Temperature dependence of the optical spectral weight in the cuprates: Role of electron correlations

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We compare calculations based on the Dynamical Mean-Field Theory of the Hubbard model with the infrared spectral weight \(W(\Omega, T)\) of \(\text{La}_{2-x}\text{Sr}_x\text{CuO}_4\) and other cuprates. Without using fitting parameters we show that most of the anomalies found in \(W(\Omega, T)\) with respect to normal metals, including the existence of two different energy scales for the doping- and the \(T\)-dependence of \(W(\Omega, T)\), can be ascribed to strong correlation effects.

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The rich and complex optical response of high-temperature superconducting cuprates (HTS) is still attracting broad interest in the scientific community. In particular, much recent work, both experimental\textsuperscript{1,2,3,4,5,6,7,8,9,10,11} and theoretical\textsuperscript{6,7,8,9,10,11} was focused on the temperature dependence of the Cu-O plane infrared spectral weight

\begin{equation}
W(\Omega, T) = \int_0^\Omega d\omega \sigma_1(\omega),
\end{equation}

where \(\sigma_1(\omega)\) is the real part of the in-plane optical conductivity. In principle \(W(\Omega, T)\) depends on both the cut-off \(\Omega\) and the temperature \(T\). When \(\Omega \rightarrow \infty\), the \(\Omega\)-sum rule for the optical conductivity is recovered. In this case \(W \propto n\), where \(n\) is the carrier density for a continuous system and therefore it is independent of \(T\). In a tight-binding model \(W(\Omega, T)\) is instead related to the carrier kinetic energy\textsuperscript{12} and then retains a temperature dependence. In this framework, the behavior of \(W(\Omega, T)\) provides crucial information on the energetic balance at \(T_c\). Indeed, it has been reported that the onset of superconductivity both in underdoped and in optimally doped \(\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+y}\) (BSCCO) is associated with a gain in kinetic energy\textsuperscript{13,14} as opposed to the slight kinetic energy loss in conventional BCS superconductors. Moreover, restricted sum rules, where in Eq. (1) \(\Omega \leq \omega_p\), with \(\omega_p\) the plasma edge, have been studied experimentally in BSCCO\textsuperscript{15} \(\text{YBa}_2\text{Cu}_3\text{O}_{7-y}\) (YBCO)\textsuperscript{15} and \(\text{La}_{2-x}\text{Sr}_x\text{CuO}_4\) (LSCO)\textsuperscript{16} both in the normal and in the superconducting phases. In the latter work, it has been shown that the quadratic \(T\)-dependence

\begin{equation}
W(\Omega, T) \simeq W(\Omega, 0) - B(\Omega)T^2,
\end{equation}

holds for any infrared cut-off \(\Omega\) between \(\sim 0.1\text{eV}\) and \(\sim 1\text{eV}\), both in LSCO and in a conventional metal like gold. In both systems, when \(\Omega\) increases, the \(T\)-dependent term obviously increases, while the thermal response \(B(\Omega)\) decreases. However, while in gold both \(W(\Omega, 0)\) and \(B\) are controlled by the same energy scale (a hopping rate \(t\)), in LSCO the former scale is by one order of magnitude larger than the latter one. Moreover, in LSCO a sizable temperature dependence of \(W(\Omega, T)\) is found unexpectedly even for \(\Omega \sim \omega_p\), while in gold \(B(\Omega \approx \omega_p) = 0\).

Expression (2) is easily recovered, at least for large cut-offs, if electron-electron interactions are neglected. If, for the sake of simplicity, one models the Cu-O plane with an effective single-band tight-binding model with nearest-neighbor hopping \(t\) only, one has \(W(\Omega \approx \omega_p, T) = -(\pi e^2)/(2d)E_{\text{kin}}\), where \(e\) is the electric charge, \(d\) is the dimensionality of the system and \(E_{\text{kin}}\) is the carrier kinetic energy. For a non-interacting system we can write \(E_{\text{kin}}\) as an integral over the density of states (DOS) and expand it \(\text{à la Sommerfeld},\) obtaining Eq. (2) with \(W(\omega_p) \propto E_{\text{kin}}(T = 0)\). Under these simplifying assumptions, \(E_{\text{kin}}(T = 0) \propto t\) and \(B \propto t^{-1}\) (for a quasi-flat DOS, e.g., \(B = \pi^2 e^2/(24t d)\)), so that both quantities are controlled by the same energy scale \(t\). As long as electronic correlations are neglected, this result cannot be significantly changed by more realistic models. It has been recently shown that the addition of a next-nearest neighbor hopping \(t'\) which controls the position of the Van Hove singularity cannot explain the observations in LSCO or BSCCO\textsuperscript{16}.

