Evidence of two species of carriers from the far-infrared reflectivity of $Bi_2Sr_2CuO_6$

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An optical ''pseudogap'' is often assumed to open at low T in the ''anomalous Drude'' absorption, which models the optical conductivity $\sigma(\omega)\propto\omega^{-1}$ of high-T_c superconductors by a linewidth $\Gamma\approx 10^3$ cm^{-1} varying with ω. In the $\sigma(\omega)$ of $Bi_2Sr_2CuO_6$ measured down to 10 cm^{-1}, we have resolved, instead, two components separated by a deep minimum: (i) a normal Drude term with $\Gamma=35$ cm^{-1} at 30 K, in very good agreement with transport data; (ii) a strong band peaked in the far infrared (FIR), likely due to bound charges, whose tail exhibits the ω^{-1} dependence. As the FIR peak softens for $T\rightarrow 0$, it opens a pseudogaplike depression in $\sigma(\omega)$ accordingly to ordinary sum rules.

Several studies have been published during recent years on the possible implications for high-T_c superconductivity of the ''pseudogap.'' This term indicates a depression in the low-energy continuum of states of many high-T_c superconductors (HCTS's), that has been observed by different techniques below a characteristic temperature $T^*\gg T_c$.[1] The pseudogap has been observed also in the optical spectra of a number of metallic cuprates. These include underdoped systems like $Bi_2Sr_2CuO_6$ with $T_c=8$ K,[2] and $HgBa_2Ca_2Cu_3O_{8+y}$ with $T_c=121$ K,[3] overdoped cuprates like $La_{2-x}Sr_xCuO_4$ with $x>0.18$,[4] and $Tl_2Ba_2CuO_{6+y}$,[5] and even metals close to optimum doping like $HgBa_2Ca_2Cu_3O_{8+y}$ with $T_c=130$ K,[3] and $Bi_2Sr_2CaCu_2O_{8+y}$ with $T_c=90$ K.[6] In these experiments, the free-carrier contribution to the real part $\sigma(\omega)$ of the optical conductivity is described by an ''anomalous Drude'' term[1]

$$\sigma(\omega)=\frac{\omega_D^2/4\pi}{\Gamma^*(\omega)+[m^*(\omega)/m]^2\omega^2/\Gamma^*(\omega)}, \qquad (1)$$

where ω_D is a constant plasma frequency, while both the scattering rate Γ^* and the effective mass m^* depend on the photon energy ω. This model has been originally introduced to fit the $\sigma(\omega)\propto\omega^{-1}$ law which replaces the ω^{-2} behavior of conventional metals in the mid-infrared spectra of HCTS's. The use of Eq. (1) leads to large scattering rates for the carriers in the cuprates. At $T\sim T^*$, $\Gamma^*\approx 1500$ cm^{-1} in the far infrared, for both $La_{2-x}Sr_xCuO_4$ and $HgBa_2Ca_2Cu_3O_{8+y}$. Below T^*, Γ^* decreases to ~ 500 cm^{-1} in the former compound, to ~ 1000 cm^{-1} in the latter.[3,4] Thus, even if the opening of an optical pseudogap below T^* is displayed by $\sigma(\omega)$, its amplitude is generally extracted from the above drop in the far-infrared (FIR) part of $\Gamma^*(\omega)$. The idea of an optical pseudogap is therefore intrinsically related to the anomalous Drude approach of Eq. (1) and to the assumption that the mid-infrared absorption of cuprates is dominated by some anomalous Fermi liquid.

However, the above picture leads to some inconsistencies. For instance, in $Bi_2Sr_2CaCu_2O_{8+y}$ samples with similar T_c's, the pseudogap determined optically (~ 700 cm^{-1}) (Ref. 6) is much larger than that found in angle-resolved photoemission (0–260 cm^{-1}, depending on the direction in the k space).[7] This discrepancy can hardly be explained, if one also considers that the optical absorption results from an average on the Fermi surface. Moreover, an optical pseudogap is observed in $La_{2-x}Sr_xCuO_4$,[4] although no such effect appears in the spin-lattice relaxation rate of this cuprate.[1] Finally, in the metallic spectra of Refs. 3–5 $\sigma(\omega)$ surprisingly decreases for $\omega\rightarrow 0$, for any T of the normal phase. This decrease is observed on the low-frequency side of a strong FIR peak *which is systematically associated with the observation of the pseudogap*. However, the FIR peak is attributed[3,4] to scattering of the free carriers by oxygen atoms randomly distributed in the Cu-O planes. As this explanation does not seem to be related to the existing theoretical models of the pseudogap,[1] the link between this latter and the FIR peak remains obscure.

