Anisotropic optical conductivity of $Nd_{2-x}Ce_xCuO_4$ thin films

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Optical conductivity spectra $\sigma_1(\omega)$ of $Nd_{2-x}Ce_xCuO_4$ thin films, measured by the reflectance-transmittance method (R-T method) which has been proposed to investigate FIR spectroscopy, are investigated based on the anisotropic pairing model. Precise measurements of the frequency-dependent conductivity $\sigma_1(\omega)$ enable us to examine quantitatively the nature of the superconducting gap through FIR properties for the first time in high-T_c superconductors. We show that the behavior of optical conductivity $\sigma_1(\omega)$ is consistent with the anisotropic superconducting gap and is well explained by the formula for d-wave pairing in the FIR region. Our results suggest that the electron-doped cuprate superconductors $Nd_{2-x}Ce_xCuO_4$ have nodes in the superconducting gap.

Keywords: electron-doped superconductors, optical conductivity, anisotropic superconducting gap, R-T method

I. Introduction

Oxide high-T_c superconductors have been investigated intensively over the last decade.[1,2] d-wave superconductivity is now well established for hole-doped superconductors. However, there is a class of high-T_c superconductors doped with electrons,[3,4] for which both s-wave[5] and d-wave pairing[6–8] have been reported. $Nd_{2-x}Ce_xCuO_4$ is a typical example of electron-doped materials and the symmetry of Cooper pairs has been a controversial issue. It is important to examine the symmetry of Cooper pairs in the study of high-T_c superconductors.

Far-infrared (FIR) spectroscopy is a powerful technique to investigate the nature of the superconducting gap. The conventional FIR spectroscopy based on a Kramers-Kronig transformation, however, is rather unfavorable for studying electronic properties in the low energy regime of the FIR region.[9,10] Since the superconducting gap Δ in $Nd_{2-x}Ce_xCuO_4$ is very small, there have been no reports on the study of the nature of the superconducting gap of $Nd_{2-x}Ce_xCuO_4$ through such techniques, although there have been a number of reports on the FIR spectroscopy of $Nd_{2-x}Ce_xCuO_4$.[11,12] Recently, a new method to examine FIR spectroscopy has been developed without the need for evaluating the Kramers-Kronig transformation.[13] In this method, the optical conductivity is estimated from the data of reflectance spectra $R(\omega)$ and transmittance spectra $T(\omega)$ by substituting them into a set of coupled equations. The new method is free from the conventional difficulties in the FIR region since we do not need the aid of the Kramers-Kronig transformation.[14] This method is referred to as the R-T method in this paper.

The purpose of this paper is to investigate FIR optical properties of $Nd_{2-x}Ce_xCuO_4$ obtained by the R-T method from the viewpoint of unconventional superconductors. We will show that the available data for the optical conductivity and transmittance are well explained by the d-wave pairing model in the clean limit. The value of the superconducting gap is estimated as $2\Delta \sim 50-60$ cm^{-1}, which is consistent with the available value estimated by scanning tunneling spectroscopy.[5]

II. Theory and Measured Conductivity

The frequency-dependent conductivity $\sigma(\omega)$ has been calculated by Mattis and Bardeen,[15] Abrikosov *et al.*[16] and Skalski *et al.*[17] for isotropic superconductors. The original Mattis-Bardeen theory was tested for a conventional type-I s-wave superconductor, where the coherence length ξ and magnetic penetration depth λ satisfy $\xi \gg \lambda$. The opposite limit $\xi \ll \lambda$ (London limit) was also examined for s-wave pairing by field theoretical treatments.[17] For the high-T_c compounds of type-II superconductor with small coherence length, the formula in the London limit is appropriate for optical conductivity measurements. Recently, the conductivity $\sigma(\omega)$ of an unconventional superconductor has been examined theoretically in the London limit.[18–22] The current response function, from which the optical conductivity is derived, is given by[18]

