Absence of a Loss of In-Plane Infrared Spectral Weight in the Pseudogap Regime of $Bi_2Sr_2CaCu_2O_{8+\delta}$

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The *ab*-plane reflectance of $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi-2212) thin films was measured in the $30-25\,000$ cm^{-1} range for one underdoped ($T_c = 70$ K), and one overdoped sample ($T_c = 63$ K) down to 10 K. We find similar behaviors in the temperature dependence of the normal-state infrared response of both samples. Above T_c, the effective spectral weight, obtained from the integrated conductivity, does not decrease when T decreases, so that no opening of an optical pseudogap is seen. We suggest that these are consequences of the pseudogap opening in the $\mathbf{k} = (0, \pi)$ direction and of the in-plane infrared conductivity being mostly sensitive to the $\mathbf{k} = (\pi, \pi)$ direction.

The normal state of high-T_c superconductors, in particular the pseudogap state, is highly debated [1]. A number of experiments in underdoped samples (nuclear magnetic resonance (NMR) [2–4], specific heat [5], uniform susceptibility [6]) show the opening of a gap in the density of states at some temperature $T^* > T_c$. Similarly, Raman spectroscopy points to a depletion of scattered intensity at low energy [7]. Nevertheless, the very existence of T^* is disputed [4]. Tunneling data exhibit a broad dip above T_c [8], over the same energy range of the superconducting gap. Angle-resolved photoemission spectroscopy (ARPES) measurements display a shift of the leading edge of the energy dispersion curve spectrum in the $(0, \pi)$ direction [9]. They also show that significant arcs of the Fermi surface are still present in the (π, π) direction down to T_c [10].

The in-plane dc resistivity in underdoped cuprates shows a deviation from the high temperature linear behavior. In Bi-2212, the temperature where this deviation occurs agrees with T^* as determined by other techniques (NMR, ARPES) [11,12]. The real part $\sigma_1(\omega, T)$ of the *ab*-plane infrared (IR) conductivity of underdoped samples exhibits a depression developing as T ($> T_c$) decreases within an energy range extending typically from 200 to 1500 cm^{-1} for the YBCO, Bi-2212, and Tl-2201 compounds [13]. This loss of spectral weight (SW) was assigned either to the pseudogap opening or to a shift of SW towards very low frequencies [13]. It is obviously difficult to discriminate between these two approaches due to experimental limitations in the low frequency part of the spectrum (usually restricted to $\omega \gtrsim 30$ cm^{-1}). A suppression of the optical scattering rate $1/\tau(\omega)$ in underdoped samples below a typical constant energy scale ~ 700 cm^{-1}, irrespective of temperature and doping, was suggested to be the infrared signature of the pseudogap state [13–15]. None of these IR data convincingly defines a T^* due to the small number of experimentally available temperatures [16].

Indeed, to the best of our knowledge, no systematic study of the detailed temperature dependence of the *in-plane* optical conductivity has been performed to date. In this work, we report such a study in Bi-2212 thin films for a set of 15 temperatures. Thanks to their highly reflective surface and their large area (typically $\geq 5 \times 5$ mm^2), thin films allow to resolve *reliably* relative changes in reflectance as small as 0.2%. This unprecedented accuracy allows us to look for the opening of a pseudogap by tracking a *transfer of spectral weight* from *low to high energies,* possibly occurring below $T^* > T_c$. Our data rule out such a direct signature of a pseudogap in the IR response, as they show the *opposite behavior,* namely spectral weight is collected at low energies as temperature decreases.

Thin films of Bi-2212 were epitaxially grown by RF magnetron sputtering onto $SrTiO_3$ substrates. As the temperature is decreased from room temperature, their in-plane resistivity ρ_{ab} departs from an initial T-linear variation, developing a slight positive curvature in the overdoped case, and a negative one in the underdoped states. The latter behavior occurs below a temperature T^*, commonly assigned to the onset of a pseudogap. The phase diagram thus derived was entirely compatible with the one obtained from, e.g., ARPES measurements in single crystals and similar films [17,18]. Our spectra were recorded in (i) an underdoped (UD) film (thickness 2400 Å), with $T_c(R = 0) = 70$ K, and a relatively broad (onset-offset) transition width (~ 15 K), and $T^* \sim 170$ K, according to the resisivity curve, and (ii) an overdoped (OD) film (thickness 3000 Å), with $T_c(R = 0) = 63$ K, and a transition width ~ 5 K.