A first hint comes from the optical conductivity of $Nd_{1.88}Ce_{0.12}CuO_4$ (NCCO), partially reported in Ref. 8. A peak develops in the infrared, which softens considerably as T is lowered. This shift causes at higher energies a depression in $\sigma(\omega)$ around 1000 cm^{-1}, very similar to the pseudogap reported by the authors cited above. This suggests that, in NCCO, the optical pseudogap opens by a transfer of spectral weight to the FIR peak as this softens and narrows, as requested by ordinary sum rules. One should then focus on this peak at finite frequency, attributed in NCCO to charges bound to the lattice via a polaronic coupling.[8] However, $Nd_{1.88}Ce_{0.12}CuO_4$ is electron doped and semiconducting, even if very close to the insulator-to-metal transition. In order to study in greater detail a system where the ''anomalous-Drude + pseudogap'' model has already been applied[2] and with the lowest carrier density, we have measured the reflectivity of $Bi_2Sr_2CuO_6$ (BSCO), from 400 to 8 K and from 15 000 to 10 cm^{-1}.

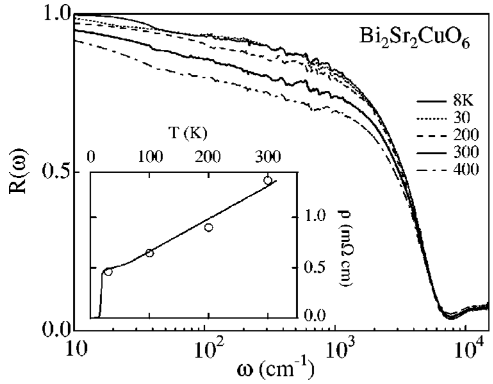

FIG. 1. Reflectivity $R(\omega)$ of the BSCO film at five temperatures from 400 to 8 K. The inset shows the resistivity $\rho(T)$ of the film from 300 to 4 K. The open circles are the ρ values obtained by extrapolating $\sigma(\omega,T)$ in Fig. 3 to $\omega=0$.

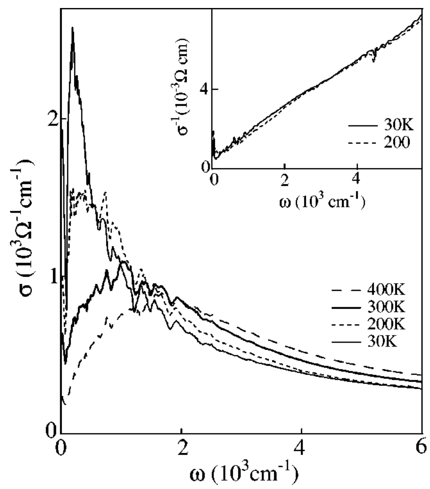

FIG. 2. Optical conductivity $\sigma(\omega)$ of the BSCO film, as extracted from $R(\omega)$ in Fig. 1 by Kramers-Kronig transformations. The inset shows that, on the high-frequency side of the FIR peak, $\sigma^{-1}(\omega)$ is linear with ω both at 200 K and 30 K.

By extending the measurements in BSCO to the very-far-infrared region of frequencies, we have first resolved from the FIR peak a normal Drude contribution which is in agreement with the dc conductivity σ_{dc} and the London penetration depth of the material. Basing on these results we show that, at least in BSCO: (i) $\sigma(\omega,T)$ cannot be explained in terms of a one-component model like the anomalous Drude of Eq. (1); (ii) the pseudogap does not open in the continuum of states of some kind of (anomalous) free carrier. On the contrary, we show that: (i) only the coexistence of two types of carriers may account for the optical absorption; (ii) the optical pseudogap is an effect created by the temperature-dependent absorption of some weakly bound charges. In such context, the phase-separation model proposed by Emery and Kivelson[9] is found to be in good agreement with the present observations.