$$K_{ij}(\mathbf{q},\omega_m) = -\frac{e^2k_F^2}{m^2c}\sum_k \hat{k}_i\hat{k}_j\frac{1}{\beta}\sum_n Tr[G(\mathbf{k}_+,\epsilon_n + \omega_m)G(\mathbf{k}_-,\epsilon_n)], \quad (1)$$

where $\mathbf{k}_\pm = \mathbf{k} \pm \mathbf{q}/2$ and $\epsilon_n = (2n+1)\pi/\beta$. The single-particle matrix Green's function is given by

$$G(\mathbf{k},\epsilon_n) = \frac{i(\epsilon_n - \Sigma(\epsilon_n))\tau^0 + \xi_k\tau^3 + \Delta_k\tau^1}{(\epsilon_n - \Sigma(\epsilon_n))^2 + \xi_k^2 + \Delta_k^2}, \quad (2)$$

where Δ_k is the anisotropic order parameter and $\Sigma(\epsilon_n)$ is the self-energy due to impurity scattering. τ^i $(i = 0,1,\cdots)$ denote Pauli matrices. Since we consider the case where $\xi \ll \lambda$ holds, the real part of optical conductivity is well approximated by the formula in the London limit:

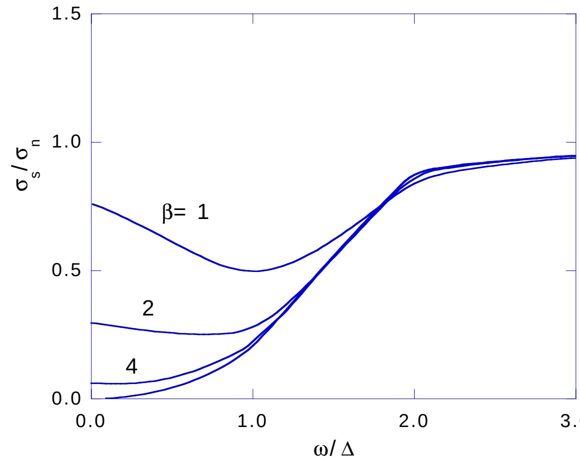

FIG. 1. Optical conductivity as a function of ω for several values of temperature. From the top $T/\Delta = 1/\beta = 1, 1/2, 1/4$ and 0.

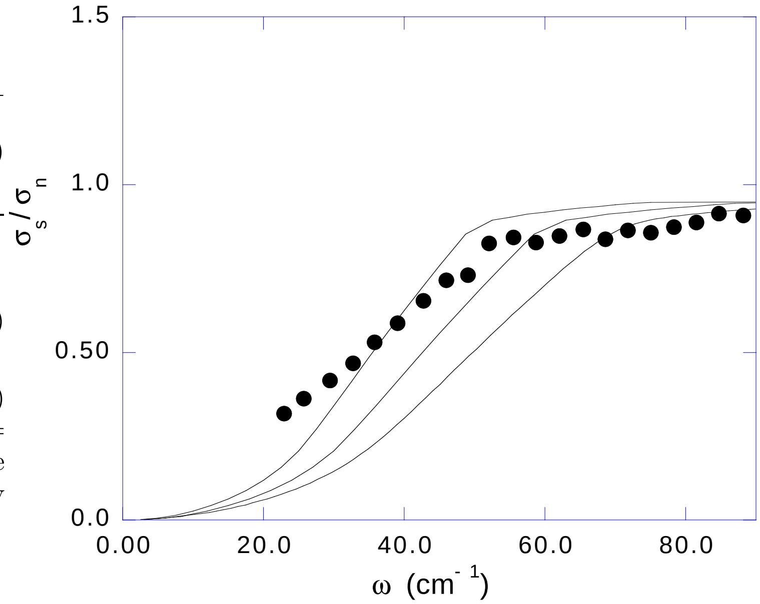

FIG. 2. Optical conductivity (circles) by the R-T method and theoretical predictions at $T = 0$ (solid curves). From the left $2\Delta = 50\mathrm{cm}^{-1}$, $60\mathrm{cm}^{-1}$ and $70\mathrm{cm}^{-1}$.

