Sum rules and electrodynamics of high-\(T_c\) cuprates in the pseudogap state.

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We explore connections between the electronic density of states (DOS) in a conducting system and the frequency dependence of the scattering rate \(1/\tau(\omega)\) inferred from infrared spectroscopy. We show that changes in the DOS upon the development of energy gaps can be reliably tracked through the examination of the \(1/\tau(\omega)\) spectra using the sum rules discussed in the text. Applying this analysis to the charge dynamics in high-\(T_c\) cuprates we found radically different trends in the evolution of the DOS in the pseudogap state and in the superconducting state.

One of the most enigmatic properties of cuprate high-temperature superconductors is the pseudogap in the spectrum of low-energy excitations developing primarily in weakly doped materials at a temperature \(T^*\) well above the superconducting transition \(T_c\). \[1\] First discovered through experiments probing spin-related behavior the pseudogap also leads to a characteristic modification of a variety of properties in the charge sector. Some aspects of the pseudogap have lead to an interpretation of this phenomenon in terms of a "precursor" of the superconducting gap whereas another school of thought is proposing scenarios based on spin- or charge-density wave fluctuations. \[2\] Using the novel sum rule analysis of the optical constants of cuprates measured for \(\Theta\) and (0; \(\pi\)) regions. Nevertheless, there are profound consequences of the pseudogap state for the in-plane carrier dynamics in many cuprates. The spectroscopic signatures of the pseudogap are best resolved in the spectra of the frequency dependent scattering rate \(\tau(\omega)\):

\[
\frac{1}{\tau(\omega)} = \frac{\omega_p^2}{4\pi} \text{Re} \left( \frac{1}{\sigma(\omega)} \right),
\]

where \(\omega_p\) is the plasma frequency and \(\sigma(\omega) = \sigma_1(\omega) + i\sigma_2(\omega)\) is the complex conductivity. \[4\] In the underdoped compounds (left panels in Fig. 1) the spectra of \(1/\tau(\omega)\) are depressed at \(T < T^*\) and reveal a characteristic threshold structure at \(\Theta \simeq 500 - 600\) cm\(^{-1}\). \[2\] At \(\omega < \Theta\) the scattering rate measured at \(T < T^*\) increases faster than linear with a crossover to a less rapid (nearly linear) increase at frequencies above the \(\Theta\) threshold. This behavior is universal and is observed in a large variety of materials. \[2\] As doping progresses to optimal and overdoped similar depression of \(1/\tau(\omega)\) occurs only at \(T \ll T_c\) (right panels in Fig. 1).

Conventional interpretation of this results is based on phase space arguments: a partial gap reduces the amount of final states available for scattering of the nodal quasi-particles therefore leading to a depression of \(1/\tau(\omega)\) for energies below the pseudogap. The above argument is corroborated by a close correspondence between the spectra of the scattering rate extracted from IR optics and the form of the electronic self-energy obtained from the angular resolved photoemission spectroscopy (ARPES). \[1\] In this paper we propose a new analysis suggesting a direct relationship between the behavior of \(1/\tau(\omega)\) and the prominent features of the electronic density of states (DOS) \(D(\omega)\). To elaborate on a connection between the spectra of \(1/\tau(\omega)\) (Eq. 1) and the essential characteristics of the DOS in the studied materials we first turn to the BCS model. We calculated the conductivity of an \(s\)-wave superconductor both in the clean and dirty limits \[12\] and determined \(1/\tau(\omega)\) from Eq. 1. A striking result is that the form of \(1/\tau(\omega)\) reproduces the key features of \(D(\omega)\) in a BCS superconductor including the gap at \(2\Delta\) followed by a sharp peak (Fig. 2). The divergence of the actual DOS at the gap edge is not found in \(1/\tau(\omega)\). Also, the peak in \(1/\tau(\omega)\) may occur somewhat above the gap energy and its location depends on the strength of (impurity) scattering. Nevertheless, the overall character of the single-particle density of states is reproduced in the \(1/\tau(\omega)\) spectra. Similar connection also holds for \(d\)-wave superconductors. \[13\]

