APPLIED PHYSICS LETTERS VOLUME 77, NUMBER 3 17 JULY 2000

Submillimeter spectroscopy of tilted $Nd_{1.85}Ce_{0.15}CuO_{4-\delta}$ films: Observation of a mixed *ac*-plane excitation

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(Received 21 March 2000; accepted for publication 24 May 2000)

The anisotropic conductivity of a series of tilted $Nd_{1.85}Ce_{0.15}CuO_{4-\delta}$ thin films was measured by quasioptical spectroscopy in the frequency range $6\ cm^{-1}<\nu<40\ cm^{-1}$. Two characteristic features have been observed in the low-temperature transmission spectra. The first one at $\nu=12\ cm^{-1}$ was shown to reflect the c-axis plasma frequency of $Nd_{1.85}Ce_{0.15}CuO_{4-\delta}$. The second feature represents a mixed ab-plane/c-axis excitation. The frequency of this resonance may be changed in a controllable way by rotating the polarization of the incident radiation. © *2000 American Institute of Physics.* [S0003-6951(00)03629-9]

Among the cuprate superconductors, $Nd_{2-x}Ce_xCuO_4$ (NCCO) has attracted considerable interest because it can be considered as an electron-doped compound,[1] which reveals the (T') tetragonal structure and is extremely two-dimensional.[1,2] Earlier in-plane microwave experiments on NCCO (Refs. 3–5) appeared to be consistent with the conventional s-wave BCS predictions. However, recent data including flux quantization[6] and penetration depth at microwave[7] and at rf frequencies[8] have provided strong experimental evidence for d-wave symmetry of the pairing state.

In contrast to a number of ab-plane experiments, there exists minimal information concerning the c-axis properties of NCCO, primarily due to the small dimensions of the samples along the c-axis. Most experiments on c-axis dynamics[9,10] were carried out using ceramic NCCO samples. Alternatively, the method of grazing-incidence reflection is known to be a powerful technique in extracting c-axis properties of strongly anisotropic superconductors.[11] The experiments using, instead of the grazing reflection, a tilted sample[12] and normal-incidence geometry may be considered as a modification of this method.

In this letter, we present the results of submillimeter-wave ($6\ cm^{-1}<\nu<40\ cm^{-1}$) experiments on tilted $Nd_{1.85}Ce_{0.15}CuO_{4-\delta}$ films at low temperatures. The spectra reveal two features below the superconducting transition, which are identified as: (i) plasma resonance along the c-axis, and (ii) mixed ab-plane/c-axis excitation. Using the measurements at different polarizations of the incident radiation, it was possible to separate the effective conductivity at a tilted angle into components within the ab plane and along the c axis.

The experiments were carried out on different $Nd_{1.85}Ce_{0.15}CuO_{4-\delta}$ films yielding similar results. For simplicity, the results from only one film are presented. The films were prepared using a two-beam-laser deposition on yttrium-stabilized ZrO_2 substrates.[13] X-ray analysis revealed the c-axis orientation of the films. The substrate of the film was tilted from the (001) plane by an angle $\alpha=0.8°\pm0.4°$. Therefore, the film was also tilted by the same angle from the ideal c-axis orientation. The ac-susceptibility measurements revealed a narrow superconducting transition ($\Delta T[10\%-90\%]=0.9\ K$) with the onset temperature of 20.1 K. The film thickness, estimated from the deposition time, was $d=170\ nm$.

The transmission experiments in the frequency range $6\ cm^{-1}<\nu<40\ cm^{-1}$ were carried out in a quasioptical arrangement,[14] which allows the measurements of both, transmission and phase shift of a film on a substrate. The properties of the substrate were determined in a separate experiment. Utilizing the Fresnel optical formulas for the complex transmission coefficient of the substrate–film system, the absolute values of the complex conductivity $\sigma^*=\sigma_1+i\sigma_2$ were determined directly from the observed spectra without any approximations. The radiation in this method is continuously tunable in frequency and is linearly polarized. This allows one to perform experiments for different directions of the currents induced in the sample. The geometry of the transmission experiment is represented in the inset of Fig. 1.

Figure 1 shows the transmission spectra of a $Nd_{1.85}Ce_{0.15}CuO_{4-\delta}$ thin film at $T=6\ K$ and at $T\approx T_C$. A new feature that can be observed in the spectrum of the superconducting state is a transmission peak at $\nu=12\ cm^{-1}$. This feature may be called ‘‘antiresonance’’ because it corresponds to an *increase* in transmission. In addition to this antiresonance, a suppression of the low-temperature transmission is seen in the frequency range $18\ cm^{-1}<\nu<30\ cm^{-1}$. This suppression corresponds to a broad resonance and is somewhat masked by the interference pattern caused by the substrate.

