High-Temperature Optical Spectral Weight and Fermi-liquid Renormalization in Bi-Based Cuprate Superconductors


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The optical conductivity \(\sigma(\omega)\) and the spectral weight \(W(T)\) of two superconducting cuprates at optimum doping, \(\text{Bi}_2\text{Sr}_{2-x}\text{La}_x\text{CuO}_8\) and \(\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8\), have been first measured up to 500 K. Above 300 K, \(W(T)\) deviates from the usual \(T^2\) behavior in both compounds, even though \(\sigma(\omega \rightarrow 0)\) remains larger than the Ioffe-Regel limit. The deviation is surprisingly well described by the \(T^4\) term of the Sommerfeld expansion, but its coefficients are enhanced by strong correlation, as shown by the good agreement with dynamical mean field calculations.

Since the discovery of high-\(T_c\) cuprates, the research has been obviously focused on their “low-temperature” properties. However, the electronic correlations, which are expected to play a central role in the low-\(T\) phenomena like superconductivity and pseudogap, are also likely to affect the cuprate properties at higher \(T\). In this respect, the high-\(T\) behavior can provide direct information about the real nature of the fermionic excitations, which, at low \(T\), may be masked by competing ordering phenomena. Indeed, it was suggested that high-\(T\) effects such as the violation of the Ioffe-Regel (I-R) limit for resistivity saturation and the quasiparticle (QP) thermal decoherence, are a hallmark of the same Hubbard physics which controls the superconductivity and pseudogap, are also likely to affect the real nature of the fermionic excitations, which, at low \(T\), may be masked by competing ordering phenomena.

We focus our investigation on the optical spectral weight

\[
W(\Omega, T) = \int_0^\infty \sigma(\omega, T) d\omega,
\]

where \(\sigma(\omega, T)\) is the real part of the \(ab\)-plane optical conductivity, and \(\Omega\) is a cutoff frequency. \(W\) is a model-independent quantity which provides important information on the evolution of the electronic dynamics with temperature [2–8].

For \(\Omega \rightarrow \infty\), the standard \(f\)-sum rule implies that \(W\) is independent of \(T\). However, useful “restricted sum rules” can be defined for finite \(\Omega\)’s. If, for example, \(\Omega = \omega_p\) [i.e., the screened plasma frequency which in the following will be approximated with the plasma edge in the reflectivity \(R(\omega)\)], a tight-binding model with nearest-neighbor hopping provides

\[
W(\omega_p, T) = -\frac{\pi e^2 a^2}{2\hbar^2 V} K(T) \\
= W_0 - B(\omega_p) T^2 + C(\omega_p) T^4,
\]

where \(a\) is the lattice constant, \(V\) is the sample volume, and \(C\) is an approximate equality in Eq. (2) comes from the Sommerfeld expansion of the kinetic energy \(K(T)\) up to the fourth order in \(T\). In the literature, Eq. (2) is typically limited to the second order (\(C = 0\), as the \(T^2\) dependence of \(W\) is well verified in many metals below room temperature, including several high-\(T_c\) superconductors [2,4]. Despite this “conventional” behavior, the cuprates show peculiar features [4]: (i) the \(T^2\) dependence extends up to \(\Omega\)’s far larger than \(\omega_p\); (ii) unlike in ordinary metals, where \(W_0\)-which accounts for all the carriers in the conduction band—and the “thermal” coefficient \(B(\omega_p)\) are governed by the same nearest-neighbor hopping rate \(t_0\), in cuprates \(B \propto t_r^{-1}\) with \(t_r = t_0/10\). While some authors [7] attribute these effects entirely to the existence of a finite cutoff, in Ref. [8] a quantitative agreement has been found between experimental data and dynamical mean field theory (DMFT) calculations, taking into account both the finite cutoff and the strong correlation present in high-\(T_c\) cuprates.

