Observation of the Transverse Optical Plasmon in SmLa$_{0.8}$Sr$_{0.2}$CuO$_{4-\delta}$

Diana Dulić$^1$, A. Pimenov$^2$, D. van der Marel$^1$, D. M. Broun$^{3,4}$, Saeid Kamali$^2$, W. N. Hardy$^3$, A. A. Tsvetkov$^1$, I.M. Sutjaja$^5$, Ruixing Liang$^3$, A. A. Menovsky$^3$, A. Loidl$^2$ and S. S. Saxena$^1$

$^1$Laboratory of Solid State Physics, Materials Science Centre, Nijenborgh 4, 9747 AG Groningen, The Netherlands
$^2$Experimentalphysik V, University of Augsburg, Universitätsstr. 2, 86135 Augsburg, Germany
$^3$Department of Physics and Astronomy, University of British Columbia, Vancouver, BC, V6T 1Z1, Canada
$^4$Interdisciplinary Research Centre in Superconductivity and Department of Physics, Cavendish Laboratory, University of Cambridge, Madingley Road, Cambridge CB3 0HE, United Kingdom
$^5$Van der Waals - Zeeman Institut, University of Amsterdam, The Netherlands

We present microwave and infrared measurements on SmLa$_{0.8}$Sr$_{0.2}$CuO$_{4-\delta}$, which are direct evidence for the existence of a transverse optical plasma mode, observed as a peak in the c-axis optical conductivity. This mode appears as a consequence of the existence of two different intrinsic Josephson couplings between the CuO$_2$ layers, one with a Sm$_2$O$_3$ block layer, and the other one with a (La,Sr)$_2$O$_{2-\delta}$ block layer. From the frequencies and the intensities of the collective modes we determine the value of the compressibility of the two dimensional electron fluid in the copper oxygen planes.

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In 1966 A.J. Leggett predicted for superconductors with two bands of charge carriers a collective oscillation corresponding to small fluctuations of the relative phases of the two condensates, briefly indicated as excitons below the superconducting gap $\Delta$. In principle these excitons should be observable with electromagnetic radiation, but in practice the effect on the infrared optical properties of most superconducting materials has been too small to be observable, except for, as we will demonstrate in the present paper, the bi-layer cuprate superconductors. The cuprate high temperature superconductors naturally form weakly coupled stacks of superconducting layers $\parallel$. Some members of this family, e.g. Bi$_2$Sr$_2$CaCu$_2$O$_8$, have two superconducting layers per unit cell. These materials are realizations of a two-band superconductor, satisfying the following unique conditions: (i) For polarization of the electric field perpendicular to the conducting planes the metallic screening is very weak due to strong anisotropy of the static and dynamical electrical conductivity. (ii) The dipole selection rules allow optical transitions which resonate at the Josephson exciton energy.

In Ref. $\parallel$ two of us (DvdM and AAT) calculated the dielectric function for cuprate superconductors with two CuO$_2$ planes per unit cell, using the Lawrence-Doniach model $\parallel$ with alternating coupling constants (the 'multilayer model'). A direct consequence was the presence of a transverse optical plasma mode, polarized perpendicular to the planes for propagation along the planes. Similar to a transverse optical phonon, and in contrast to the transverse Josephson plasma resonance (JPR) in single layer cuprates, this mode is observable as an optically allowed absorption in measurements of the optical conductivity. In Fig. 1 the currents and charge oscillations of these modes are sketched. Recently the unusual 'bump' in the c-axis conductivity of YBCO $\parallel$ and Bi$_2$2212 $\parallel$, has been attributed to such a mode. However, due to the close proximity to the optical phonons it was not possible to separate these electronic collective modes from the optical lattice vibrations $\parallel$, which complicates the quantitative analysis of this interesting phenomenon.

