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## Anomalous millimeter-wave absorption in the superconducting phase of $La_{2-x}Sr_xCuO_4$

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Direct measurements of the complex conductivity spectra of thin-film  $La_{2-x}Sr_xCuO_4$  are made at frequencies of 5–40 cm<sup>-1</sup>. Narrow, intense Drude-type excitation is observed in the superconducting phase. © 1998 American Institute of Physics. [S0021-3640(98)01017-2]

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Measurements of the complex dynamic conductivity  $\sigma = \sigma_1 + i\sigma_2$  of hightemperature superconductors (HTSCs) yield important information about the nature of pairing, the quasiparticle density of states, and the charge-carrier scattering mechanisms.<sup>1,2</sup> In pure samples of the cuprate YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub>, which is the best-studied cuprate to date, the linear temperature dependence<sup>1,3</sup> of the penetration depth in the limit  $T \rightarrow 0$  and the intense absorption band in the millimeter (MM) and submillimeter (SBMM) conductivity spectra  $\sigma_1$  at temperatures  $T < T_c$ ,<sup>4,5</sup> which leads to a peak in the temperature dependence  $\sigma_1(T)$ ,<sup>4,6,7</sup> are currently viewed as evidence of *d*-type pairing or, at least, the presence of zeros of the order parameter on the Fermi surface.<sup>8-10</sup> In this connection, the problem of expanding the experimental data for other HTSCs is topical. In the present work, we performed measurements of the complex conductivity spectra of another superconducting cuprate La<sub>2-x</sub>Sr<sub>x</sub>CuO<sub>4</sub> in the millimeter and submillimeter ranges (5-40 cm<sup>-1</sup>).

We investigated a high-quality La<sub>1.84</sub>Sr<sub>0.16</sub>CuO<sub>4</sub> (LSCO) film 59 nm thick, deposited by molecular epitaxy on a plane-parallel, isotropic SrLaAlO<sub>4</sub> substrate approximately 1 mm thick. The *c* axis of the film was oriented perpendicular to the plane of the substrate. The method of synthesis and the structure and electrophysical properties of the film are described in Ref. 11. The superconducting transition temperature  $T_c$ =38.5 K, with a transition width of 1.5 K, which is comparable to the parameters of the best bulk samples.

The energy transmittance spectra  $Tr(\nu)$  and the phase spectra  $\varphi(\nu)$  of a wave transmitted through a film–substrate sandwich were measured on a laboratory BWT spectrometer (which utilizes a backward wave tube as the radiation source).<sup>12</sup> The optical constants of the film were calculated directly from these spectra (without using the

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FIG. 1. Spectra of the real (a) and imaginary (b) parts of the conductivity of LSCO in the normal and superconducting phases. Solid lines — least-squares result for the model (1) described in the text.

Kramers–Kronig relations) using general formulas for the complex transmittance of a bilayer system.<sup>13</sup> The optical parameters of the substrate were measured in advance. A detailed description of this method for measuring the MM–SBMM spectra of superconducting films on dielectric substrates is given in Ref. 14.

Figure 1 displays the wide-range  $(5-400 \text{ cm}^{-1})$  spectra of the real  $\sigma_1(\nu)$  and imaginary  $\sigma_2(\nu)$  parts of the complex conductivity of LSCO. To complete the picture, the original MM–SBMM data  $(5-40 \text{ cm}^{-1})$  are supplemented by higher frequency IR spectra  $(50-400 \text{ cm}^{-1})$ , obtained from measurements of the reflectance of a LSCO single crystal.<sup>15</sup> The well-known decrease<sup>2,16</sup> in the real part of the conductivity with decreasing temperature in the superconducting (SC) phase is observed above 30 cm<sup>-1</sup>.

The main result of the present work is the observation of an intense absorption band in the  $\sigma_1$  spectra in the superconducting phase of LSCO in the MM–SBMM region and is demonstrated in Fig. 1a. As the temperature decreases from 50 to 5 K the MM–SBMM conductivity increases severalfold; the dispersion-free behavior of  $\sigma_1$  in the normal phase is replaced at temperatures  $T < T_c$  by Drude-type dispersion in the spectra<sup>17</sup> —  $\sigma_1$  decreases as the frequency increases. This absorption band becomes narrower and more intense as the temperature decreases. The temperature evolution of the band leads to the appearance of a wide maximum in the dependence  $\sigma_1(T)$  below  $T_c$  (Fig. 2).

