Optical spectroscopy as a probe of gaps and kinetic electronic energy in p- and n-type cuprates

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Abstract

The real part of the optical in-plane conductivity of p- and n-type cuprates thin films at various doping levels was deduced from highly accurate reflectivity measurements. We present here a comprehensive set of optical spectral weight data as a function of the temperature $T(>T_c)$, for underdoped and overdoped samples. The temperature dependence of the spectral weight is not universal. Using various cut-off frequencies for the spectral weight, we show that n-type $Pr_{2-x}Ce_xCuO_4$ and p-type $Bi_2Sr_2CaCu_2O_{8+\delta}$ exhibit both similarities and striking differences. The Fermi surface is closed in overdoped metallic samples. In underdoped $Pr_{2-x}Ce_xCuO_4$ samples, it clearly becomes discontinuous giving rise to a "pseudogap" signature. It is argued that such a signature is subtle in underdoped $Bi_2Sr_2CaCu_2O_{8+\delta}$.
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1. Introduction

The properties of a doped Mott insulator and the role of strong correlations stand among the present great challenges in condensed matter physics, and high critical temperature superconductivity in cuprates is one of those. Unlike conventional metals, various energy scales are present in the normal state of these materials, and vary throughout the phase diagram.

One energy scale is related to the pseudogap, evidenced by many experimental techniques [1]. Angular resolved photoemission spectroscopy (ARPES) of hole doped cuprates yields a "low energy" pseudogap, opening below a (doping dependent) temperature T^*. This pseudogap is traced by the shift with respect to the Fermi level, of the leading edge of the energy dependent curve (EDC) along the $(0, \pi)$ direction of the **k** space. It is argued that a "high energy" pseudogap develops, also along the $(0, \pi)$ direc-

tion, associated with a broad feature which tracks the low energy pseudogap [2]. As for infrared spectroscopy, which is the topic of this paper, hole doped cuprates reveal a pseudogap feature only through the depletion of the low energy optical scattering rate [3].

In electron-doped cuprates, a "low energy" pseudogap was observed by tunneling spectroscopy [4]. A "high energy" pseudogap is seen in ARPES also as a broad feature in the EDC [5], however along the $(0.65\pi, 0.3\pi)$ direction. The infrared in-plane conductivity reveals a pseudogap at a comparable energy scale. The temperature T^* where this "pseudogap" opens depends on doping [6,7]. T^* is best tracked, as shown further, by measuring the spectral weight $W(\Omega, T)$, i.e., the integral of the real part $\sigma_1(\omega)$ of the optical conductivity up to some cut-off frequency Ω:

$$W(\Omega, T) = \int_O^\Omega \sigma_1(\omega, T)\, d\omega \qquad (1)$$

Another energy scale is relevant to cuprates, namely the electronic kinetic energy E_K. In a single band, tight-binding model with nearest-neighbor interactions, E_K is related to

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the spectral weight, provided the cut-off frequency Ω is chosen so as to restrict to the free carrier contribution [8]. The electronic kinetic energy scale reflects the strength of the correlations [9]. Its temperature dependence remains unsettled, from a theoretical point of view and from the experiments, where this temperature dependence varies from sample to sample [10–13]. Such discrepancies suggest that the T dependence of E_K (or of the spectral weight) is not universal.

In this communication, we take advantage of our measurements in both n-type $Pr_{2-x}Ce_xCuO_4$ (PCCO) and p-type $Bi_2Sr_2CaCu_2O_{8+\delta}$ (BSCCO) thin films. Relying on the metallic behavior of the overdoped samples, whether n- or p-type, we note that the underdoped compounds deviate from this metallic behavior at some temperature T^*. We argue that such deviations, although quite different in PCCO and BSCCO, are associated with the onset of a "pseudogap". One caveat though: we use the term "pseudogap" for the sake of simplicity, and we mean a density gap. However it may relate to different physics. This still vividly debated issue is beyond the scope of this communication.

2. Experimental

Samples are thin films with thicknesses ranging from 2000 to 3000 Å. Details about their preparation and characteristics and about the experimental procedure can be found elsewhere [7,14].