Both in the two-band exciton model and in the multilayer model the collective modes are oscillations of the relative phase of the two condensates, the inertia of which is due to the finite Josephson coupling between the two condensates. In Ref. $\parallel$ the restoring force was provided by the fact, that if $\delta N$ electrons are added to a plane, the free energy increases with an amount $\delta F = \mu \delta N + \delta N^2/(2Kn^2)$. Here $\mu$, $K$, and $n$ are the chemical potential, the electronic compressibility, and the electron density respectively. For a Fermi-liquid $Kn^2 = \partial N/\partial \mu$ corresponds to density of states at the Fermi level. In neutral fluids the compressibility causes propagation of sound, whereas for electrons it causes the dispersion of plasmons. On the other hand, in Ref. $\parallel$ the restoring force is provided by the long range Coulomb interaction between each set of planes. Recently this model

![FIG. 1. Snapshot of the currents (arrows) and charges (± signs) of the transverse optical JPR. This mode carries a finite net polarization transverse to the direction of propagation.](image-url)
was extended by taking into account the compressibility

Dispersal of the JPR in the cuprates due to the compressibility term has been previously described by Koyama and Tachiki.

SmLa$_{0.8}$Sr$_{0.2}$CuO$_{4-\delta}$ forms the $T^*$ structure, with superconducting planes alternatingly separated by two types of blocking layers, a flourite-type layer of Sm$_2$O$_2$ ($T^*$ type) and a rocksalt-type (La,Sr)$_2$O$_2$-$\delta$ ($T$ type) block layer. The lattice parameter is 12.57 Å, and the distances between CuO$_2$ layers across the SmO and the LaO barriers are 6.13 Å, and 6.43 Å respectively. Because the coupling across both blocking layers is weak, the Josephson plasma resonances are well below the optical phonon frequencies, allowing a clean separation between the lattice vibrations and the electronic modes. Recently two longitudinal Josephson plasma resonances have been observed in embedded powder samples of this material using the far-infrared sphere resonance method, but the transverse optical plasma mode could not be detected.

Here we present infrared optical spectra of single crystal SmLa$_{0.8}$Sr$_{0.2}$CuO$_{4-\delta}$. We observed the transverse optical plasma mode and the two longitudinal Josephson plasma resonances. The influence of the compressibility on the intensity and the frequency of the observed resonances is large, a characteristic which these collective modes have in common with the two-band excitons predicted in 1966. Using a quantitative analysis of the intensities and peak positions of the three collective modes we extract the values of the intrinsic Josephson coupling constants and the compressibility of the two dimensional charge fluid in the planes.

The single crystals of SmLa$_{0.8}$Sr$_{0.2}$CuO$_{4-\delta}$ were grown at the University of Amsterdam using a four mirror image furnace. They were annealed at 900 °C for 24 hours to homogenize the strontium concentration. The crystal was cut into several slabs for the various experiments, and then annealed in a high pressure oxygen furnace at the University of British Columbia. The samples were slow cooled (one °C per hour) from 500°C to 350°C at 80 bars oxygen pressure, held at 350°C for 48 hours and then quenched by opening the furnace. The large crystal faces (containing the c-axis) were polished to an optical finish using diamond polishing pads. The magnetization, as measured in a field of 2 Oe with $H$ parallel to $c$, gave a midpoint $T_c$ of about 16K with a transition width of ±2K.

Three types of experiments were performed: (a) polarized reflection measurements using a Fourier transform spectrometer in the frequency range between 12 and 1000 cm$^{-1}$, and the temperature range from 4 to 300K; \textit{in-situ} evaporated gold films were used as a reference; (b) polarized transmission measurements in the frequency range from 4 to 20 cm$^{-1}$ using a Mach-Zehnder interferometer arrangement, which allows measurements of both transmission and phase shift (from the Fresnel formulae and the relation between the dielectric function and optical conductivity: $\epsilon = 4\pi i\sigma/\omega$). The real part of the optical conductivity, the real part of the dielectric function and the loss function $Im(-1/\epsilon)$ are shown in Fig. (a), (b), and (c), respectively.