The above standard behavior of non-interacting systems, in which the same \(t\) controls both \(W(\Omega, 0)\) and \(B\), is manifestly at variance with experimental evidence in cuprates. In particular, in LSCO\textsuperscript{17} \(E_{\text{kin}}(T = 0)\) leads to a \(t_0\) of the order of several hundreds meV, consistent with independent estimates of the bandwidth based on photoemission spectra or band-structure calculations\textsuperscript{18}. On the other hand, the experimentally determined value of \(B\) is unexpectedly large and leads to \(t_T \simeq 20\text{meV}\),\textsuperscript{17} smaller than \(t_0\) by one order of magnitude. As it was proposed in Ref.\textsuperscript{17} the existence of two distinct energy scales points towards the separation between low- and high-energy physics that is characteristic of strongly correlated electronic systems, and suggests that the large difference between \(t_0\) and \(t_T\) can be due to the proximity to a Mott insulator.
The present paper is aimed at putting this hint on firmer grounds by calculating the optical conductivity of a strongly correlated single-band Hubbard model, by Dynamical Mean-Field Theory (DMFT). We are aware that this approach will not provide a complete description of the optical properties of cuprates, where spatial correlations and other effects definitely play a role. However, we want to understand whether the Coulomb repulsion, even in its simplest reliable description, provides the basic ingredient to explain the one-order-of-magnitude discrepancy between the $T = 0$ spectral weight and the thermal coefficient $B$. We will assume reasonable values for the parameters of the calculation, but in this context it would be of little significance to finely tune them in order to fit the experimental results. Those values, indeed, should be certainly modified as other effects beyond DMFT were included.

The Hamiltonian of the Hubbard model reads

$$\mathcal{H} = -t \sum_{<ij>,\sigma} c_{i\sigma}^\dagger c_{j\sigma}^\vphantom{\dagger} + U \sum_i \left( n_{i\uparrow} - \frac{1}{2} \right) \left( n_{i\downarrow} - \frac{1}{2} \right) + \mu \sum_i (n_{i\uparrow} + n_{i\downarrow}),$$

(3)

where $c_{i\sigma}(c_{i\sigma}^\dagger)$ are annihilation(creation) operators for fermions of spin $\sigma$ on site $i$, $n_{i\sigma} = c_{i\sigma}^\dagger c_{i\sigma}$, $t$ is the hopping amplitude, $U$ is the local Hubbard repulsion, $\mu$ is the chemical-potential and the sum in the first term is restricted to nearest-neighbors only.

In order to solve (3) we resort to the DMFT, presently the most reliable tool to treat the physics of doped Mott insulators. DMFT is based on the neglect of spatial fluctuations, but fully retains local quantum dynamics, and density-driven Mott transitions. The DMFT maps the lattice model onto an impurity model subject to a semi-circular DOS of half-bandwidth $D$. Despite deliberately excluding phases with given spatial arrangement, DMFT allows for the simultaneous description of low- and high-energy features, and has provided the first unified treatment of the correlation- and density-driven Mott transitions. The DMFT maps the lattice model onto an impurity model subject to a self-consistency equation which contains the information about the original lattice. Here we consider, as it is often done, an infinite-coordination Bethe lattice with a semi-circular DOS of half-bandwidth $D$. The impurity model has been solved by Exact Diagonalization at finite temperatures. The number of levels in the bath has been fixed to $N_b = 4 \div 5$, which, despite being small, gives a good description of thermodynamic properties, at least at intermediate temperatures. Following Ref. 15 we assume $D = 4t = 1.2eV$, and $U = 3D = 12t$, so that the antiferromagnetic superexchange $J = 4t^2/U \simeq 100$ meV is compatible with experimental estimates. As we discuss in the following, our choice is related to the message we aim to bring with this paper.

Before turning to the analysis of optical properties, we briefly remind how the strongly correlated phases arising by doping a Mott insulator are characterized within DMFT. For large enough $U/D$, the half-filled system is a Mott insulator, with a spectrum made by two broad incoherent features, the Hubbard bands, which are centered around $\pm U/2$ with a width of the order of the bare bandwidth. As the system is doped, a quasiparticle (QP) peak appears around the Fermi level, whose weight is measured by the residue $Z = (1 - \partial \Sigma(\omega)/\partial \omega|_{\omega=0})^{-1}$, $\Sigma(\omega)$ being the (momentum-independent) self-energy. $Z$, which tends to vanish when the Mott transition is approached, is small in the strongly correlated regime, reflecting the loss of metallic behavior due to the Coulomb repulsion. In the hole-doping case which we discuss here, the QP peak is located close to the lower Hubbard band.