Figure 2 shows the effective conductivity of the film as

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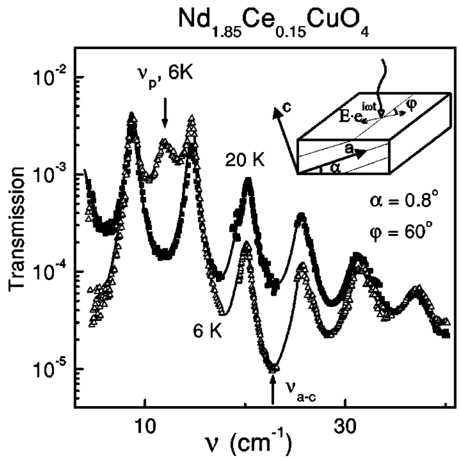

FIG. 1. Submillimeter-wave transmission spectra of a $Nd_{1.85}Ce_{0.15}CuO_{4-\delta}$ film at $T=T_C$ (20 K) and at $T=6$ K. Lines are drawn to guide the eye. Arrows indicate the position of the c-axis plasma frequency and of a mixed ac-plane excitation. The inset shows the geometry of the transmission experiment.

calculated from transmission and phase-shift spectra for $T=6$ K. Because the exact expressions for the complex transmission coefficient were used, the substrate interference patterns are absent in this presentation. The effective conductivity is shown for different orientations of incident radiation and is labeled by the angle φ (inset of Fig. 1).

The comparison of Figs. 1 and 2 clearly shows that a zero crossing of σ_2 and a peak in the loss function $\mathrm{Im}(1/\varepsilon^*)$ correspond to the antiresonance at $\nu=12\,\mathrm{cm}^{-1}$. The suppression of the transmission at $\nu\sim 23\,\mathrm{cm}^{-1}$ yields a peak in the real part of conductivity. Interestingly, both features exhibit a completely different dependence upon the polarization plane angle (φ). As will be seen below, the antiresonance at $12\,\mathrm{cm}^{-1}$ corresponds to the c-axis plasma frequency and is, therefore, independent of incident radiation polarization. In contrast, the peak at $23\,\mathrm{cm}^{-1}$ ($\varphi=60°$) represents the mixed ab-plane/c-axis excitation. The degree of mixing may be controlled by the geometry of the experiment, which explains the φ dependence of the peak position.

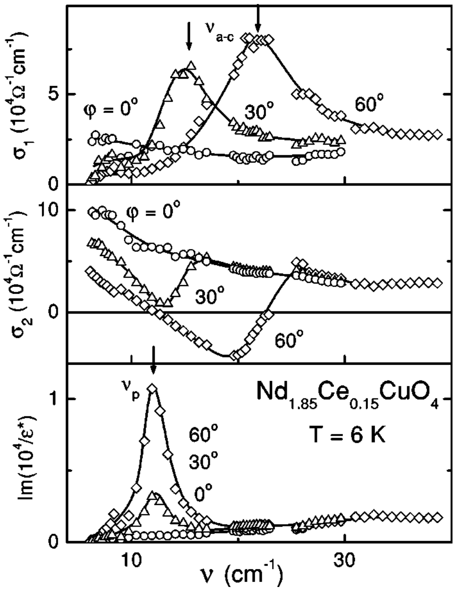

FIG. 2. Low-temperature conductivity spectra of a $Nd_{1.85}Ce_{0.15}CuO_{4-\delta}$ film for different orientations of the incident radiation. Lines are drawn to guide the eye. Upper panel: σ_1. Middle panel: σ_2. Lower panel: imaginary part of the loss function $\mathrm{Im}(1/\varepsilon^*)$. Arrows indicate the position of the c-axis plasma frequency and of the mixed ac-plane excitation.

The spectra for $\varphi=0°$ correspond to currents flowing only within the ab planes and resemble the spectra of a superconductor below the gap frequency: σ_1 has only a weak frequency dependence and is suppressed by approximately a factor of 3 compared to the normal-state conductivity, whereas σ_2 is strongly enhanced compared to the normal state and reveals approximately a $1/\omega$ frequency dependence. From its slope the low-frequency penetration depth $\lambda_{ab}(6\text{ K})=230\pm 30\,\mathrm{nm}$ can be estimated.

In order to calculate the effective conductivity of a tilted sample, consider a free-standing film of thickness d in a uniform electromagnetic field $Ee^{-i\omega t}$, as shown in the inset of Fig. 1. For simplicity, the film is assumed to be thin compared to the penetration depth, $d\ll\lambda$. In this case, the current and field distribution may be considered to be uniform. Taking into account the charges formed at the surface, the following equation for the effective conductivity is obtained:

$$\sigma_{\text{eff}}=\frac{-i\varepsilon_0\omega(\sigma_a\cos^2\alpha+\sigma_c\sin^2\alpha)+\sigma_a\sigma_c}{-i\varepsilon_0\omega+\sigma_a\sin^2\alpha+\sigma_c\cos^2\alpha}. \qquad (1)$$

Here, ε_0 is the permittivity of free space, σ_a (σ_c) is the complex conductivity in the ab plane (along the c axis), and ω is the angular frequency.