The sample investigated here is a thick BSCO film, highly oriented with the *c* axis orthogonal to the surface. It has been grown by liquid phase epitaxy on a $LaGaO_3$ substrate.[10] Its resistivity $\rho(T)$, obtained from standard four-points measurements, is reported by a full line in the inset of Fig. 1. It shows a linear dependence on T from 300 to about 65 K, where a slight change of slope is observed. The superconducting transition has its onset at $T_c=20$ K, with a width of 5 K. The ρ value at 300 K (1.4×10^{-3} Ω cm) is intermediate between that of a good BSCO single crystal (0.3×10^{-3} Ω cm),[11] and that of a polycrystalline pellet of the same material (2.7×10^{-3} Ω cm).[12] This may be attributed to the presence of grain boundaries in the well oriented *a*-*b* plane of the present film. However, eventual grain boundaries are not expected to affect the infrared measurements. Indeed, the reflectivity $R(\omega)$ of the BSCO film, relative to a gold-plated reference and reported in Fig. 1, is typical of the best single crystals. The $R(\omega)$ spectra, with the electric field polarized in the *a*-*b* plane, were collected using a Bomem DA8 interferometer coupled to the infrared synchrotron radiation beamline SIRLOIN of the LURE laboratory at Orsay. Helium-cooled bolometers, mercury-cadmium-tellurium or silicon detectors were used, depending on the frequency range under investigation. The large size of the film (0.5×0.5 cm^2) and the good performances of the apparatus have allowed us to measure $R(\omega)$ down to unusually low values of ω (~10 cm^{-1}). The film thickness (1.8 μm) was such that no correction for the substrate contribution to $R(\omega)$ was needed; see Fig. 1. Therefore the optical conductivity $\sigma(\omega)$ was obtained by simple, canonical Kramers-Kronig transformations of $R(\omega)$. In the normal metallic phase ($T>T_c$), $R(\omega)$ has been extrapolated from $\omega=10$ cm^{-1} to $\omega=0$ by a Drude-Lorentz fit, in the superconducting phase by a London conductivity, as usually done; see Ref. 13. Moreover, $R(\omega)$ has been extrapolated from 15 000 cm^{-1} up to 320 000 cm^{-1} by using the data of Ref. 14 that have been extended to higher frequencies by a ω^{-4} law.

The conductivities $\sigma(\omega,T)$ corresponding to the raw reflectivities of Fig. 1 are shown in Fig. 2. At any T, $\sigma(\omega,T)$ exhibits a broad band peaked in the far infrared (FIR peak). Its high-frequency side behaves as ω^{-1} up to 6000 cm^{-1}, as shown in the inset of Fig. 2 for $T=200$ and 30 K. The tail of the FIR peak accounts therefore for the frequency dependence of the so-called anomalous-free-carrier absorption usually observed in metallic cuprates. The peak frequency (~800 cm^{-1} at 400 K) increasingly softens and narrows as T is lowered, until it reaches 110 cm^{-1} at 30 K. In the mid-infrared, a broad pseudogap opens for $T\rightarrow0$. This feature is less deep than that observed in other cuprates (see Refs. 3–5), most likely because the doping of the present BSCO sample is nearly optimal. This pseudogap is just due to the redshift and the narrowing of the FIR peak for decreasing T, which produce a transfer of spectral weight towards low frequencies. Indeed, the effective number of carriers per unit cell n_{eff} is constant with T within a few percent, for all temperatures of the normal phase, see the inset in Fig. 3. n_{eff} has been evaluated from the relation

$$n_{eff}=\frac{2m^*V}{\pi e^2}\int_{\omega_{min}}^{\omega_{max}}\sigma(\omega)\,d\omega, \qquad (2)$$

where V is the cell volume, $\omega_{min}=10$ cm^{-1}, $\omega_{max}=7500$ cm^{-1}, and m^* has been assumed equal to the free electron mass.

The results of Fig. 2 are quite similar to those reported in Ref. 2 for a single crystal of BSCO with $T_c=8$ K, whose

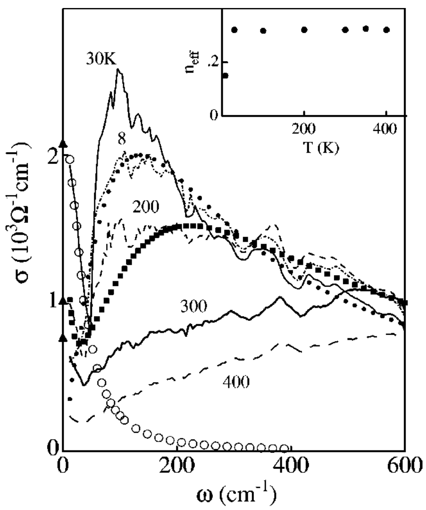

FIG. 3. Expansion in the FIR of the $\sigma(\omega)$ of Fig. 2. The open circles are the best fit to a normal Drude term, given by Eq. (3), here proposed only for the 30 K curve. The dots and the squares are best fits of Eqs. (4) and (5) to data at 8 and 200 K, respectively. The triangles on the ordinate axis represent the values of σ_{dc} measured at the same temperatures, also reported in the inset of Fig. 1. The inset shows the spectral weight calculated from Eq. (2) (see text).