We note that the MM–SBMM absorption band observed in the superconducting phase of LSCO (like the analogous band in YBCO at these frequencies<sup>4</sup>) agrees qualitatively with models in which the energy gap has zeroes on the Fermi surface; these give rise to a narrow absorption peak at low frequencies.<sup>9,10</sup>

To obtain quantitative estimates of the parameters of the observed absorption band, the experimental spectra were analyzed using a phenomenological model for the complex conductivity.<sup>6</sup> To describe the MM–SBMM section of the spectrum we used the Drude



FIG. 2. Temperature dependence of the real part of the conductivity of LSCO for three fixed frequencies.

model and the  $\delta$  function at zero frequency in the spectrum of  $\sigma_1(\nu)$ , which is responsible for dc superconductivity (with allowance for its contribution to  $\sigma_2(\nu) \propto 1/\nu$ ), and the absorption band in the IR region was modeled by a Lorentzian whose parameters were found to be practically temperature-independent:

$$\sigma^*(\omega) = \sigma_1(\omega) + i\sigma_2(\omega) = \frac{n_n e^2}{m} \frac{1}{(\gamma - i\omega)} + \frac{n_s e^2}{m} \left[\frac{\pi}{2}\delta(0) + i\frac{1}{\omega}\right] + \text{``IR Lorentzian.''}$$
(1)

Here  $n_s$  and  $n_n$  are the densities of the paired and unpaired carriers, respectively, and  $\gamma$ is the relaxation rate of the unpaired carriers. The spectra were analyzed by the leastsquares method under the condition that the following sum rule is satisfied:  $n_s + n_n$  $= n_0 = \text{const}(T).^{18}$ 

The presence of substantial dispersion in the MM–SBMM spectra of  $\sigma_1$  and  $\sigma_2$  in the superconducting phase made it possible to determine positively the temperature dependence of the relaxation rate  $\gamma$  (Fig. 3) and to estimate the plasma frequencies of the periodice of the relaxation rate  $\gamma$  (Fig. 5) and to estimate the plasma frequencies of the paired,  $(\omega_{pl}^s)^2 = 4 \pi n_s e^2/m$ , and unpaired,  $(\omega_{pl}^n)^2 = 4 \pi n_n e^2/m$ , electrons. At T = 5 K one has  $\omega_{pl}^s/2\pi = 3900 \pm 800$  cm<sup>-1</sup> and  $\omega_{pl}^n/2\pi = 7800 \pm 1500$  cm<sup>-1</sup>. Hence we find for the "total" plasma frequency  $\omega_{pl}^0/2\pi = \sqrt{(\omega_{pl}^s)^2 + (\omega_{pl}^n)^2}/2\pi = 8700 \pm 1700$  cm<sup>-1</sup>, which agrees with the estimate<sup>16</sup> obtained from IR measurements on LSCO:  $\omega_{pl}^0/2\pi$ = 6300 cm<sup>-1</sup>. For the individual densities of the paired  $(x_s = n_s/n_0)$  and unpaired  $(x_n = n_s/n_0)$  $=n_n/n_0$ ) electrons we obtain  $x_s = (\omega_{pl}^s/\omega_{pl}^0)^2 \approx 20\%$  and  $x_n = (\omega_{pl}^n/\omega_{pl}^0)^2 \approx 80\%$ , i.e., even at the lowest temperatures most of the electrons in LSCO remain unpaired. For the London penetration depth we have  $\lambda_L = c/\omega_{pl}^s = 0.4 \ \mu\text{m}$ , which agrees well with the data from kinetic (0.4  $\mu$ m),<sup>19</sup> microwave (0.4  $\mu$ m),<sup>20</sup> IR (0.43  $\mu$ m),<sup>15</sup> and  $\mu$ SR (0.3  $\mu$ m)<sup>21</sup> measurements.

In summary, we measured the complex conductivity spectra of  $La_{2-x}Sr_xCuO_4$  in the millimeter and submillimeter regions of the spectrum  $(5-40 \text{ cm}^{-1})$ . Strong anomalous absorption in the superconducting phase was observed in the spectra of the real part of



FIG. 3. Temperature dependence of the relaxation rate  $\gamma$  obtained by analyzing the spectra of  $\sigma_1$  and  $\sigma_2$ . The large decrease in  $\gamma$  in the superconducting phase is responsible for the wide maximum in the temperature dependence of  $\sigma_1$  (Fig. 2).

the conductivity at these frequencies. From the experimental data obtained we determined the plasma frequency of  $La_{2-x}Sr_xCuO_4$  and the temperature dependence of the relaxation rate of the unpaired carriers that give rise to the observed absorption.