Fig. 1 displays the optical conductivity for two PCCO and BSCCO underdoped samples in a temperature range where pseudogaps were claimed previously to be identified. A clear dip and a maximum at ~ 1700 cm^{-1} develop in the PCCO sample as T decreases. A dip and a maximum at ~ 800 cm^{-1} are hardly visible at 75 K in the case on underdoped BSCCO. If such maxima relate to a density gap fea-
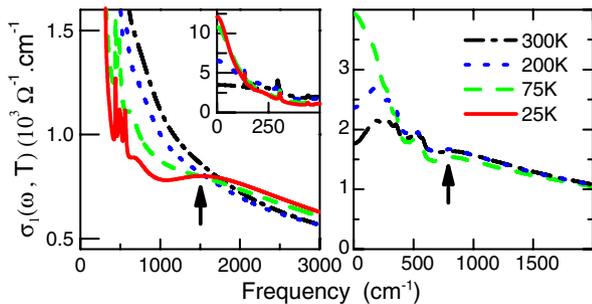
ture, then upon the opening of such a gap, states are transferred from the Fermi energy to a higher energy. This reflects as a spectral weight transfer toward high energy at the temperature where the gap opens. We expect this transfer of spectral weight to be best observed when integrating up to a cut-off frequency Ω (Eq. (1)) of the order of the gap energy.

Figs. 2 and 3 show the change of spectral weight for a set of cut-off frequencies spanning the range $\Omega = 500–20{,}000$ cm^{-1} in PCCO (Fig. 2) and BSCCO (Fig. 3). The Ω cut-off frequencies are the same in a single family, but not in-between the two families. The values were selected in order to display the characteristic energy scales where changes in the temperature dependence are best seen.

We translated the spectral weight in units of a carrier density per copper N_{eff} [13]. An estimate of the actual effective carrier density per copper is obtained for $\Omega = 8000$ cm^{-1}. We find $N_{eff}(300 \text{ K}) = 0.22$ and 0.26 for the overdoped and underdoped PCCO respectively, 0.32 and 0.35 for the underdoped and overdoped BSCCO respectively [15]. The difference $N - N_{eff}(300 \text{ K})$ versus T is displayed in Figs. 2 and 3. The scatter of the points at 20,000 cm^{-1} in the right panel of Fig. 2 gives an upper limit for our data accuracy (± 0.002 in units of N_{eff}) [16].

Both overdoped samples (left panels, Figs. 2 and 3) exhibit a similar behavior: $N - N_{eff}(300 \text{ K})$ increases steadily as T decreases, approximately by the same amount.
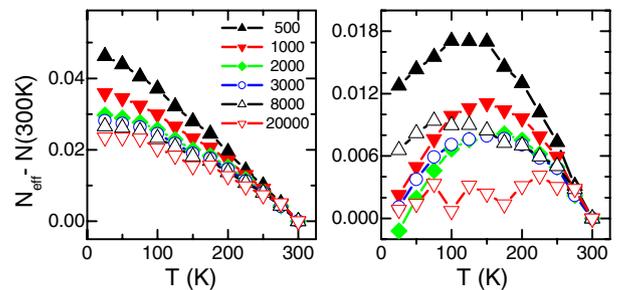

Fig. 2. Change of spectral weight – translated into the effective carrier density N_{eff} per Cu – versus temperature, for the underdoped – $x = 0.13$, $T_c = 15$ K – and overdoped – $x = 0.17$, $T_c = 15$ K – PCCO films. The cut-off frequencies Ω are indicated in the left panel.