Our data were collected with a Bruker IFS 66v interferometer ($30-7000$ cm^{-1}). Near-infrared and visible data ($4000-25\,000$ cm^{-1}) were measured in a Cary 5E grating spectrometer. In the overlapping spectral range, measurements agree within 1.5%. The spectra were measured for 15 temperatures between 300 and 10 K. A He gas flow

cryostat allows to stabilize the temperature within ± 0.2 K. Figure 1 shows the reflectivity of the two samples, for a restricted set of temperatures, up to 1500 cm^{-1}.

Studying thin films precludes the use of the Kramers-Kronig (KK) transform to obtain the conductivity, due to the contribution of the substrate to the experimentally measured reflectivity. Therefore, in order to derive the optical conductivity of the films, we fit the reflectance of the film on top of a substrate using an attempt dielectric function for the film, and the optical constants of SrTiO$_3$ that we have experimentally determined for each temperature. The model dielectric function that fits the reflectance involves Lorentz and Drude oscillators, thus warranting causality. The procedure is described in detail elsewhere [19,20]. An interesting outcome of this approach is that it provides what can be considered our best guess for the extrapolation of the dielectric function in the energy range which is not available experimentally, i.e., $\omega \lesssim 30$ cm^{-1}.

A systematic error in the absolute value of the reflectivity ($\pm 0.5\%$) is irrelevant since we are only interested in its relative changes with temperature. The change of a spectrum versus temperature is defined within $\Delta_T \mathcal{R} \leq 0.2\%$, and the accuracy of the fit is $\Delta_F \mathcal{R} \leq 0.5\%$. This results in a typical error of 5% in the conductivity [21]. After completing the fitting procedure, we obtain all the relevant spectral functions for the film. We show in Fig. 2(a) the real part of the conductivity $\sigma_1(\omega, T)$ for the UD sample, at the same temperatures as in Fig. 1. Figure 2(b) displays similar results for the OD sample. The dc resistivity derived from our $\sigma_1(\omega, T)$ data are consistent with dc transport measurements on similar films. The so-called
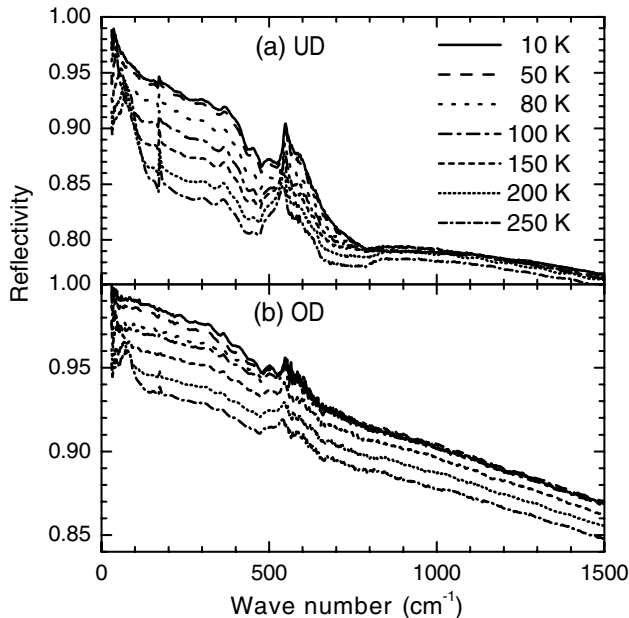
pseudogap effect shows up in the UD sample [Fig. 2(a)] as a depression in the conductivity above T_c, in the range $\sim 200-1500$ cm^{-1}, as observed previously in single crystals. In the OD sample, a similar depletion of the conductivity occurs above T_c in the same spectral range. The question is whether the observed loss is balanced by the increase of spectral weight at low energy.

