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Electron-energy loss spectra and plasmon resonance in cuprates

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The consequences of the non-Drude charge response in the normal state of cuprates and the effect of the layered structure on electron-energy loss spectra are investigated, both for experiments in the transmission and the reflection mode. It is shown that in the intermediate doping regime the plasmon resonance has to be nearly critically damped as a result of the anomalous frequency dependence of the relaxation rate. This also implies an unusual low-energy dependence of the loss function. Both facts are consistent with experiments in cuprates. Our study based on the *t*-*J* model shows good agreement with measured plasmon frequencies.
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In metals the investigation of the plasmon resonance is used as a standard tool to extract information about charge carriers. As is the case with several other electronic properties, the plasmon response also is quite unusual in cuprates. This can be realized from the dielectric measurements[1] which reveal a very broad plasmon resonance in the electron-energy loss (EEL) function and hence a poorly defined plasma frequency ω_p. One of the experimental approaches for measuring the plasmon dispersion $\omega_p(\mathbf{q})$ is the electron-energy-loss spectroscopy (EELS).[2] So far the EELS has not been employed very frequently in cuprates, also due to the ambiguities in the interpretation. The EELS in the transmission mode has been performed mostly for $Bi_2Sr_2CaCu_2O_8$ (BSCCO),[3,4] showing an optic plasmon with a quadratic dispersion. Similar spectra, restricted to $q=0$, have been obtained for $La_{2-x}Sr_xCuO_4$ (LSCO) by analyzing the dielectric reflectivity.[5] Recently higher-resolution EELS spectra have been measured[6] for BSCCO in the reflection mode, displaying an acousticlike plasmon at small wave vectors $\mathbf{q}$. In both experiments the plasmon resonance appears heavily damped, although not overdamped.

So far there have been only few theoretical studies of EEL, i.e., relevant to the quite specific electronic structure of cuprates. It has been realized early, in analogy with the layered electron gas,[7] that weakly coupled layers of mobile electrons in CuO planar structures can lead, in addition to optic plasmons, also to acoustic plasmon branches.[8] Recently, an important relation of the EEL function and the related dielectric function $\epsilon(\mathbf{q},\omega)$ to the mechanism of the superconductivity in cuprates has also been stressed.[9]

Another important aspect follows from the observation that the charge dynamics in the normal state of cuprates is anomalous and not consistent with the Drude-type relaxation. Charge as well as spin fluctuations have been instead phenomenologically described within the marginal-Fermi-liquid (MFL) scenario.[10,11] In particular, the latter has been shown to apply in the optimum-doping regime to the optical conductivity $\sigma(\omega)$, which can be expressed within a generalized Drude form with an effective (MFL-type) relaxation rate $\tau^{-1}\sim 2\pi\lambda(\omega+\xi T)$.[11] More recently, the same anomalous dynamical behavior has been found in numerical studies within the *t*-*J* model at intermediate doping,[12–14] revealing a more precise form of this diffusion behavior. Closely related to the dielectric function $\epsilon(\mathbf{q},\omega)$ and the EELS in cuprates are calculations of the density fluctuations $N(\mathbf{q},\omega)$, studied within the *t*-*J* model both analytically[15] and numerically.[16,14]

The aim of this contribution is to discuss the consequences of the established non-Drude planar optical conductivity for the loss function and EELS experiments, both in the transmission and in the reflection mode. In particular we shall investigate the intrinsic damping of the plasmon mode.