Fig. 1. Left panel: real part $\sigma_1(\omega,T)$ of the optical conductivity of the underdoped PCCO sample – $T_c = 15$ K. The inset shows the conductivity in the range 0–500 cm^{-1} over the full scale along the Y axis. An expanded view in the main left panel shows the changes of $\sigma_1(\omega)$ with temperature over the range 0–3000 cm^{-1}. The arrow shows a clear maximum in the PCCO conductivity at 25 K, just above T_c. Right panel: real part $\sigma_1(\omega,T)$ of the optical conductivity of the underdoped BSCCO sample – $T_c = 70$ K. $T = 300$ K, 200 K, 75 K, 25 K (the latter is not shown, lying below T_c for BSCCO). The arrow shows a tenuous maximum in the BSCCO conductivity at 75 K, just above T_c.
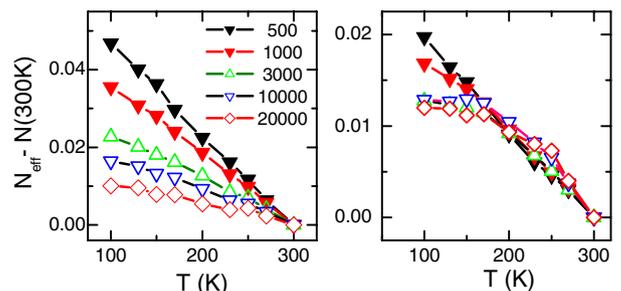

Fig. 3. Change of spectral weight – translated into the effective carrier density N_{eff} per Cu – versus temperature, for the underdoped – $T_c = 70$ K – and overdoped – $T_c = 63$ K – BSCCO films. The cut-off frequencies Ω are indicated in the left panel.

This increase results from a single mechanism, which does not depend upon the details of the Fermi surface: the quasiparticle (QP) scattering rate is overall decreasing as the temperature decreases, thus putting more and more spectral weight in the Drude-like peak at low energy. $N - N_{eff}(300\,K)$ is the smallest at the largest cut-off; this is the expected trend since the f-sum rule must be eventually satisfied at large frequencies. Note that in the case of BSCCO, at 20,000 cm^{-1}, $N - N_{eff}(300\,K)$ lies indeed almost within the experimental error, while the f-sum rule is not fulfilled yet at this energy in PCCO.

The point we wish to emphasize is that the overdoped samples exhibit a metallic behavior, associated with a closed Fermi surface, and there is no hint as T decreases, of a modification of this Fermi surface.

In underdoped PCCO (Fig. 2, right panel), as T is lowered from 300 K, the spectral weight increases as observed in the overdoped samples: this is a signature of a metallic state. The material remains metallic down to 25 K, as certified by a Drude-like peak (shown in the inset of Fig. 1) in $\sigma_1(\omega, T)$. Below $\sim$150 K, the trend is reversed: $N - N_{eff}(300\,K)$ decreases. This can be understood without the need of a specific model. The optical conductivity is an average over the $\mathbf{k}$ space. Since a metallic behavior is present down to 25 K, the observed decrease implies that the Fermi surface becomes discontinuous possibly due to a partial gap (pseudogap) opening along specific $\mathbf{k}_0$ values (identified by ARPES). The QP states which vanish at those $\mathbf{k}_0$ values no longer contribute to the Drude peak, and are transferred at high energy, hence a decrease of the low energy spectral weight below 150 K. The characteristic energy scale of this pseudogap lies in the range 2000–3000 cm^{-1}. It corresponds to the maximum seen in the optical conductivity at $\sim$1700 cm^{-1} (Fig. 1, left panel). It is also is compatible with ARPES data [5].