Before we address this point, we touch upon the scattering rate, defined as

$$\frac{1}{\tau(\omega)} = \frac{2\pi}{Z_0} \Omega_p^2 \, \mathrm{Re}\left[\frac{1}{\sigma(\omega)} \right]. \tag{1}$$

The plasma frequency Ω_p (in cm^{-1}) was obtained by integrating $\sigma_1(\omega)$ up to 1 eV. $Z_0 = 377$ Ω is the vacuum impedance. Taking $\sigma(\omega)$ in Ω^{-1} cm^{-1}, one gets $1/\tau(\omega)$ in cm^{-1}. $1/\tau(\omega)$ is shown in the insets of Figs. 2(a) and 2(b). As we are focusing on the normal state, we have only plotted $1/\tau(\omega)$ above T_c. In the UD sample, when the temperature decreases, a gradual depletion of $1/\tau(\omega)$ below ~ 700 cm^{-1} is observed [inset of Fig. 2(a)]. A similar, less pronounced, depletion below the same energy is also observed in the OD sample already above T_c [inset of Fig. 2(b)]. This fact can already be traced in those single crystals where doping was changed only by varying the amount of oxygen, and has been ascribed in the underdoped regime to the opening of an optical pseudogap [13].

Our data show distinctly the connection between the increase of the conductivity and the depression of the low energy scattering rate in *both samples*. The most striking result is the narrowing and increase of the low frequency

FIG. 1. Reflectivity of (a) the underdoped and (b) the overdoped film. The sharp peaks at 550, 120, and below 100 cm^{-1}, especially visible in the underdoped sample, are the phonon modes of the substrate.
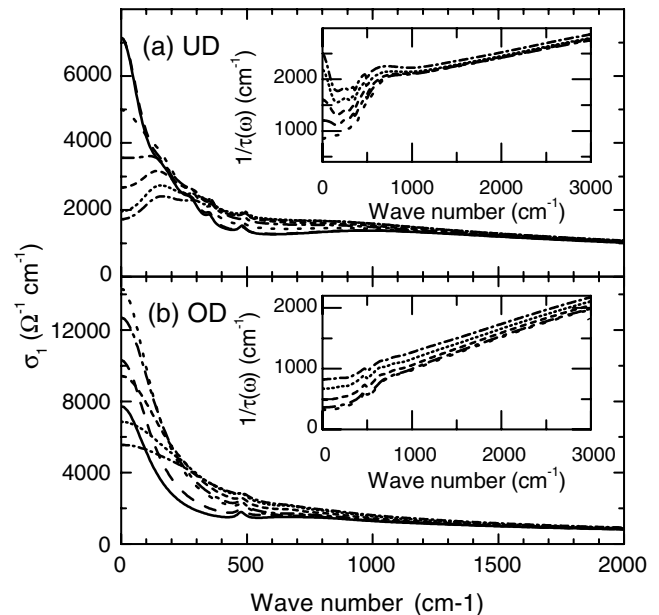

FIG. 2. Real part of the conductivity (from fit) of (a) the underdoped and (b) the overdoped film. The insets show the scattering rate for $T > T_c$ (see text), using $\Omega_p = 16\,000$ and $16\,600$ cm^{-1}, respectively. The line types refer to the same temperatures as in Fig. 1. The conductivity extrapolated from 30 cm^{-1} down to zero results from the fit (see text).

Drude-like peak in the UD sample conductivity occuring *in the superconducting state,* with no obvious loss of spectral weight associated with the formation of the condensate. In contrast, in the OD sample, the conductivity drops below T_c, and the missing area is clearly visible. The superconducting state is discussed elsewhere [22].