$R(\omega)$ was measured from 12 000 to 50 cm^{-1}. However, as evident from Fig. 3 where the FIR $\sigma(\omega,T)$ has been reported on an expanded scale, the extension of the spectra down to 10 cm^{-1} has allowed us to resolve a narrow, low-frequency absorption from the FIR peak. The dip separating these two components of $\sigma(\omega)$ is observed at all temperatures. It corresponds to a change of slope at ~ 50 cm^{-1} in $R(\omega)$, see Fig. 1, where no instrumental effects are present, e.g., beam splitter transmittance minima or data file overlaps. The low-frequency component of the absorption increases for $\omega \to 0$, extrapolates to the $\sigma_{dc}(T)$ values measured in the same sample (full triangles), and disappears below T_c. The open circles in Fig. 3 show that this feature, presumably peaked at $\omega = 0$, is very well fitted at 30 K by a *normal Drude* term

$$\sigma(\omega) = \frac{\omega_D^2/4\pi}{\Gamma_D + \omega^2/\Gamma_D}, \tag{3}$$

with plasma frequency $\omega_D = 2100$ cm^{-1} and scattering rate $\Gamma_D = 35$ cm^{-1}. At 100 (200) K one obtains, instead, $\omega_D = 1800(1600)$ cm^{-1} and $\Gamma_D = 35(40)$ cm^{-1}. If one extrapolates the Drude fits to obtain $\sigma(0,T)$ at all temperatures, one obtains the values reported by open circles in the inset of Fig. 1 together with the dc resistivity (full line) measured in the same sample. The agreement is within a few percent.

The results reported in Figs. 2 and 3 point towards a multicomponent model for $\sigma(\omega)$ and indicate a coexistence of free and bound charges. The latter may be polaronic in nature and may aggregate at low temperatures, as proposed for the NCCO system in Ref. 8. Emery and Kivelson[9] have presented a picture where free carriers are scattered by arrays of (dynamical) bound charges which carry local dipoles. Following these authors, the optical conductivity can be written as

$$\sigma(\omega,T) = \sigma_a + \sigma_b = \frac{e^2 A}{\omega}\chi_2(\omega,T) + (e^*)^2 \omega \chi_2(\omega,T) \tag{4}$$

with

$$\chi_2(\omega,T) = c \tanh(\hbar\omega/2k_B T)\frac{\Gamma_p}{\Gamma_p^2 + \omega^2}. \tag{5}$$

In Eq. (4), σ_b is the contribution of the bound charges, modeled as c dipoles of charge e^*. σ_a takes into account the scattering of the free carriers of charge e by these dipoles. Γ_p determines both the FIR peak frequency and width, while A is a constant whose expression is reported in Ref. 9.

Equation (4) was compared unsuccessfully with the optical conductivity measured in La_2CuO_{4+y}.[15] On the contrary, good fits of Eq. (4) to the present BSCO data are obtained at all temperatures, as reported in Fig. 3 for $T > T_c$ (200 K) and $T < T_c$ (8 K). Remarkably, by using the two parameters c and Γ_p (whose values are 230 cm^{-1} at 200 K, 135 cm^{-1} at 8 K), one reproduces the Drude term, the asymmetric FIR peak, and the softening and narrowing of this latter for decreasing T.

Below T_c the Drude peak disappears from the measuring range and the FIR peak loses the intensity corresponding to the underlying Drude tail. This effect, which has been reproduced in the fit by fixing $\sigma_a(\omega, T = 8\ \mathrm{K}) = 0$, allows one to perform a further check of the present analysis. In fact, the loss of spectral weight observed below T_c in the inset of Fig. 3 provides an estimate of the London penetration depth in the film, as given by the Ferrell-Glover sum rule[16]

$$\lambda_L^2 = \frac{c^2}{8\int_{\omega_{min}}^{\omega_{max}} [\sigma_n(\omega,30K) - \sigma_s(\omega,8K)]\, d\omega}, \tag{6}$$

where here c is the speed of light and the other symbols have a obvious meaning. For $\omega_{min} = 10$ cm^{-1} and $\omega_{max} = 15\,000$ cm^{-1} one obtains $\lambda_L = 300 \pm 10$ nm, a value in excellent agreement with that obtained for $Bi_2Sr_2CuO_6$ from transport measurements (310 nm).[11] On the other hand, no estimate of the superconducting gap can be made, due to the strong FIR peak which overshadows the collapse of the Drude component below T_c.