An intriguing attribute of the model spectra displayed in Fig. 2 is revealed by the integration of \(1/\tau(\omega)\). It appears that the area removed from the intra-gap region is fully recovered due to the overshoot of the spectra at \(\omega > 2\Delta\). The reason for the above "sum rule" can be clarified by the following considerations. Expressing \(\sigma(\omega)\) Eq. 1 through the dielectric function \(\epsilon_1(\omega) + i\epsilon_2(\omega)\) one finds \(1/\tau(\omega) = [\omega_p^2/\omega](\epsilon_2/(\epsilon_1^2 + \epsilon_2^2 + 1 - 2\epsilon_1))\). The term on the RHS in the latter expression is to be compared with the integrand in the well-known sum rule \[14\]:

\[
\int_0^\infty \frac{d\omega}{\omega} \frac{\omega}{\epsilon_1 + \epsilon_2} = \frac{\pi}{2}
\]

The difference between the two terms arising from \(1 - 2\epsilon_1\) in the denominator of the \(1/\tau(\omega)\) expression results in less than 1% correction at frequencies \(\omega < 0.5\omega_p\). Therefore the balance between the areas associated with the intra-gap region and the overshoot in the \(1/\tau(\omega)\) spectra that can be expressed as:

\[
\int d\omega[1/\tau^A(\omega) - 1/\tau^B(\omega)] \simeq 0
\]
is in fact expected from the sum rule arguments. In Eq. 3 indexes $A$ and $B$ refer to different states of the studied system (e.g.: normal, pseudogap, superconducting). If the $\frac{\pi}{2}$ sum rule (Eq. 2) and Eq.3 are applied to the results plotted in Fig. 2 this analysis provides an additional support for the idea of the direct correspondence between the $1/\tau(\omega)$ spectra and the $D(\omega)$. Indeed, the BCS model suggests that the states removed from the intra-gap region are recovered at energies above the gap. This fundamental conservation is also manifested in the behavior of $1/\tau(\omega)$ (Fig. 2). Moreover, in those situations when $[1 - 2\varepsilon_1] \ll \varepsilon_1^2 + \varepsilon_2^2$ the DOS conservation can be quantitatively verified using Eq. 3.

In Fig. 3 we show an experimental example confirming connections between the structure seen in $1/\tau(\omega)$ spectra and the features of the DOS. We studied the response of the single crystals of Cr which is a spin-density wave (SDW) antiferromagnet with the Neel temperature $T_N = 312$ K. The results of the scattering rate analysis are reported in Fig. 3 for the first time. We find that at $T > T_N$ the absolute value of $1/\tau(\omega)$ increases as $\omega^2$ in accord with the Fermi-liquid (FL) theory. At 10 K the SDW gap is fully developed giving rise to a non-trivial form of the $1/\tau(\omega)$ spectra. The scattering rate is suppressed at $\omega < 500$ cm$^{-1}$ but then overshoots the 320 K spectrum with a maximum at $\omega \approx 900$ cm$^{-1}$. This behavior is similar to the results for the optimally doped cuprates plotted in Fig. 1. The main difference is that cuprates show a linear "background" in $1/\tau(\omega)$ as opposed to the $\omega^2$ background seen in the data for Cr. It is appropriate to compare the overshoot in the $1/\tau(\omega)$ spectra with the calculations for a BCS superconductor since the BCS theory is believed to produce an accurate representation of the DOS in a SDW system. While unmistakable similarities are revealed by such a comparison, it is important to keep in mind that in Cr only a part of the Fermi surface is affected by the SDW state. Therefore the gap in the DOS is incomplete which may account for a more gradual increase of $1/\tau(\omega)$ in the vicinity of the gap energy compared to theoretical prescriptions in Fig. 2. As pointed out above, the conservation of the DOS may be reflected through the balance of the "intra-gap" and "overshoot" areas (Eq. 3) provided the $1 - 2\varepsilon_1$ correction is insignificant. We found that in Cr the latter correction amounts to about 0.1% of the $\varepsilon_1^2 + \varepsilon_2^2$ at $\omega < 10,000$ cm$^{-1}$. Thus it is hardly surprising that Eq.3 is fulfilled for Cr with the accuracy of 10% with integration limited to $\omega_c = 1500$ cm$^{-1}$. Similar behavior of $1/\tau(\omega)$ spectra is also found in charge-density wave materials as well as in the heavy fermion materials revealing hybridization gap.