$$\sigma_{ij}(\omega) = -\frac{c}{\omega}\lim_{q\to 0}\mathrm{Im}K_{ij}(\mathbf{q},\omega). \quad (3)$$

The expressions of $K_{ij}(\mathbf{q},\omega)$ for real and imaginary parts were given in ref. 20. Our focus is the collisionless limit of the normalized conductivity to compare it with the data for $Nd_{2-x}Ce_xCuO_4$ since $\xi \ll \ell$ holds for the mean-free path ℓ. For anisotropic superconducting order parameter Δ_k such that the average over the Fermi surface vanishes $\langle\Delta_k\rangle = 0$, the expression for $\sigma_{xx}(\omega)$ in the collisionless limit on the plane is simply given by[19]

$$\frac{\sigma_{xx}(\omega)}{\sigma_n} = \frac{1}{2\omega}\int_{-\infty}^{\infty}dx\langle\mathrm{Re}\frac{|x|}{(x^2-\Delta_k^2)^{1/2}}\rangle\langle\mathrm{Re}\frac{|x-\omega|}{[(x-\omega)^2-\Delta_k^2]^{1/2}}\rangle[\tanh(\frac{\beta x}{2}) - \tanh(\frac{\beta(x-\omega)}{2})], \quad (4)$$

which is an angle-dependent generalization of the Mattis-Bardeen formula. For the d-wave symmetry, the average over the Fermi surface denoted by the angular brackets is defined as

$$\langle\mathrm{Re}\frac{x}{(x^2-\Delta_k^2)^{1/2}}\rangle = \mathrm{Re}\int\frac{d\phi}{2\pi}\frac{x}{[x^2-(\Delta\cos(2\phi))^2]^{1/2}}, \quad (5)$$

where the order parameter is factorized as $\Delta_k = \Delta\cos(2\phi)$. In Fig.1, we show the behaviors of $\sigma_s(\omega) \equiv \sigma_{xx}(\omega)$ as a function of ω for several values of temperature T. The FIR behaviors reflect the lines of nodes on the Fermi surface.

FIR reflection $R(\omega)$ and transmission $T(\omega)$ measurements were performed for $Nd_{2-x}Ce_xCuO_4$ $(x = 0.15)$ thin films deposited by laser ablation onto (001) MgO substrates. The thickness of the $Nd_{2-x}Ce_xCuO_4$ thin film was about 40 nm. T_c was estimated to be ∼20K. The electric field of the FIR radiation was predominantly parallel to the a-b plane. The conductivity spectra were evaluated by the R-T method from the data for $R(\omega)$ and $T(\omega)$ at $T = 4.3$ and 30K.[14]

The R-T method provides us with reliable spectroscopic data in the FIR region due to which a comparison between the experimental data and that of theoretical analysis is possible. In the R-T method, both the refrectance spectra $R(\omega)$ and the transmittance spectra $T(\omega)$ are measured experimentally from which a set of coupled equations is derived describing the transmittance and reflectance of a thin film on a substrate. The coupled equations are solved numerically by the Newton method to determine the optical conductivity. This method is free from the difficulties in the FIR region which occur commonly in the conventional method employing a Kramers-Kronig transformation. In Fig.2, we show the observed data and theoretical curves at $T = 0$ for $2\Delta = 50$, 60 and 70 cm^{-1}. The experimental data $\sigma_1(3.4K)/\sigma_1(30K)$ normalized by the normal state values at $T = 30$K are shown in Fig.2. It is obvious from the experimental results that there is no evidence of a true gap, which is suggestive of an anisotropic superconducting gap, since the spectral weight of conductivity should vanish for $\omega \leq 2\Delta$ at $T = 0$ in conventional isotropic superconductors. It is also shown in Fig.2 that they are well fitted by the curve with $2\Delta = 50$ cm^{-1}, which is consistent with the value estimated by scanning tunneling spectroscopy measurements.[5]