The microscopic models, such as the Hubbard model and the *t*-*J* model,[17] studied in connection with electronic properties of cuprates and other materials with strongly correlated electrons, do not incorporate the long-range Coulomb interaction. The latter is necessary for the appearance of plasmon oscillations. It is quite straightforward to include the long-range interaction within the random-phase approximation (RPA), where the dielectric function $\epsilon(\mathbf{q},\omega)$ and the dynamical density susceptibility $\chi(\mathbf{q},\omega)$ can be represented as

$$\epsilon(\mathbf{q},\omega)=1+V_{\mathbf{q}}\chi_0(\mathbf{q},\omega),$$

$$\chi(\mathbf{q},\omega)=\frac{\chi_0(\mathbf{q},\omega)}{1+V_{\mathbf{q}}\chi_0(\mathbf{q},\omega)}, \qquad (1)$$

where $\chi_0(\mathbf{q},\omega)$ is the electron-density susceptibility which includes the short-range correlations. In cuprates the hopping-matrix element for electrons between layers is rather small, e.g., as manifested in the large resistivity anisotropy, so that $\chi_0(\mathbf{q},\omega)$ should depend only on the planar component $\mathbf{q}_\parallel$, i.e., $\chi_0(\mathbf{q}_\parallel,\omega)$. On the contrary, the Coulomb matrix element $V_{\mathbf{q}}$ still depends on the three-dimensional (3D) wave vector $\mathbf{q}=(\mathbf{q}_\parallel,q_z)$. The general expression for $V_{\mathbf{q}}$ has been given in Refs. 7 and 8. For simplicity we restrict our discussion to small $\mathbf{q}$, i.e., $q_zd<1$, $q_\parallel a<1$, where d,a denote the interlayer distance and the planar cell dimension, respectively. In this regime one gets

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$$V_{\mathbf{q}}=e^2/\epsilon_0(\epsilon_{\parallel}q_{\parallel}^2+\epsilon_{\perp}q_z^2), \tag{2}$$

where $\epsilon_{\parallel}$ and $\epsilon_{\perp}$ are planar and interlayer $\omega\to\infty$ dielectric constants, respectively, due to screening of nonconduction electrons. The loss function is given by[2]

$$I(\mathbf{q},\omega)=-\mathrm{Im}\frac{1}{\epsilon(\mathbf{q},\omega)}=V_{\mathbf{q}}\,\mathrm{Im}\,\chi(\mathbf{q},\omega). \tag{3}$$

In the following we shall focus on the long-wavelength limit $q\to 0$ and on the regime of higher frequencies $\omega\sim\omega_p$. Here it follows from the continuity equation,

$$\chi_0(\mathbf{q}_{\parallel},\omega)=\frac{iq_{\parallel}^2\tilde{\sigma}(\omega)}{\omega e^2}, \tag{4}$$

where $\tilde{\sigma}(\omega)$ is the (complex) planar conductivity, which includes short-range correlation effects.

Let us comment here on the validity of the RPA approximation Eq. (1) together with the relation (4). The main question is whether we treat the long-range Coulomb repulsion consistently with the short-range correlations. If we consider the regime $q\to 0$ only the long-range character of $V_{\mathbf{q}}$ is important and we recover the thermodynamic relation (for $q=q_{\parallel}\to 0$) $\epsilon(\omega)=1+i\tilde{\sigma}(\omega)/\epsilon_0\omega$, which incorporates correctly the correlation effects. In the same way one can argue for the validity of the analysis for finite but small q. A caution is however needed when $q_{\parallel}a\gtrsim 1$ (which is not the subject of this paper) where corrections to the RPA analysis could become substantial and moreover also short-range properties of the interaction $V_{\mathbf{q}}$ become dominant.

The optical conductivity $\sigma(\omega)=\mathrm{Re}\tilde{\sigma}(\omega)$ in cuprates has been studied extensively,[1] revealing anomalous behavior consistent with the MFL scenario.[10,11] There have also been numerous theoretical calculations of $\sigma(\omega)$ at $T=0$, using exact diagonalization of finite-size models for strongly correlated electrons in cuprates.[18] Here we restrict our attention to the t-J model,[17] which incorporates the interplay between antiferromagnetic (AFM) spin correlations governed by the exchange J and the motion of electrons governed by the hopping parameter t, and the exclusion of double occupation of sites. Note that within this model the only relevant parameters are J/t and the concentration c_h of holes introduced by doping. It is expected that the t-J model can represent (even quantitatively) quite realistic low-energy properties of electrons in cuprates.[14] Similar results should also follow for the Hubbard model, provided that the strong correlation regime $J\ll t, U\ll t$ is considered. It is established that the inclusion of the next-nearest-neighbor hopping t' can improve the agreement with experiments.[18] However there are indications that the qualitative behavior of $\sigma(\omega)$ is less sensitive to the introduction of t',[14] therefore we omit such terms in this study.