In the present Letter we investigate the in-plane optical conductivity of two cuprates in their normal phase up to 500 K, in order to study the behavior of \(W(\Omega, T)\) towards the I-R limit. The samples are two single crystals at optimum doping, \(\text{Bi}_2\text{Sr}_{1.6}\text{La}_{0.4}\text{CuO}_8\) (Bi-2201) and \(\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8\) (Bi-2212), grown by the floating-zone (FZ) technique [9]. \(\text{Bi}_2\text{Sr}_{1.6}\text{La}_{0.4}\text{CuO}_8\) is a single Cu-O layer cuprate, with maximum critical temperature

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$T_c^\text{max} \approx 33$ K. In this system, whose optical spectra at low doping were reported previously [10], optimum doping occurs at $x = 0.4$, which corresponds to 0.16 holes per Cu [11,12]. Bi$_2$Sr$_2$CaCu$_2$O$_8$ is the well-known double-layer cuprate with $T_c^\text{max} \approx 93$ K.

The $ab$-plane resistivity $\rho(T)$ is represented in the inset of Fig. 1. Above the sharp superconducting transitions at $T_c = 33$ K for Bi-2201 and 93 K for Bi-2212, both curves display the linear behavior typical of optimally-doped cuprates. The reflectivity $R(\omega)$ was measured at near-normal incidence, shortly after cleaving the samples, with a Michelson interferometer between 40 and 22 000 cm$^{-1}$, at several $T > T_c$, stable within $\pm 1$ K. The reference in the infrared (visible) range was a gold (silver) film evaporated in situ onto the sample, which was mounted in a closed-cycle cryostat below room $T$, heated inside an optical vacuum chamber above 300 K. The intensity reflected both by the sample and reference was measured at every $T$, for any spectral range, in order to compensate the thermal displacements of the sample holder. The chemical stability of both samples was checked by measuring $R(\omega)$ at 300 K after every high-$T$ cycle. $R(\omega)$ is shown at the lowest and highest temperatures in the inset of Fig. 1 for both Bi-2201 and Bi-2212. As shown by the vertical arrows in the inset, at any $T$ the plasma edge is at $\omega_p \approx 8000$ cm$^{-1}$ in Bi-2201 and $\omega_p \approx 10 000$ cm$^{-1}$ in Bi-2212.

The real part $\sigma(\omega)$ of the $ab$-plane optical conductivity was finally obtained from $R(\omega)$ through standard Kramers-Kronig transformations. First extrapolations of $R(\omega)$ to $\omega = 0$ were based on Drude-Lorentz fits, which provided a $\sigma(0)$ which deviated from the $\sigma_{ab}$ measured at the same $T$ within $\pm 1\%$. Afterwards, these fits were adjusted exactly to $\sigma_{ab}$. The extrapolations to high frequency were based on the data of Ref. [13] up to 40 eV and on a power law beyond this energy. The resulting $\sigma(\omega)$ is shown in Figs. 1(a) and 1(b) at selected temperatures between $T_c$ and 500 K, for Bi-2201 and Bi-2212, respectively. In both samples the edge of the lowest electronic band appears at $\omega \approx 10 000$ cm$^{-1}$. The Drude peak in the far infrared broadens with increasing temperature, becoming a flat contribution at 500 K.

In the insets of Fig. 2, the spectral weight $W(\omega_p)$, as obtained from $\sigma(\omega)$ by Eq. (1), is shown as a function of $T$ for both Bi-2201 and Bi-2212. The error bars have been estimated by assuming a 1% error on the raw $R(\omega)$ throughout the measuring range. As one can see from the Figure, the $T^2$ dependence predicted by Eq. (2) limited to the second order (dotted line) well fits the measured $W(\omega_p)$ only for $T \leq 300$ K. A strong deviation from this behavior is instead evident above room temperature for both Bi-2201 and Bi-2212. One may wonder whether this is due to the system approaching the I-R limit, where the quasiparticle picture breaks down because the electron mean free path $\ell$ becomes comparable with the lattice constant $a$ [1].

To evaluate the resistivity at the I-R limit, following Ref. [1] one may assume a cylindrical Fermi surface of radius $k_F = \sqrt{2\pi/a}$, with $a = 0.383$ nm and height $2\pi/c$. Here, $c \approx 1.23$ nm for Bi-2201 [14] and $c \approx 0.765$ nm for Bi-2212 [15] is the average separation between Cu-O planes. At the I-R limit $\ell = a$, and hence

$$\rho_{ab}^{\text{I-R}} = \frac{2\pi\hbar c}{e^2k_Fa} \approx 0.055(c/a_0)\text{ m}\Omega\text{ cm}, \quad (3)$$

where $a_0$ is the Bohr radius. One thus obtains $\rho_{ab}^{\text{I-R}} \approx 1.3$ m$\Omega$ cm for Bi-2201 and $\rho_{ab}^{\text{I-R}} \approx 0.8$ m$\Omega$ cm for Bi-2212. The corresponding $\sigma_{ab}^{\text{I-R}}$ are marked in Figs. 1(a) and 1(b) by arrows close to the vertical axis. As one can see, even at the maximum temperature investigated, $\sigma(\omega)$ is much larger than the I-R limit, indicating
that a trace of coherent fermionic excitations still survives in both Bi-based systems.