A transmission curve is also presented in Fig.3, where T_S/T_N, the ratio of the transmission in the superconducting (T =3.4K) to that in the normal state (T =30K), is the experimentally measured quantity. The following phenomenological expression for T_S/T_N is employed to determine the transmission curve theoretically,[23]

$$\frac{T_S}{T_N} = \frac{1}{[T_N^{1/2} + (1 - T_N^{1/2})(\sigma_1/\sigma_n)]^2 + [(1 - T_N^{1/2})(\sigma_2/\sigma_n)]^2}, \qquad (6)$$

where σ_1 and σ_2 are real and imaginary parts of the conductivity $-(c/\omega)K(\mathbf{q}, \omega)$ for $\mathbf{q} \to 0$, respectively. T_N is determined as $T_N \simeq 0.08$ from the expression for the ratio of the power transmitted with a film to that with no film given as

$$T_N = 1/[1 + \sigma_n d \frac{Z_0}{n+1}]^2. \qquad (7)$$

Here, d is the film thickness, n is the index of refraction of the substrate, and Z_0 is the impedance of free space. We have assigned the following values; $d = 4 \times 10^{-6}$cm, $n = 3.13$, $Z_0 = 377\Omega$, $\sigma_n \approx 7 \times 10^3 \Omega^{-1}$cm^{-1}. Apparently, the ω-dependence of measured transmittance agrees with the theoretical curve for $2\Delta = 50$ ~60cm^{-1} as shown in Fig.3. The agreement between the observed quantities and theoretical curve is significant, which should be compared to the isotropic BCS prediction calculated from the Mattis-Bardeen equations.[24]

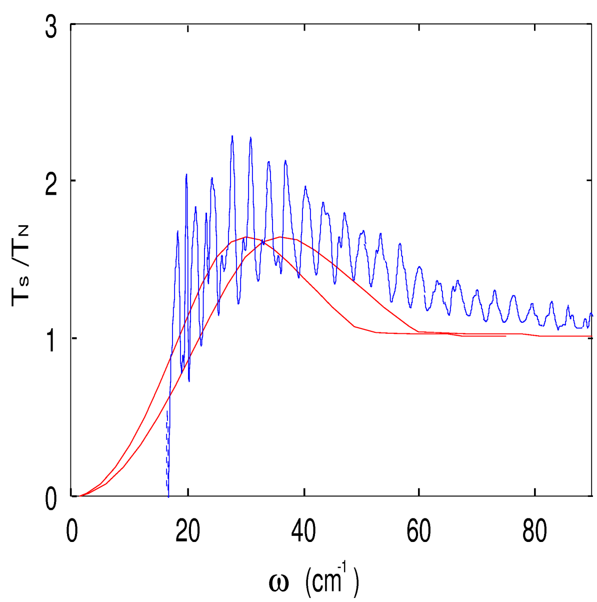

FIG. 3. Observed transmittance and the theoretical curve at $T = 0$ (solid curve) for $2\Delta = 50$cm^{-1} (left) and 60cm^{-1} (right).

III. Summary

We have successfully made a comparison between experimental data and theoretical data for the optical conductivity of $Nd_{2-x}Ce_xCuO_4$ in the FIR region for the first time. We have shown that there is a reasonable agreement between the optical conductivity $\sigma_1(\omega)$ observed by the R-T method and theoretical analysis without adjustable parameters except the superconducting gap. An estimate of 50~60 cm^{-1} for the superconducting gap is consistent from both the experimental and theoretical aspects. The FIR optical conductivity suggests that the superconducting gap of electron-doped $Nd_{2-x}Ce_xCuO_4$ is an unconventional one with nodes on the Fermi surface. The anisotropic nature of electron-doped superconductors is consistent with the recent researches performed for the one-band and three-band Hubbard models.[25–28] If the superconducting gap is anisotropic for the electron-doped superconductors, there is a possibility that both the hole-doped and electron-doped cuprates superconductors are governed by the same superconductivity mechanism.

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