Recent numerical studies using the finite-temperature Lanczos method (for a review see Ref. 14) of the t-J model, with realistic parameters $J/t=0.3$, reveal results for $\sigma(\omega)$, which are quite consistent with experiments in the normal state of cuprates. In particular, it has been found in the intermediate doping regime, $0.1<c_h<0.3$, that in a broad frequency range $\omega<\omega^*\sim 3t$ $\sigma(\omega)$ follows a universal law,[13]

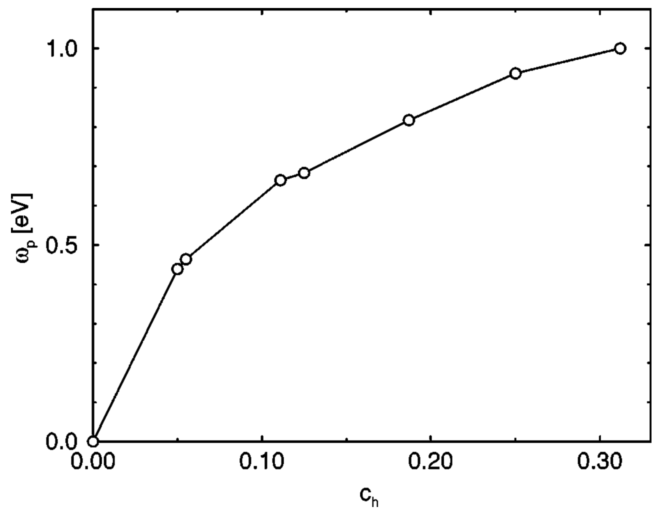

FIG. 1. Plasma frequency ω_p calculated within the t-J model assuming parameters relevant for LSCO: $t=0.4$ eV, $J=0.3$ t, $\epsilon_\infty=4$, and $d=0.66$ nm.

$$\sigma(\omega)=C_0\frac{1-e^{-\omega/T}}{\omega}, \quad \omega<\omega^*. \tag{5}$$

This anomalous diffusion has its origin in the fast decay of current-current correlations within a disordered spin background and seems to be generic for correlated systems,[19] at least at intermediate temperatures $T>T^*$.

On the other hand, in a tight-binding model one can always express the complex conductivity $\tilde{\sigma}(\omega)$ in terms of a memory function $M(\omega)$ as

$$\tilde{\sigma}(\omega)=\frac{ie^2\mathcal{K}t/d}{\omega+M(\omega)}, \quad \mathcal{K}=-\frac{E_{kin}}{2Nt}, \tag{6}$$

where $\mathcal{K}$ is the dimensionless kinetic-energy density (per cell) at a given T. For the case of a weak scattering $|M(\omega_p)|\ll\omega_p$, Eqs. (1), (2), (4), and (6) yield the plasma frequency

$$\omega_p=\sqrt{\frac{e^2\mathcal{K}t}{\epsilon_0\epsilon_{\parallel}d}}. \tag{7}$$

$E_{kin}=\langle H_{kin}\rangle$ is quite well established at $T=0$ from small-system studies,[18] and it is expected to vary only slightly at low $T<J$.[14] Note that at very low doping, e.g., $c_h<0.05$, assuming independent holes one expects $E_{kin}\propto c_h$. For higher doping the behavior remains qualitatively similar, although the slope $d|E_{kin}|/dc_h$ is reduced. Taking into account our numerical values for E_{kin} obtained for $J=0.3t$ and systems with $N=16,18,20$ sites, as well as the corresponding number of holes $0<N_h<5$, we get the results for $\omega_p(c_h=N_h/N)$ shown in Fig. 1. In the evaluation of Eq. (7) we assume $t\sim 0.4$ eV (Ref. 17) and use parameters relevant for LSCO, i.e., $\epsilon_{\parallel}=4$ and $d=0.66$ nm.[1,5]