In Ref. the dielectric function has been derived for a material with two superconducting layers of infinitesimal thickness, with two different Josephson coupling constants $J$ and $K$, distances between the neighboring planes $x_Jd$ and $x_Kd$, $x_J + x_K = 1$, and $d$ is the lattice constant along the $c$-direction. The total dielectric function was shown to be of the form

$$\frac{\epsilon^2}{\epsilon(\omega)} = \frac{\tilde{\varepsilon}_J\omega^2}{\omega(\omega+i\gamma_J) - \omega_J^2} + \frac{\tilde{\varepsilon}_K\omega^2}{\omega(\omega+i\gamma_K) - \omega_K^2}, \quad (1)$$

Here the average background dielectric constant is $\epsilon_{av}^2 = (x_J/\epsilon_J^* + x_K/\epsilon_K^*)^{-1}$, and $\epsilon_J^*$, $\omega_J$, and $\gamma_J$ are the local

![FIG. 2. The c-axis reflectivity of SmLa$_{0.8}$Sr$_{0.2}$CuO$_{4-\delta}$: 4K-solid line, 30K-dotted line. Inset: blown up region from 11 to 50cm.$^{-1}$](image-url)
background dielectric constants the screened Josephson plasma frequencies and the quasi-particle damping characterizing the two types of junctions. \( \text{Re} (\omega) \) has two zero crossings corresponding to the two longitudinal plasmons, and a pole at the transverse optical plasma frequency. The transverse and the longitudinal plasma resonances show up as a peaks in the optical conductivity and the loss function respectively. The transverse optical plasma frequency \( \omega_T = (\varepsilon_r \omega_K^2 + \tilde{z}_T \omega_T^2)^{1/2} \), and its inverse lifetime \( \gamma_T = \tilde{z}_T \gamma_K + \tilde{z}_T \gamma_T \) are fixed by the values of \( \omega_T, \omega_K, \gamma_T, \gamma_K \), and the weight factors \( \tilde{z}_K = 1 - \tilde{z}_I \), which are a function of the relative volume fractions, the local dielectric functions, and the compressibility which we will discuss below.

The two longitudinal plasma frequencies can be read directly from the peak positions in the loss function, providing \( \omega_I/2\pi c = 7 \text{cm}^{-1} \) and \( \omega_K/2\pi c = 12.6 \text{cm}^{-1} \). The background dielectric constant is obtained by analyzing the reflectivity spectra of Fig. 3, providing \( \varepsilon_n^\prime = 23.0 \pm 0.5 \). The intensities of the two peaks in the loss function, or, more accurately, the intensity of the peak in \( \sigma \) at \( \omega_T \), provide the weight factor \( \tilde{z}_K: \int \sigma_{\text{peak}} (\omega) d\omega = \tilde{z}_K \varepsilon_n^\prime (\omega_K^2 - \omega^2) / \omega_T^2. \) This way we obtain \( \tilde{z}_K = 0.04 \pm 0.005 \), and \( \tilde{z}_I = 0.96 \pm 0.005 \). In Fig. 4 we show the theoretical spectra of the dielectric function, and the loss-function using these parameters together with the experimental spectra at 4 K. The damping factors were adjusted to the experimental linewidth of the loss-peaks, \( \gamma_I/2\pi c = \gamma_K/2\pi c = 0.5 \text{cm}^{-1}. \)

From the microwave data we have obtained an absolute measurement [16] of the c-axis penetration depth, which is 45 \( \mu \text{m} \) at 3K. The penetration depth is \( \lambda_c = \lim_{\omega \rightarrow 0} \epsilon^\prime (\omega) / (\omega \epsilon^\prime (\omega)) \), which in the framework of Eq. 4 is \( \varepsilon_n^\prime \lambda_c^2 / c^2 = \tilde{z}_I \omega^2 / \tilde{z}_T \omega_K^2. \) With the experimental values for \( \tilde{z}_I, \omega_T \), and \( \omega_K \) determined above, this provides \( \lambda_c = 47 \mu \text{m} \), in excellent agreement with the microwave results. In Fig. 5 the separate temperature dependencies of \( \lambda_c, \lambda_c, \) and \( \lambda_c, \) are given in the normalized form \( \lambda_c^2 (3K) / \lambda_c^2 (T) \); \( \lambda_c \) is obtained from a 1 GHz cavity perturbation measurement, and \( \lambda_c, = c / (\sqrt{\epsilon T \omega}) \), \( \lambda_c, = c / (\epsilon \omega K \omega K) \) from the transmission measurement. As seen from Fig. 5, both \( \lambda_c, \) and \( \lambda_c, \) exhibit similar, characteristic, power law temperature dependence \( \lambda_c^2 (3K) / \lambda_c^2 (T) = 1 - T^\gamma, \gamma \sim 3 \). This establishes that both resonances are indeed Josephson plasmons.