Since a set of 15 temperatures is available, we can look for a loss of spectral weight as the temperature is lowered. This would be the *actual signature of a pseudogap* opening in the vicinity of the Fermi level. We have therefore integrated the conductivity in order to derive an effective carrier density up to ω_M, according to

$$N_{\text{eff}} \propto \int_{0^+}^{\omega_M} \sigma_1(\omega)\,d\omega . \qquad (2)$$

In order to display the thermal evolution starting from room temperature for various ω_M in a single plot, we have normalized $N_{\text{eff}}(\omega_M, T)$ with respect to $N_{\text{eff}}(\omega_M, 300\,\text{K})$. We show in Fig. 3, for both samples, the temperature variation of this normalized effective carrier density (equivalent to a normalized SW) for a set of ω_M values spanning the full experimental frequency range. When $\omega_M = 100$ cm^{-1}, the SW *increases* sharply as the temperature decreases down to T_c, showing that spectral weight is collected at low frequency in the normal state. As ω_M is increased, the transfer of SW becomes less apparent. The data for $\omega_M = 5000$ and $10\,000$ cm^{-1} (not shown) can hardly be distinguished from those for

20 000 cm^{-1}. This shows that SW going to low frequency ($\omega_M < 100$ cm^{-1}) comes mostly from the midinfrared range. The most important conclusion is that, for decreasing T, and for all ω_M values considered (up to 20 000 cm^{-1}), there is a systematic *increase of the spectral weight,* both in the UD and OD samples. This observation is at odds with the opening of a pseudogap which would yield a *decrease* of SW. Note that a break in this trend of increasing SW as T decreases is seen only at the superconducting transition, where the SW hardly decreases in the UD sample, and drops sharply in the OD sample (Fig. 3) [22].

So far, we have integrated Eq. (2) starting at zero frequency, using the computed $\sigma_1(\omega)$ for $\omega < 30$ cm^{-1}. In order to check the sensitivity of our results to a low frequency cutoff, we have done a similar calculation, starting the integration from 30 and from 50 cm^{-1}. The result for the UD case is shown for $\omega_M = 1000$ cm^{-1} in the inset of Fig. 3. Introducing a low frequency cutoff results in a decrease of SW already at $T \lesssim 100$ K, and a broad maximum develops in temperature as the cutoff is increased. A similar behavior is observed for the overdoped sample. This being an artifact, confirms that no detectable loss of SW is seen, making it impossible to locate T^* from the thermal evolution of the effective carrier density. Actually, from all the observations described above, it is clear that there is enough room in the low frequency part of the spectrum to balance the small depression of spectral weight observed in the conductivity data. Our data strongly suggest that the pseudogap has no clear-cut signature in the infrared conductivity. Both the depletion in the scattering rate and the occurrence of a narrow peak in the real part of the conductivity are settling gradually as the temperature is decreased.

Since the existence of a pseudogap is established through a variety of experimental techniques, in both the spin and the charge channels, why does the optical conductivity appear not to display this phenomenon? One logical hypothesis is that the conductivity probes mostly the quasiparticles (QP's) along the (π, π) direction, where no pseudogap opens [9,10]. Indeed, recent ARPES experiments on *optimally* doped Bi-2212 suggest that, as single-particle scattering rates near the nodes and transport rates exhibit similar energy and temperature dependencies, the in-plane transport might be dominated by nodal excitations [23,24].

One may explore how reasonable the above hypothesis is in a simple framework. In the semiclassical approximation, the finite-frequency conductivity is [25]:

$$\sigma(\omega) \propto \int \frac{v_{\mathbf{k}}^2}{\tau^{-1}(\varepsilon_{\mathbf{k}}) - i\omega} \left(\frac{\partial f}{\partial \varepsilon} \right)_{\varepsilon = \varepsilon_{\mathbf{k}}} d^2 k, \qquad (3)$$