It is evident that Eq. (5) cannot be modeled with small $M(\omega)$. It is useful to present $\tilde{\sigma}(\omega)$ in the generalized Drude form,[1]

$$\tilde{\sigma}(\omega)=\frac{i\omega_p^2}{\eta(\omega)[\omega+i\gamma(\omega)]} \tag{8}$$

with an effective relaxation rate (inverse relaxation time) $\gamma(\omega)$ and the mass renormalization $\eta(\omega)$,

$$\gamma(\omega)=\frac{M''(\omega)}{\eta(\omega)}, \quad \eta(\omega)=1+\frac{M'(\omega)}{\omega}. \tag{9}$$

Since $\eta(\omega_p)\neq 1$, it would be more appropriate to define $\omega_p^*=\omega_p/\sqrt{\eta(\omega_p^*)}$, which should more directly apply to the $q_z=0$ plasmon resonance, relevant for optical mesurements and for the transmission EELS. Our numerical $T>0$ results for $\sigma(\omega)$ and consequently for $M(\omega)$ (Refs. 13 and 14) reveal, however, quite consistently for doping concentrations $0.05<c_h<0.3$ that $\eta(\omega_p)\lesssim 1$, hence $\omega_p^*\sim\omega_p$.

A peculiar feature of the charge dynamics in cuprates is the non-Drude behavior of $\sigma(\omega)$, e.g., as represented by the MFL scenario,[10] or more specifically by Eq. (5), which should be valid at least near optimal doping. Within the broader MFL concept the dependence $\gamma(\omega,T)$ (Refs. 11 and 12) is given by

$$\gamma=\tilde{\lambda}(\omega+\xi T), \quad \tilde{\lambda}=2\pi\lambda. \tag{10}$$

This behavior is obeyed in cuprates up to $\omega\gtrsim\omega_p$. The dimensionless constant $\tilde{\lambda}$ obtained from numerical studies is not small. Estimates for various compounds fall in the range $0.5<\tilde{\lambda}<0.9$.[14]

Although $\tilde{\lambda}$ appears as a free parameter within the MFL proposal, this is not the case with Eq. (5), which also exhibits the MFL variation [Eq. (10)] apart from logarithimic corrections. Assuming that $\sigma(\omega>\omega^*)=0$, one can analytically evaluate $\tilde{\sigma}(\omega)$ and consequently $M(\omega)$. It is easy to express γ in two regimes:

(a) For $\omega\ll T\ll\omega^*$ the expansion in ω yields

$$\tilde{\lambda}=\frac{\pi}{3+2\ln T/\omega}, \quad \xi=2, \tag{11}$$

(b) while for $T\ll\omega\ll\omega^*$ we get

$$\tilde{\lambda}=\frac{\pi}{2(1+\ln\omega/T)}. \tag{12}$$

For the relevant range of T in the normal state of cuprates and the experimental range of ω (with reliable results), Eq. (12) should apply to experiments. The best overall fit of the numerical results[12,14] for $T\ll t$ and $\omega<t$ is achieved with $\tilde{\lambda}\sim 0.6$ and $\xi\sim 2.7$, also quite consistent with experiments.[1]

If we insert the generalized Drude expression (8) and Eq. (2), into Eqs. (4) and (1), we get

$$\epsilon(\mathbf{q},\omega)=1-\frac{\tilde{\omega}_p^2(\mathbf{q})}{\eta(\omega^2+i\omega\gamma)}, \tag{13}$$

where $\tilde{\omega}_p(\mathbf{q})$ is the effective plasmon frequency for a layered system,

$$\tilde{\omega}_p(\mathbf{q})=\omega_p\frac{q_\parallel}{\sqrt{q_\parallel^2+(\epsilon_\perp q_z/\epsilon_\parallel)^2}}. \tag{14}$$