The knowledge of the single-particle spectral function can be directly used to compute the optical conductivity (and the associated spectral weight) because vertex corrections vanish in the infinite coordination limit in which DMFT becomes exact and $\sigma_1(\omega)$ (summed over all the spatial directions) is simply given by

$$\sigma_1(\omega) = \frac{2e^2}{\pi} \int d\epsilon \int d\nu N(\epsilon)V(\epsilon)[\text{Im}[G(\epsilon, \nu)]$$

$$\times \text{Im}[G(\epsilon, \nu + \omega)] f(\nu) - f(\nu + \omega)$$

(4)

where $N(\epsilon)$ is the DOS of the non-interacting system, and $V(\epsilon) = (D^2 - \epsilon^2)/3$ is the square current vertex of the infinite-coordination Bethe lattice we are considering. $G(\epsilon, \nu)$ and $G(\epsilon, \nu + \omega) = (\omega - \epsilon + \mu + \Sigma(\omega))^{-1}$ is the retarded Green functions of the lattice system.

The behavior of $\sigma_1(\omega)$, already discussed in Refs. 17,18 clearly mirrors the above described spectral features of a doped Mott insulator, and presents three main features, namely (i) a Drude contribution at low frequencies (say up to 0.1$D$) arising from optical excitations within the QP resonance, (ii) a mid Infrared (MIR) peak (at $\omega \sim 0.5 \div 1.0D$) resulting from the transitions between the lower Hubbard band and the unoccupied part of the quasi-particle peak and, finally, (iii) an insulating-like contribution at higher frequencies (at $\omega \sim U$) mainly due to transitions between the Hubbard bands.

Now we discuss how the above described behavior reflects in the temperature dependence of the optical spectral weight $W(\Omega, T)$. As it has been done for experimental data, we consider two values of the cut-off: $\Omega = 0.1D$, which is expected to include in the integral only the contribution from the Drude peak, and $\Omega = 1.5D$, which is in a sense the plasma edge of our model, i.e., contains both the Drude weight and the MIR bands, excluding from the integral the small but finite high-energy contributions.

In Fig. 1 the ratio $W(T)/W(0)$ is plotted for both the above $\Omega$ values and for different doping levels. One can see that our model calculation nicely follows the $T^2$ behavior in the range $0.008D < T < 0.022D$ (i.e., between 100 and 300K), as experimentally found in cuprates. The agreement with observations is not limited to the $T^2$ law, but extends to two important properties which...
TABLE I: Results of the fits in Fig. 1 for selected doping values. First and second columns: values of the coefficient $B$ (in units of $1/D$) for low- and high-frequency cut-off, respectively (note that, for $U = 0$, $B \approx 2/D$ in the whole doping range here considered). Third and fourth columns: corresponding spectral weights. Last two columns: kinetic energy and quasiparticle weight.

| $x=0.07$ | $58 \pm 5$ | $27 \pm 3$ | $0.03D$ | $0.11D$ | $0.17D$ | $0.12$ |
| $x=0.12$ | $48 \pm 5$ | $16 \pm 4$ | $0.08D$ | $0.10D$ | $0.21D$ | $0.20$ |
| $x=0.15$ | $43 \pm 5$ | $15 \pm 4$ | $0.09D$ | $0.18D$ | $0.22D$ | $0.22$ |
| $x=0.19$ | $51 \pm 5$ | $13 \pm 5$ | $0.11D$ | $0.20D$ | $0.24D$ | $0.27$ |
| $x=0.26$ | $33 \pm 7$ | $11 \pm 3$ | $0.14D$ | $0.24D$ | $0.27D$ | $0.35$ |

are not accounted for by non-interacting calculations. Namely, we find that (i) the inclusion of strong correlations determines a substantial increase in the coefficient $B$ controlling the thermal excitations, and that (ii) the enhancement is more pronounced for the lower value of $\Omega$. The results for $B(\Omega = 0.1D, 1.5D)$ and $W_0(\Omega) = W(\Omega, T = 0)$ determined by fitting the numerical data with Eq. $2$ are reported in Table I. For the sake of comparison we also report the $T = 0$ values of the quasiparticle weight $Z$ and for the full kinetic energy $E_{\text{kin}}$, i.e., $W(\Omega \to \infty, T = 0)$. The large value of $B$ reproduced by the DMFT can be mainly ascribed to the renormalized effective bandwidth $2D^* = 22D$ determined by correlations. If however the same effective bandwidth controlled also the zero-temperature spectral weight in our calculation, a strong inconsistency between theory and experiment would appear for this last quantity, whose experimental value is compatible with photoemission estimates of the hopping parameter, in turn of the order of the bare hopping.