In conclusion, the present far-infrared study of BSCO questions the widely accepted ''anomalous Drude + pseudogap'' model, through the identification in $\sigma(\omega,T)$ of *a narrow, normal Drude absorption well resolved from a broad FIR peak*. The small value of the free-carrier scattering rate ($\Gamma_D < 50$ cm^{-1}) is consistent with a metal in an extremely clean limit, where the carriers move in well ordered, nearly defect-free Cu-O planes. Comparable values of Γ_D can be obtained in doped semiconductors by the technique of ''modulation doping,'' namely, by spatially separating the impurities, which provide the free carriers, from the planes where these latters are free to move. In BSCO the Cu-O planes seem then to be very clean, while the eventual impurities and defects are mostly distributed out of plane.

The ω^{-1} dependence of $\sigma(\omega)$ in the mid infrared, often invoked to justify the need for an anomalous Drude model, is due to the tail of the peak centered at finite frequencies and should be ascribed to the bound charges. Similarly, the optical pseudogap (see Fig. 2) is a depression in $\sigma(\omega)$ created by the bound-charge absorption as this latter narrows and softens for decreasing T. This effect is required by the conservation of n_{eff} with temperature, here fulfilled within a few percent. In this respect, any optical determination of the temperature T^* where the pseudogap starts opening seems questionable, as typically the FIR peak is observed even at $T > T^*$.

The present observations strongly support the coexistence of free and weakly bound charges in BSCO, responsible for the normal Drude absorption and the FIR peak, respectively. This latter is very similar to the polaronic feature observed in semiconducting cuprates at low doping. Its softening for decreasing T, followed by a saturation below $T \sim 150$ K, has already been attributed in $Nd_{2-x}Ce_xCuO_4$ to the formation of polaronic aggregates. This approach is confirmed by the present observations in BSCO, which are remarkably well fitted by a phase-separation model based on the scattering of the free carriers by dynamical arrays of weakly bound charges.

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[1] For a review, see T. Timusk and B. Statt, Rep. Prog. Phys. **62**, 61 (1999).
[2] A. A. Tsvetkov, J. Schützmann, J. I. Gorina, G. A. Kaljushnaia, and D. Van der Marel, Phys. Rev. B **55**, 14 152 (1997).
[3] J. J. McGuire, M. Windt, T. Startseva, T. Timusk, D. Colson, and V. Viallet-Guillen (unpublished).
[4] T. Startseva, T. Timusk, M. Okuya, T. Kimura, and K. Kishio, Physica C **321**, 135 (1999).
[5] A. V. Puchkov, T. Timusk, S. Doyle, and A. M. Hermann, Phys. Rev. B **51**, 3312 (1995).
[6] A. V. Puchkov, D. N. Basov, and T. Timusk, J. Phys.: Condens. Matter **8**, 10 049 (1996).
[7] M. Randeria and J.-C. Campuzano, Proceedings of the International School of Physics ''Enrico Fermi,'' Course CXXXVI, 1997 (IOS Press, Amsterdam, 1998).
[8] S. Lupi, P. Maselli, M. Capizzi, P. Calvani, P. Giura, and P. Roy, Phys. Rev. Lett. **83**, 4852 (1999).
[9] V. J. Emery and S. A. Kivelson, Phys. Rev. Lett. **71**, 3701 (1993).
[10] G. Balestrino, M. Marinelli, E. Milani, A. Paoletti, and P. Paroli, J. Appl. Phys. **70**, 6939 (1991).
[11] S. Martin, A. T. Fiory, R. M. Fleming, L. F. Schneemeyer, and J. V. Waszczak, Phys. Rev. B **41**, 846 (1990).
[12] G. Xiao, M. Z. Cieplak, and C. L. Chien, Phys. Rev. B **38**, 11 824 (1988).
[13] M. A. Quijada, D. B. Tanner, R. J. Kelley, M. Onellion, H. Berger, and G. Margaritondo, Phys. Rev. B **60**, 14 917 (1999).
[14] I. Terasaki, S. Tajima, H. Eisaki, H. Takagi, K. Uchinokura, and S. Uchida, Phys. Rev. B **41**, 865 (1990).
[15] M. A. Quijada, D. B. Tanner, F. C. Chou, D. C. Johnston, and S-W. Cheong, Phys. Rev. B **52**, 15 485 (1995).
[16] M. Tinkham and R. A. Ferrell, Phys. Rev. Lett. **2**, 331 (1959).