It is apparent from Fig.2 and Fig. 3 that the spectra of $1/\tau(\omega)$ capture the gross characteristics of the density of states in a conducting system especially in those situations when the DOS is (partially) gapped. We now return to the data for cuprates focusing on the implications of Eqs. 2-3 for the understanding of the charge response across the phase diagram. As pointed out above, the dominant features of the $1/\tau(\omega)$ spectra obtained for underdoped crystals in the pseudogap state are similar to those seen at $T \ll T_c$ in the optimally doped samples. On a closer examination one finds differences as well. Suppression of $1/\tau(\omega)$ at $\omega < \Theta$ in the pseudogap state does not lead to the development of the overshoot between the spectra measured at $T \ll T^*$ and $T > T^*$. The data for the optimally doped crystals taken at $T \ll T_c$ and $T \approx T_c$ does reveal an overshoot at $\omega \approx 800$ cm$^{-1}$. In the latter case, the area under the overshoot is balanced out by the reduction of $1/\tau(\omega)$ at low energies with the accuracy better than 10% in Tl$_2$Ba$_2$CuO$_6$ crystal and about 15% in the YBa$_2$Cu$_3$O$_{6.95}$ sample. This balance is expected from Eqs. 2-3 because the contribution of $1 - 2\varepsilon_1$ term is about 0.1% of $\varepsilon_1^2 + \varepsilon_2^2$ for either of the above superconductors. Thus the form of the $1/\tau(\omega)$ spectra for the optimally doped system is in accord with the notion of the transfer of the states from the intra-gap region to a peak at $\omega > \Theta$. This behavior can be naturally attributed to the opening of the superconducting energy gap. Although, the absolute values of the optical constants for the underdoped compounds are also in the regime when the Eq. 3 ought to be satisfied, data gives no indications even for a partial recovery of the area associated with the far IR depression of $1/\tau(\omega)$ is the pseudogap state.

The fact that in cuprates the spectra taken above and below $T^*$ "violate" Eqs. 2-3 within the frequency range of Fig. 1 signifies the differences in the changes of the low energy DOS in the pseudogap state from the changes associated with superconductivity. Essentially, the analysis described above indicates that the states removed from $\omega < \Theta$ at $T < T^*$ disappear from the energy interval reliably sampled in our experiments (0.5 eV). This result is in accord with the tunneling studies of the temperature dependence of the electronic DOS in underdoped cuprates. Indeed, data obtained at $T \ll T_c$ reveals a transfer of the electronic states from the intragap region in the quasiparticle peaks just above the gap. On the contrary, the formation of the pseudogap only leads to a depletion of DOS without visible traces of recovery of $D(\omega)$ at least at $h\omega < 0.1$ eV. Specific heat measurements are also in accord with our findings since these experiments suggest a reduction of the DOS in the pseudogap state within the energy window accessible to thermodynamic probes (\approx 500 K). Yet, another indication of an energy scale extending beyond 0.1-0.5 eV that is involved into the pseudogap state response is provided by the IR studies of the interlayer $c$-axis conductivity $\sigma_c(\omega)$. Non-conservation of the low-energy spectral weight $N_{eff}(\omega) = \int_0^\infty d\omega' \sigma_c(\omega')$ for $\omega < 0.2 - 0.4$ eV is inferred from the oscillator strength sum rule these $c$-axis results parallel our findings deduced from the examination of the in-plane scattering rate using Eqs. 2-3.
Thus one can conclude that both spectroscopic and thermodynamic studies of cuprates reveal anomalous changes of $D(\omega)$ in the pseudogap state distinct from the DOS effects associated with superconductivity. Hence, these results argue against a common origin of the pseudogap state and superconducting state.