Moreover, one finds that the deviation of $W(\omega_p, T)$ from the $T^2$ behavior is satisfactorily reproduced at all temperatures if one fits to data the whole Eq. (2), namely, if one includes the $T^4$ term of the Sommerfeld expansion (dashed lines in both insets of Fig. 2).

In order to check for the generality of such behavior, the cutoff frequency $\Omega$ in Eq. (1) was varied from $\omega_p/2$ to $3\omega_p/2$, and deviations from the $T^2$ dependence quite similar to those in the insets of Fig. 2 were always found. The resulting set of $W(\Omega, T)$ values were fit up to the $T^4$ term, and the coefficients $B(\Omega)$ and $C(\Omega)$ were determined in terms of $W_0$.

The results are shown in Fig. 3. We obtained $b(\omega_p) = B/W_0 \approx 4.0 \times 10^{-7}$ K$^{-2}$ in Bi-2201 and $b(\omega_p) = 2.0 \times 10^{-7}$ K$^{-2}$ in Bi-2212. Figure 3 also shows the $T^4$ coefficient $C(\Omega)$ in the Sommerfeld expansion. In both compounds, $c(\Omega) = C/W_0$ decreases with $\Omega$ like $b(\Omega)$. Therefore, within errors, the ratio $c/b$ in Fig. 3(c) is independent of $\Omega$ and, surprisingly, also the same in both optimally doped samples. At the screened plasma frequency one has $c(\omega_p) = 6.1 \times 10^{-13}(3.6 \times 10^{-13})$ K$^{-4}$ and

$$
c(\omega_p)/b(\omega_p) = (1.5 \pm 0.4) \times 10^{-9}((1.8 \pm 0.5) \times 10^{-6}) \text{ K}^{-2}$$

for Bi-2201 (Bi-2212).

The strong temperature dependence of $W$ in the low-$T$ regime and the deviations from the $T^2$ behavior at intermediate $T$, suggest to investigate the role of electron correlations in renormalizing the Sommerfeld coefficients $B$ and $C$. To this purpose, we have performed DMFT calculations [16] for a single-band Hubbard model with realistic values of the hopping parameters for both Bi-based compounds [17]. Following the scheme presented in Refs. [8,18], such calculations qualitatively reproduce the absolute (material dependent) value of $W$. The temperature behavior can be reproduced even quantitatively when rescaling $W$ with its (extrapolated) value at $T = 0$ value $W_0$.

The comparison between experimental and DMFT values of $W(T)/W_0$ is reported in Fig. 2 for a local Coulomb interaction $U = 12t_0$ and optimal doping (0.16 holes per Cu). The agreement with data is excellent, as DMFT results (blue diamonds in Fig. 2) capture both the

![FIG. 2 (color online). Temperature-dependent optical spectral weight $W(\omega_p, T)$ of optimally doped (a) Bi-2201 and (b) Bi-2212, normalized to the (extrapolated) value at $T = 0$. The I-R data (red circles) are compared with DMFT results for the restricted sum rule (blue diamonds) of the single-band Hubbard model. Also shown are theoretical calculations for the noninteracting system ($U = 0$) and the lowest-order Sommerfeld expansion, where the coefficient $B$ is simply rescaled by the QP DMFT weight ($Z$ scaled). In panel (a) DMFT results for the total sum rule are displayed for comparison (green squares). In the inset the dotted (dashed) line indicates the fit performed on $W(\omega_p, T)$ data using Eq. (2) up to the second (fourth) order.](https://www-prl.aps.org/105/77002/figure2)

