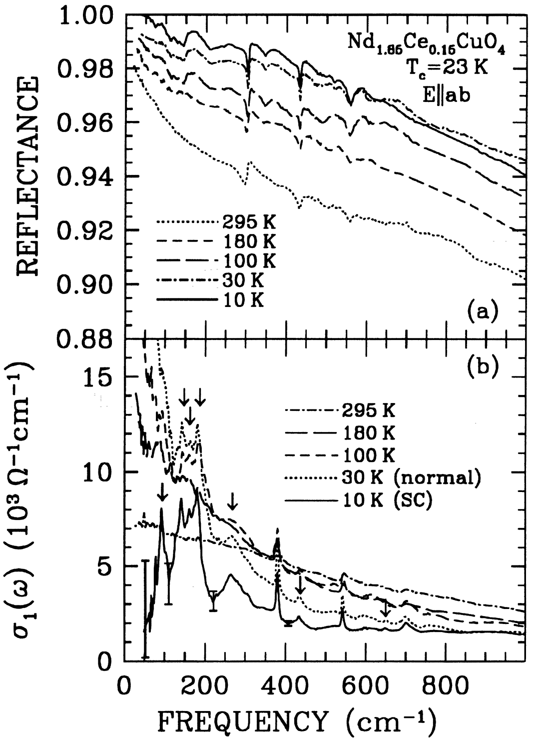

Fig. 1. (a) The reflectance of $Nd_{1.85}Ce_{0.15}CuO_4$ ($T_c = 23$ K) for $E \| ab$) at several temperatures above T_c (295, 180, 100 and 30 K), and below T_c at 10 K from ≈ 30 to $1000\ \mathrm{cm}^{-1}$. At low temperatures, the reflectance is over 95% below $\approx 1000\ \mathrm{cm}^{-1}$. (b) The conductivity at the same temperatures over the same frequency range. The arrows indicate the new spectral features observed below ≈ 100 K, while the error bars are an estimate of the uncertainty in the conductivity below T_c at 10 K.

the same frequency range. The free-carrier component may be characterized as a Lorentzian centered at zero frequency which is narrowing rapidly with decreasing temperature. There are a number of new spectral features which appear below ≈ 100 K, which are thought to be mainly phonons. Interestingly, this is the same temperature at which the resistivity changes from linear to quadratic behavior.

Below T_c the conductivity decreases rapidly at low frequency, this 'missing area' in the conductivity is due to the transfer of spectral weight from the free carriers into the delta function at zero frequency. The strength of the condensate $\delta \propto \omega_{pS}^2$, where the plasma frequency of the condensate (ω_{pS}) has been determined using two different techniques:

1. the optical conductivity sum rule $\omega_{pS}^2 = (120/\pi) \int_0^\infty [\sigma_{1n}(\omega) - \sigma_{1s}(\omega)]\, \mathrm{d}\omega$, where $\sigma_{1n}(\omega)$ is the conductivity in the normal state just above T_c, and $\sigma_{1s}(\omega)$ is the conductivity for $T \ll T_c$;
2. an analysis of the real part of the dielectric function based on a clean limit approach where all the free carriers condense into the δ function, which yields $\epsilon_1(\omega) = \epsilon_\infty' - \omega_{pS}^2/\omega^2$ for $T \ll T_c$.

Both methods yield $\omega_{pS} \approx 10\,000\ \mathrm{cm}^{-1}$ [17]. The penetration depth is also related to the plasma frequency of the condensate, $1/\lambda(T) = 2\pi\omega_{pS}(T)$, giving $\lambda_{ab} \approx 1600$ Å at 10 K.

It has been noted that in many of the cuprate-based high temperature superconductors $\delta \propto T_c$ [18], which is shown in Fig. 2 for a variety of materials. However, the value of δ for $Nd_{1.85}Ce_{0.15}CuO_4$ is much larger than the low T_c for this system would suggest, indicating that this material is not like other cuprate superconductors.

An examination of the conductivity in Fig. 1(b) below T_c indicates that the large errors associated with the conductivity at low frequency (assuming an error of $\pm 0.1\%$ in the reflectance) precludes an unambiguous determination of the nature of the energy gap in this system. However, the low value of the electron–phonon coupling constant λ_{tr} determined from transport measurements [19], and the absence of any phonon anomalies due to a Holstein mechanism [20] at or below T_c, suggests that while a BCS-type mechanism may not be ruled out, the superconductivity in this material is probably not phonon mediated.

In summary, the strength of the condensate in $Nd_{1.85}Ce_{0.15}CuO_4$ has been calculated from the optical properties. The large value for ω_{pS} (and small value of λ_{ab}) is suggestive of a 90 K superconductor, rather than the observed $T_c = 23$ K. This result, when coupled with the small value of λ_{tr} and the absence of any phonon anomalies at T_c indicates that superconductivity of this

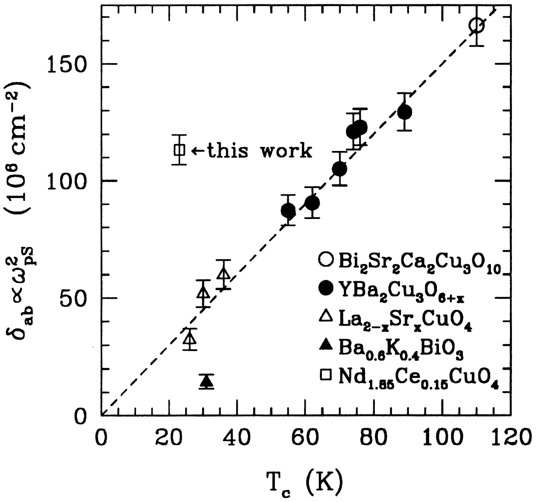

Fig. 2. The strength of the condensate $\delta \propto \omega_{pS}^2$ as a function of T_c for $La_{2-x}Sr_xCuO_4$ (open triangles), $YBa_2Cu_3O_{6+x}$ (filled circles), $Bi_2Sr_2Ca_2Cu_3O_{10}$ (open circle) [18], $Ba_{0.6}K_{0.4}BiO_3$ (solid triangle, [21], and $Nd_{1.85}Ce_{0.15}CuO_4$ (open square, this work). The dashed line is the universal Uemura line. While the majority of the underdoped and optimally-doped cuprates fall in the Uemura line, and the bismuthate just below, the value for $Nd_{1.85}Ce_{0.15}CuO_4$ obtained in this work places it well outside this linear relationship. This suggests that the nature of the superconductivity in $Nd_{1.85}Ce_{0.15}CuO_4$ is different than in other hole-doped cuprates.

material is probably not phonon mediated, yet it is also unlike other cuprate superconductors.

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