Within the approximation $\alpha\approx\sin\alpha\ll 1$ and $|\sigma_a|\gg|\sigma_c|$, Eq. (1) can be written as

$$\sigma_{\text{eff}}=\frac{\sigma_a(\sigma_c-i\varepsilon_0\omega)}{\sigma_a\alpha^2+(\sigma_c-i\varepsilon_0\omega)}. \qquad (2)$$

Two simple conclusions can be immediately drawn from this expression. If the real parts of the conductivities (σ_{1a},σ_{1c}) are only weakly frequency dependent, then (i) the effective conductivity should reveal a peak if $\mathrm{Im}[\sigma_a\alpha^2+(\sigma_c-i\varepsilon_0\omega_1)]=0$, and (ii) the inverse conductivity should show a peak (i.e., longitudinal resonance) if $\mathrm{Im}[\sigma_c-i\varepsilon_0\omega_0]=0$. Assuming that only the high-frequency dielectric constant ($-i\varepsilon_0\varepsilon_{c,\infty}\omega$) and the superconducting condensate ($in_se^2/m_c\omega$) contribute to the low-temperature c-axis conductivity, the critical frequency in case (ii) can be estimated $\omega_0^2=n_se^2/[m_c(\epsilon_\infty+1)]$, which closely resembles the expression for the screened plasma frequency. Evidently, the expression for ω_0^2 does not depend upon the tilt angle. On the contrary, a substantial angular dependence is expected for the resonance at ω_1. The change of the tilt angle may be easily achieved experimentally by rotating the polarization of the incident radiation. In this case, the effective tilt angle may be obtained as $\alpha_{\text{eff}}=\alpha_0\sin\varphi$, where $\varphi=0°$ corresponds to the polarization within the ab plane.

Using Eq. (1), it is possible to calculate the c-axis properties of the $Nd_{1.85}Ce_{0.15}CuO_{4-\delta}$ film. The dielectric function along the c axis, $\varepsilon_c^*=i\sigma_c/\omega\varepsilon_0$, is presented in Fig. 3. The data for all polarizations φ collapse into a single curve, thereby supporting the applied concept. It should be noted that the uncertainty in determination of α_0 represents the most relevant source of errors in the determination of ε_c^*. However, the variation of this angle influences only the absolute values of the conductivity and leaves the overall frequency dependence unchanged.

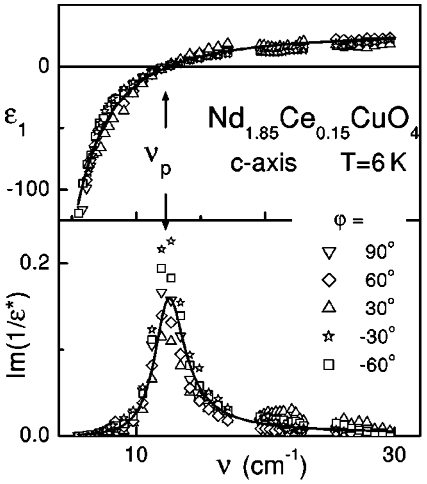

FIG. 3. Dielectric function of a $Nd_{1.85}Ce_{0.15}CuO_{4-\delta}$ film along the c-axis. Upper panel: ε_1. Lower panel: Imaginary part of the loss function $\mathrm{Im}(1/\varepsilon^*)$. Lines are calculated within the following assumptions: (i) σ_{1c} is frequency independent and (ii) only high-frequency dielectric constant ε_∞ and superonducting condensate $-(\lambda_c c/\omega)^2$ contribute to ε_1.

As demonstrated in Fig. 3, the high-frequency ($\nu > 20\,\mathrm{cm}^{-1}$) c-axis dielectric constant of $Nd_{1.85}Ce_{0.15}CuO_{4-\delta}$ is positive, which can be ascribed to the phonon contribution. A superconducting response becomes dominant for $\nu < 15\,\mathrm{cm}^{-1}$, from which the low-frequency penetration depth may be estimated as $\lambda_c(6\,\mathrm{K}) = 24 \pm 8\,\mu$. From the zero crossing of the dielectric constant, the screened plasma frequency can be extracted ($\nu_p = 12\,\mathrm{cm}^{-1}$). This value is in excellent agreement with the powder data of Shibata and Yamada[9] and with the recent results of Singley *et al.*[15]

In conclusion, the anisotropic properties of a series of slightly tilted $Nd_{1.85}Ce_{0.15}CuO_{4-\delta}$ thin films were measured in the submillimeter frequency range. Two new features were observed in the low-temperature transmission spectra which showed different polarization dependencies. The antiresonance at $\nu = 12\,\mathrm{cm}^{-1}$ corresponds to the c-axis plasma frequency of $Nd_{1.85}Ce_{0.15}CuO_{4-\delta}$. The second resonance represents a mixed ab-plane/c-axis excitation. The frequency of this resonance can be changed in a controllable way by rotating the polarization of the incident radiation.

The authors acknowledge stimulating discussions with R. Hackl, W. N. Hardy, and T. Timusk. This work was supported by BMBF (Contract No. 13N6917/0-EKM).

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