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- <sup>1</sup>D. A. Bonn and W. N. Hardy, in *Physical Properties of High Temperature Superconductors*, edited by D. M. Ginsberg, World Scientific, Singapore, 1996, Vol. 5.
- <sup>2</sup>D. B. Tanner and T. Timusk, in *Physical Properties of High Temperature Superconductors*, edited by D. M. Ginsberg, World Scientific, Singapore, 1992, Vol. 3.
- <sup>3</sup>W. N. Hardy, D. A. Bonn, D. C. Morgan et al., Phys. Rev. Lett. **70**, 3999 (1993).
- <sup>4</sup>M. C. Nuss, P. M. Mankievich, M. L. O'Malley et al., Phys. Rev. Lett. 66, 3305 (1991); A. A. Volkov,
- B. P. Gorshunov, G. V. Kozlov et al., Zh. Eksp. Teor. Fiz. 95, 261 (1989) [Sov. Phys. JETP 68, 148 (1989)].
- <sup>5</sup>P. G. Quincey, P. B. Whibberley, and J. R. Birch, Solid State Commun. 76, 1281 (1991).
- <sup>6</sup>D. A. Bonn, P. Dosanjh, R. Liang, and W. N. Hardy, Phys. Rev. Lett. **68**, 2390 (1992); D. A. Bonn, R. Liang, T. M. Riseman *et al.*, Phys. Rev. B **47**, 11314 (1993).
- <sup>7</sup>A. Frenkel, F. Gao, Y. Liu et al., Phys. Rev. B 54, 1355 (1996).
- <sup>8</sup>D. J. Scalapino, Phys. Rep. 250, 329 (1995); K. Maki and H. Won, Ann. Phys. (Leipzig) 5, 320 (1996).
- <sup>9</sup>S. M. Quinlan, P. J. Hirschfeld, and D. J. Scalapino, Phys. Rev. B 53, 8575 (1996).
- <sup>10</sup> J. P. Carbotte, C. Jiang, D. N. Basov *et al.*, Phys. Rev. B **51**, 11798 (1995); H. Yamagata and H. Fukuyama, J. Phys. Soc. Jpn. **65**, 2204 (1996).
- <sup>11</sup> J.-P. Locquet, A. Catana, E. Machler *et al.*, Appl. Phys. Lett. **64**, 372 (1994); J.-P. Locquet and E. Machler, MRS Bull. **19**, 39 (1994).
- <sup>12</sup>A. A. Volkov, Yu. G. Goncharov, G. V. Kozlov *et al.*, Infrared Phys. **25**, 369 (1985); A. A. Volkov, G. V. Kozlov and A. M. Prokhorov, Infrared Phys. **29**, 747 (1989).
- <sup>13</sup> M. Born and E. Wolf, *Principles of Optics*, Pergamon Press, Oxford, 1980, 4th edition [Russian translation, Nauka, Moscow, 1970].
- <sup>14</sup>B. P. Gorshunov, G. V. Kozlov, A. A. Volkov et al., Int. J. Infrared Millim. Waves 14, 683 (1993).
- <sup>15</sup>H. S. Somal, B. J. Feenstra, J. Schutzmann et al., Phys. Rev. Lett. 76, 1525 (1996).
- <sup>16</sup>F. Gao, D. B. Romero, D. B. Tanner et al., Phys. Rev. B 47, 1036 (1993).
- <sup>17</sup>A. V. Sokolov, *Optical Properties of Metals*, American Elsevier, New York, 1967 [Russian translation, Fizmatgiz, Moscow, 1961].
- <sup>18</sup>R. A. Ferrel and R. E. Glover, Phys. Rev. **109**, 1398 (1958).
- <sup>19</sup>J.-P. Locquet, Y. Jaccard, A. Cretton et al., Phys. Rev. B 54, 7481 (1996).

## 436 JETP Lett., Vol. 68, No. 5, 10 Sept. 1998

Pronin et al.

<sup>20</sup>T. Scibauchi, H. Kitano, K. Uchinokura *et al.*, Phys. Rev. Lett. **72**, 2263 (1994).
<sup>21</sup>Y. J. Uemura, G. M. Luke, B. J. Sternleib *et al.*, Phys. Rev. Lett. **62**, 2317 (1989).

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