![FIG. 3 (color online). Normalized coefficients $b(\Omega) = B/W_0$, $c(\Omega) = C/W_0$, and $c(\Omega)/b(\Omega)$ for Bi$_2$Sr$_{1.6}$La$_{0.4}$CuO$_6$ and Bi$_2$Sr$_2$CaCu$_2$O$_8$, as obtained from the fits to $W(T, \Omega)$. The lines are guides to the eye.](https://www-prl.aps.org/105/77002/figure3)
strong $T^2$ dependence of $W$ at low $T$ between 0 and 250 K for Bi-2201 and Bi-2212 (within $\sim 3\%$ and $\sim 1.5\%$, respectively), and the deviation from the $T^2$ behavior for $T \approx 300$ K. Even when the sum rule is extended up to infinity (see the DMFT calculations for the 2201 material, green squares in Fig. 2) $W$ is still dependent on $T$. Such a dependence is certainly weaker than that obtained for a finite cutoff, but it remains significant. This result demonstrates, that the effects of strong correlation [8] and finite cutoff [7] contribute about equally to the observed temperature dependence. The 2201 system displays a stronger $T$ dependence than the 2212 compound as the energy scale $T$ is smaller. This proves the major role of electronic correlations in determining the optical behavior of both Bi-based cuprates in the whole temperature range.

In a first approximation, the observed $B$ and $C$ enhancement can be related to the renormalization factor $Z$, which controls the QP bandwidth through

$$1/Z = 1 - \frac{\delta \Sigma(\omega = 0)}{\partial \omega}. \quad (4)$$

Here $\Sigma$ is the DMFT self-energy and for the parameters we studied $1/Z$ ranges between 6 and 8. On the basis of a simple dimensional argument one can expect that the Sommerfeld coefficients are renormalized by $Z$, as $B \sim 1/t_0 \rightarrow 1/(Zt_0)$ and $C \sim 1/t_0^3 \rightarrow 1/(Zt_0)^3$. In Fig. 2, both DMFT and experimental results are compared with curves obtained by this simple rescaling of the noninteracting $B$ coefficient by $Z$. The small deviations observed up to 250–300 K (i.e. for the coefficient $B$ [22]) can be ascribed to the $T$ dependence of the chemical potential and to smearings of the van Hove singularity due to correlation [18]. On the other hand, analytical calculations show that the simple dimensional renormalization cannot be applied to the coefficient $C$, since its value [23] also depends on frequency- and temperature-dependent scattering terms of $\Sigma$, which we found to be substantial in our DMFT calculation.

In conclusion, we have measured for the first time the Sommerfeld expansion (though for strongly correlated electrons) from $T_c$ to 500 K is a challenging result as it indicates that the low-energy fermionic excitations in both optimally doped cuprates can be described in terms of a renormalized Fermi liquid. This result calls for further work aimed at understanding its relation with the well-known deviations from the Fermi-liquid behavior attributed to the presence of a quantum critical point in the phase diagram of cuprates [24].

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16. Exact diagonalization with 6–7 sites has been used as impurity solver for DMFT calculations.
17. The hopping parameters adopted [18–20] are $t_0 = 250$ (350) meV (nearest-neighbor), $t_1/t_0 = -0.2 (-0.3)$ (diagonal), $t_2/t_1 = -0.5 (0.0)$ (second nearest neighbor) for Bi-2201 (Bi-2212).
22. For Bi-2201 (Bi-2212) one has $b(\omega_p)_{U=0} \approx 1.1 \times 10^{-6} \times 5 \times 10^{-8}$ K$^{-2}$. Once renormalized, these values become $b(\omega_p)_{\text{REN}} = b(\omega_p)_{U=0}/Z \approx 7.3 \times 10^{-7}$ (4.1 $\times 10^{-7}$) K$^{-2}$ for $Z = 0.15$ (Z = 0.12).
23. For Bi-2201 (Bi-2212) one has $c(\omega_p)_{U=0} \approx 9.4 \times 10^{-14}$ (2.7 $\times 10^{-14}$) K$^{-4}$. Once renormalized, these values become $c(\omega_p)_{\text{REN}} = c(\omega_p)_{U=0}/Z^2 \approx 2.8 \times 10^{-11}$ (1.7 $\times 10^{-11}$) K$^{-4}$ for $Z = 0.15$ (Z = 0.12).