The intensity of the peaks, represented by the weight factors, \( \tilde{z}_K = 0.04 \) and \( \tilde{z}_I = 0.96 \), can be used to estimate the value of the compressibility of the 2D electron fluids, using the expressions derived in [16]. The dielectric function is of the form given in Eq. 4 while \( \tilde{z}_K \) can be calculated from the experimental peak positions and linewidths \( \omega_K, \omega_I, \gamma_I \), and \( \gamma_K \) using

\[ \tilde{z}_K = \frac{1}{2} + \frac{z_K (z_I + z_T)}{2z_K z_I + z_T} \left( 1 - \frac{4(2\gamma)^2 \omega_K^2 \omega_T^2}{z_K (z_I + z_T) + z_T} \right) \]

(2)

Here \( \gamma \) is a dimensionless constant which is inversely proportional to the two-dimensional compressibility of the electron fluids in the planes, \( \gamma = \varepsilon_n^\prime / (4\pi c e^2 K n^2). \) The constants \( z_K \) and \( z_I \), defined as \( z_K = x_K \varepsilon_n^\prime / \varepsilon_K \) and \( z_I = \varepsilon_n^\prime / \varepsilon_I \) correspond to the weight factors in the limit of infinite compressibility (\( \gamma \rightarrow 0 \)). We can calculate them easily from the known volume fractions \( x_K \) and \( x_I \), (0.5 in the present case) and from the ratio between the \( T \) -phase and the \( T' \) -phase c-axis dielectric constants, \( \varepsilon_K^\prime / \varepsilon_I^\prime \). From infrared spectra of \( T \) -phase and the \( T' \) -phase materials [12], this ratio is known to be in the interval \( 1 \leq \varepsilon_K^\prime / \varepsilon_I^\prime \leq 1.7 \). The (numerical) inversion of Eq. 4 then provides \( K n^2 = 1.12 \pm 0.01 \text{eV}^{-1} \text{unit of CuO}_2 \). For \( \text{La}_{2-x} \text{Sr}_x \text{CuO}_4-\delta \) the linear term in the normal state specific heat [14] ranges from 0.4 mJ/g.at.K² (\( x=0.1 \)) to 1.2 mJ/g.at.K² (\( x=0.2 \)). For fermionic exci-
tations this would imply that the density of states varies from $1.2 \text{ eV}^{-1}$ ($x=0.1$) to $5.0 \text{ eV}^{-1}$ per unit of CuO$_2$ ($x=0.2$). The value of $Kn^2 = 1.1\text{eV}^{-1}$ should then be taken as an indication, that our SmLaSrCuO crystals are underdoped. This is also indicated by the $T_c$ of 16 K, and the low values of the JPR-frequencies.

Without the compressibility term, the JPR’s would have been at $6.1 \text{ cm}^{-1}$ and $8.0 \text{ cm}^{-1}$ [7]. Hence the finite electronic compressibility causes a large shift of resonance frequency and a large reduction of the intensity of the higher frequency JPR. This also explains why the 'bump' has a very weak intensity in Bi2212 [8].

In conclusion, we have studied microwave and infrared properties of SmLa$_{0.8}$Sr$_{0.2}$CuO$_{4-\delta}$. We have observed two longitudinal Josephson plasma resonances, and the transverse optical plasma mode that arises from the alternation of two different Josephson couplings between the CuO$_2$ layers. We observe a shift of frequency of the plasma modes, and a reduction in intensity of the highest mode due to the finite electronic compressibility corresponding to $1.1 \text{ eV}^{-1}$ per CuO$_2$ unit.

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FIG. 4. (a) Fit (solid line) to (a) the real part of the $c$-axis optical conductivity, (b) Real part of the $c$-axis dielectric function, (c) the loss function at 3K. The open circles are the experimental data.

FIG. 5. Temperature dependence of the $c$-axis penetration depth: open squares—measured in cavity perturbation experiment, triangles: $\lambda_{c,J}$, open circles: $\lambda_{c,K}$.

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