The spectral weight in underdoped BSCCO (Fig. 3, right panel) does not decrease whatever the cut-off frequency. It was independently experimentally established that a pseudogap is present below a temperature T^* in this material (in this specific sample, from the resistance measurement, $T^* \sim 130$ K [8]). Nevertheless, Fig. 3 (right panel) shows that, at $\Omega \simeq 3000$ cm^{-1}, the spectral weight levels off. The temperature where this levelling off occurs is $\sim$150 K. $N - N_{eff}(300\,K)$ remains constant down to T_c, showing no decrease as compared to underdoped PCCO. Moreover, this plateau appears only for cut-offs beyond 3000 cm^{-1}. At lower energy, the metallic contribution is the dominant mechanism for spectral weight redistribution. If this plateau was to be related to the onset of the pseudogap, it would imply that the spectral weight redistribution extends far beyond our full experimental range, unlike PCCO (in Fig. 2, right panel, the spectral weight is eventually conserved at 20,000 cm^{-1}). A further concern is that in underdoped BSCCO single crystals, the plateau is not seen [12]. Hence its possible interpretation in terms of a pseudogap certainly requires one more step in the analysis.
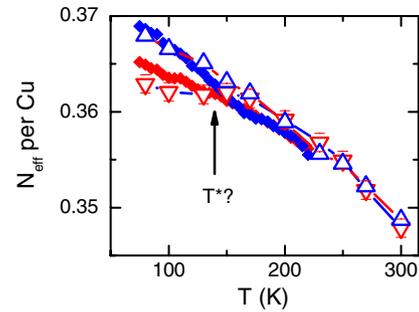

Fig. 4. Spectral weight computed for $\Omega = 8000$ cm^{-1} (films, open symbols [14]) and 10,000 cm^{-1} (crystals, full symbols [12]). For each underdoped sample, the absolute value has been normalized so as to overlap the 300 K spectral weight of the overdoped akin compound. The spectral weight of underdoped samples departs form the overdoped at $\sim$140 K.

3. Discussion

Having shown that the overdoped samples display a metallic behavior, we propose to normalize, within the experimental error, the spectral weight of the underdoped sample to the overdoped one at 300 K. This is shown in Fig. 4 for $\Omega = 8000$ cm^{-1}, for the sake of a comparison with single crystals. Similar plots can be found for PCCO samples in earlier papers [7,8]. After normalizing so, both samples appear to have a similar metallic behavior down to $\sim$140 K, within the error bars. Next we superimpose single crystals data at $\Omega = 10,000$ cm^{-1} [12], also normalizing $N_{eff}(300\,K)$ of the underdoped crystal to the overdoped one. These two different sets of data turn out to be in surprisingly good agreement. Moreover, the spectral weight increases less rapidly in the underdoped sample than in the overdoped one, below approximately the same temperature. The difference develops and reaches $\sim$0.005, still larger than our error bar. Therefore, in this respect, we observe "missing spectral weight" below $\sim$140 K. We suggest that it is a (subtle) signature of the onset of the pseudogap in underdoped BSCCO.

According to the terminology recalled above, the pseudogap in BSCCO is a "low energy" one, whereas in PCCO it would be a "high energy" one. If such a distinction is valid, then different mechanisms for the onset of these pseudogaps may very well be at stake, as suggested by numerous authors. These pseudogaps might however be related: in hole doped cuprates, the broad feature in ARPES tracks the low energy pseudogap [2]. Our contribution here is to show that both have an optical signature in terms of missing spectral weight, although the signatures are qualitatively and quantitatively different. More accurate studies of the in-plane conductivity in hole-doped materials are still necessary in order to allow a more comprehensive comparison with the pseudogap in n-type materials.

4. Conclusion

From the temperature dependence of the spectral weight in n-type PCCO cuprates, one can clearly see that the

Fermi surface breaks off due to the opening of a pseudogap. In p-type BSCCO samples, the pseudogap does not give rise to a clearcut signature. We used the metallic temperature variation of the overdoped sample as a reference variation. Doing so, the temperature dependence for the underdoped samples departs at a temperature T^* from the reference temperature dependence. This difference is large for PCCO (10%), but does not exceed 1.5% in terms of the total spectral weight in BSCCO. We suggest that in the latter compound, it is a subtle signature of the pseudogap. Whether the difference relates to a basic one *i.e.* a different mechanism for these partial gaps, remains to be settled.

Acknowledgements

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[15] We used a unit cell volume (per Cu) of 183 Å^3 for PCCO, and 228 Å^3 in BSCCO. The small differences in the N_{eff}(300 K) can depend on specific details of the technical procedure used to establish the absolute value of the spectral weight [14].
[16] The accuracy is even better at lower energies $\sim$10000 cm^{-1}, being further from our highest available measuring frequency.