$\mathbf{v_k}$ and $\varepsilon_{\mathbf{k}}$ are the carrier bare (band-structural) velocity and energy, respectively. $\tau^{-1}(\varepsilon_{\mathbf{k}})$ is the transport scattering rate, and f is the Fermi distribution function. As the derivative of f is peaked at the Fermi energy,
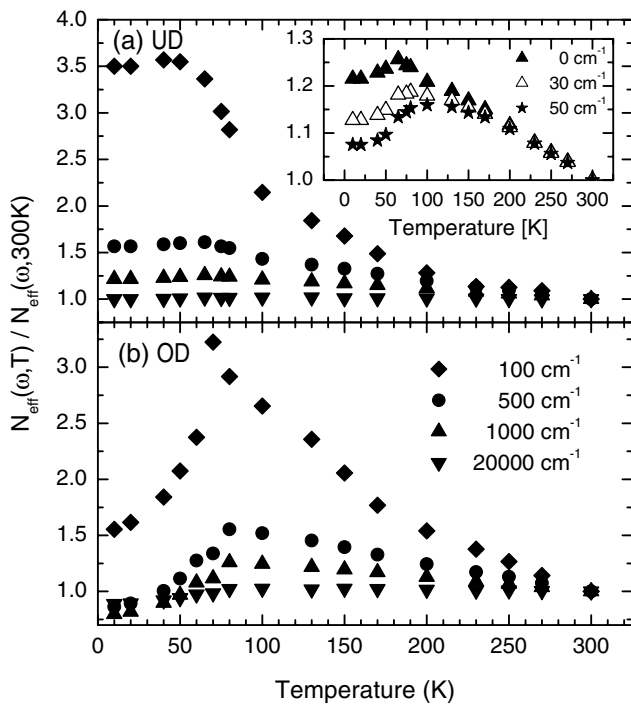

FIG. 3.   Spectral weight versus temperature for (a) the underdoped and (b) the overdoped film. Different symbols refer to different cutoff frequencies ω_M. The inset shows the effect of starting the integration from 0, 30, and 50 cm^{-1} (up to $\omega_M = 1000$ cm^{-1}).

the integration in Eq. (3) over the direction perpendicular to the Fermi surface (FS) shows that a critical weighting factor is the bare Fermi velocity $\mathbf{v}_F$, which—due to the anisotropy of the FS—is smaller in the $\mathbf{k} = (0, \pi)$ than in the $\mathbf{k} = (\pi, \pi)$ direction. Simple calculations yield $v_F(\pi, \pi)/v_F(0, \pi) \sim 5$ [26,27]. As a consequence, $\sigma_1(\omega)$ should be mostly sensitive to the nodal QP's, and no pseudogap can be seen. Furthermore, as the anisotropy of the FS holds in a sizable doping range, the trends in the T dependence of the normal-state $\sigma_1(\omega)$ should not strongly depend on doping. This is indeed what our data show (Figs. 2 and 3).

Within this framework, the low-frequency narrowing of $\sigma_1(\omega)$ when temperature decreases can be assigned to the scattering of nodal QP's (through coupling to low-energy excitations) becoming less efficient. An indirect contribution of the pseudogap may arise because fewer states are available for scattering in the "pseudogapped" regions, thus enhancing the QP's relaxation time. In the dc limit, this would imply a downward deviation of the resistivity from its high-temperature behavior.

Up to now, ARPES measurements of the *renormalized* Fermi velocity $\mathbf{v}_F^*$ in optimally doped Bi-2212 have reported an anisotropy of $v_F^*(\pi, \pi)/v_F^*(0, \pi) \sim 3\text{–}4$ in the superconducting state [24]. Our results call for a deeper study of $\mathbf{v}_F^*$ as a function of doping and temperature, and of the relevance of the bare and renormalized Fermi velocities in the determination of the optical conductivity.

Among other interpretations, a gain in the low-frequency $\sigma_1(\omega)$ can be also regarded as a precursor of the coherent $\delta(\omega)$ peak in the superconducting state, coming from either collective excitations [28] or phase fluctuations of the superconducting gap [29]. However, these models neglect the strong $\mathbf{k}$ dependence of the electronic structure, of the gap, and of the interactions in real materials. As our results demonstrate, these are crucial issues in the detailed understanding of transport phenomena in high-T_c materials.

In summary, we showed that, at $T > T_c$, the in-plane IR response of overdoped and underdoped Bi-2212 are qualitatively the same. In particular, no loss of spectral weight, therefore no pseudogap signature, is seen in the normal state conductivity. We suggest that these are consequences of the anisotropy of the Fermi velocity.

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[21] The magnitudes of the error bars given in this work arise from various numerical simulations of generic reflectivity spectra of cuprates.

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