Further support for the distinct genesis of the pseudogap and of superconducting gap is provided by the data for La$_{2-x}$Sr$_x$CuO$_4$ (La214) and Nd$_{2-x}$Ce$_x$CuO$_4$ (Nd214) materials. The scattering rate analysis of the in-plane response of these systems reveals a characteristic threshold structure at $\Theta \approx 500-600 \text{ cm}^{-1}$. In the double- or triple-layered materials the development of superconducting gap occurs approximately at the same energy as the pseudo-gap structure. However, in La214 and Nd214 compounds the signatures attributable to superconductivity are confined to energies below 40-50 cm$^{-1}$, whereas the pseudogap feature is essentially identical to what is seen in the double- layered compounds with $\Theta \approx 500 - 600 \text{ cm}^{-1}$. It is difficult to account for the difference by more than one order of magnitude between the energy scales associated with the pseudogap and with superconductivity if both effects result from the same microscopics. The formation of the pseudogap is commonly discussed in the context of the density of states such as Cr argues against a common origin of the pseudogap and of superconducting gap occurs approximately at the same energy as the pseudo-gap structure. However, in La214 and Nd214 compounds the signatures attributable to superconductivity are confined to energies below 40-50 cm$^{-1}$, whereas the pseudogap feature is essentially identical to what is seen in the double- layered compounds with $\Theta \approx 500 - 600 \text{ cm}^{-1}$. It is difficult to account for the difference by more than one order of magnitude between the energy scales associated with the pseudogap and with superconductivity if both effects result from the same microscopics. The formation of the pseudogap is commonly discussed in the context of the density of states such as Cr argues against a common origin of the pseudogap and of superconducting gap occurs approximately at the same energy as the pseudo-gap structure. However, in La214 and Nd214 compounds the signatures attributable to superconductivity are confined to energies below 40-50 cm$^{-1}$, whereas the pseudogap feature is essentially identical to what is seen in the double- layered compounds with $\Theta \approx 500 - 600 \text{ cm}^{-1}$. It is difficult to account for the difference by more than one order of magnitude between the energy scales associated with the pseudogap and with superconductivity if both effects result from the same microscopics. The formation of the pseudogap is commonly discussed in the context of the density of states such as Cr argues against a common origin of the pseudogap and of superconducting gap occurs approximately at the same energy as the pseudo-gap structure.
instance, impurities are known to suppress the QP peak as shown for Bi2212 compounds through tunneling measurements (S. Davis (unpublished)).

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**FIG. 1.** Spectra of the scattering rate determined for a variety of cuprates. Left panel displays data for underdoped materials: YBa$_2$Cu$_3$O$_6$[2] and Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$[5]. Right panel shows data for the optimally doped compounds: YBa$_2$Cu$_3$O$_{6.95}$[2] and Tl$_2$Ba$_2$CuO$_{6+\delta}$[6]. Solid lines: $T = 10$ K, gray lines: $T \simeq T_c$, dashed lines: $T = 300$ K. The in-plane response of underdoped materials in the pseudogap state is characterized by a depression of $1/\tau(\omega)$ at $\omega < 500$ cm$^{-1}$. In the optimally doped materials similar depression is observed only in the superconducting state.

**FIG. 2.** The frequency dependence of the scattering rate (Eq. 1) for a BCS superconductor in clean and dirty limit (black lines). In the normal state (gray lines) we assumed $1/\tau(\omega) = const$ in accord with the Drude model. Area balance in $1/\tau(\omega)$ spectra holds irrespective of the absolute values of the scattering rate. This attribute of the model spectra is exemplified in the right bottom panel displaying integrals of $1/\tau(\omega)$ at $T = T_c$ and $T = 0$ as a function of the cut-off frequency.

**FIG. 3.** Top panel: $1/\tau(\omega)$ spectra for Cr. At $T > T_N$ data follows the form $1/\tau(\omega) \propto \omega^2$ (dashed line) in agreement with the Fermi liquid theory. At $T = 10$ the SDW gap is fully developed leading to non-trivial form of the $1/\tau(\omega)$ spectrum described in the text. Inset shows the conductivity spectra.
Figure 1
Fig. 3