The explicit calculation of $W_0(\Omega = 1.5D)$ is instead in very good agreement with the experiments, as shown in Fig. 2. Therein the experimental points are obtained by integrating the $\sigma_1(\omega)$ of LSCO reported for different doping in Ref. [19]. Fig. 2 shows that, even if the coherent hopping scale is significantly reduced by correlation, $W_0$ is much less affected and it is still large due to the contribution of the MIR band. Indeed it involves incoherent hopping processes which contribute to the average kinetic energy, even if they have no role in $\Omega^F$ transport.

It is worth to notice that theoretical and experimental points clearly follow a similar behavior as a function of doping, with a decreasing weight when the Mott insulator is approached.

In this regard, we expect that the inclusion of spatial correlations beyond DMFT and of the antiferromagnetic superexchange $J$ would result in a non vanishing renormalized bandwidth, as predicted, e.g., by slave boson mean-field theories. At the same time, once the DMFT approximation is relaxed, vertex corrections have to be considered in the optical conductivity. The fact that in Fig. 2 the calculated values are systematically lower than the experimental ones reflects our choice to fix $U/D$ without adjusting its value to data. Anyway, the ability of the present DMFT calculation to give the correct order of magnitude of $W_0$ suggest that the effect of the superexchange and the inclusion of vertex correction may partially cancel each other, as it indeed happens within zero-temperature slave boson approaches.

In the final part of this paper we like to consider a quantity suitable to measure the variation with temperature of the optical spectral weight at large $\Omega$. Denoting here as $W(T)$ the spectral weight $W(\Omega, T)$ for $\Omega = \omega_p$ for the experiments and $\Omega = 1.5D$ for the calculation, we plot in Fig. 3 the relative variation of $W(T)$ between $T = 0$ and $T = 300 \text{ K}$ $\Delta W/W_0 = \frac{W(T) - W_0}{W_0}$.
FIG. 3: The relative variation of the spectral weight between the lowest $T$ and 300 K, as a function of doping for various DMFT calculations (squares) and with the predictions of non-interacting models (tight binding model (boh line), flat band (boh-boh line)). The dashed line is a guide to the eye. The simple inclusion of correlation effects allows one to reproduce the observed absolute values with no need for fitting parameters. Data for LSCO are obtained from Refs. 5,19, for BSCCO from Ref. 2 (BSCCO) and from Ref. 1 (BSSCO).

$[W(300K)−W(0)]/W(0)$. In the same Figure, we report for comparison the prediction of a non interacting nearest neighbor model (dotted line, see also Ref. 10). Fig. 4 clearly shows that: i) such ratio has similar values in different families of cuprates, indicating that this quantity is essentially material-independent; ii) the thermal change of $W$ does not depend on details of the Fermi surface, which are instead dependent on both material and doping; iii) DMFT, with the introduction of strong correlation effects, reconciles the calculations with the experimental infrared behavior of the cuprates, which in no way can be reproduced by more conventional hopping models.

In conclusion, we have shown that the inclusion of strong correlation effects, already in the simplest theoretical scheme given by the Hubbard model, removes the inconsistency of non-interacting models and naturally accounts for the bifurcation between a $T=0$ spectral weight which becomes rapidly comparable with the non-interacting value, and the coefficient of its temperature dependence where a renormalized effective hopping plays the major role. This neither means that the Hubbard model in the DMFT approximation can account for all properties of cuprates, nor that our analysis has the ambition to be quantitative. However we believe that a non-perturbative treatment of correlations, such as the DMFT approach, is a basic building block for a theory of the optical response and of the transport properties of high-$T_c$ materials. In our view, the next step would be the inclusion of spatial correlations by means of non-local extensions of the DMFT such as Dynamical Cluster Approximation (DCA) or Cellular DMFT. In the latter approach, the effective bandwidth is no longer bound to vanish at the Mott transition, as it should also depend on the antiferromagnetic exchange coupling $J$. As mentioned above, we finally expect that the effect of the band broadening induced by $J$ will be partially compensated by vertex corrections to the optical conductivity. These improvements would ultimately lead to a better understanding of the role played by charge and spin modulations in the optical response of cuprates.

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We notice that the enhancement of $B$ is larger than $1/Z$, suggesting a possible role for finite lifetime effects.