Note that for $q_\parallel<q_z$ the plasmon shows an acoustic dispersion, i.e., $\tilde{\omega}_p=\omega_p q_\parallel\epsilon_\parallel/q_z\epsilon_\perp\propto q_\parallel$.[7,8]

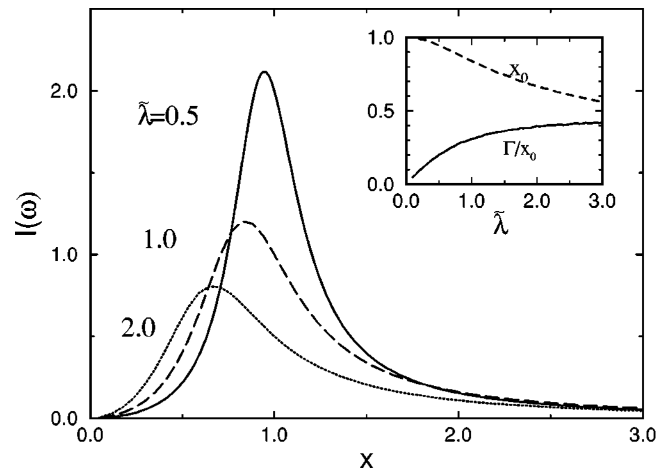

FIG. 2. Loss function I [Eq. (16)] vs $x=\omega/\tilde{\omega}_p$ for three different $\tilde{\lambda}$ values, i.e., $\tilde{\lambda}=0.5$, 1.0, and 2.0. The inset shows the ratio Γ/x_0 (solid curve) between the half-width Γ at half maximum measured at the low energy side of the peak and the renormalized plasma frequency $x_0=\omega_{max}/\tilde{\omega}_p$ (dashed curve).

With Eq. (13) the EEL function (3) can be written as

$$I(\omega)=\frac{\gamma\eta\omega\tilde{\omega}_p^2}{(\eta\omega^2-\tilde{\omega}_p^2)^2+\gamma^2\eta^2\omega^2}. \tag{15}$$

Let us restrict our attention again to the intermediate doping regime where relations (5) and (10) apply. In the range $\omega>T$, mostly relevant to experiments and for the study of the plasma resonance in particular, we insert $\gamma\sim\tilde{\lambda}\omega$ and assume $\eta(\omega)\sim 1$. Note that these simplifications could be possibly violated in the extreme acoustic limit $\tilde{\omega}_p(\mathbf{q})\to 0$. The result is

$$I(\omega)=\frac{\tilde{\lambda}x^2}{(x^2-1)^2+\tilde{\lambda}^2x^4}, \quad x=\frac{\omega}{\tilde{\omega}_p}. \tag{16}$$

It is evident from expression (16) that the width of the plasmon resonance is entirely determined by $\tilde{\lambda}$, i.e., the resonance width is $\Delta\omega/\tilde{\omega}_p\sim\tilde{\lambda}$. A further characteristic feature is the anomalous variation below the resonance, i.e., $I(\omega<\tilde{\omega}_p)\propto\omega^2$.

Figure 2 presents $I(\omega)$ for three different values of $\tilde{\lambda}$. For a typical value $\tilde{\lambda}\sim 1$ this implies that the plasmon resonance is necessarily very broad, i.e., the half-width of the peak of $I(\omega)$ is of the order of the plasma frequency itself. This seems to be a new aspect of the anomalous behavior of strongly correlated electrons in doped AFM and cuprates. In addition, there is a noticeable downward shift of the peak position with respect to $\tilde{\omega}_p$.

Let us turn to the discussion of experimental results of cuprates. The loss function $I(\mathbf{q},\omega)$ has been determined directly by EELS measurements for BSCCO (Refs. 3 and 4) and for $YBa_2Cu_3O_7$.[20] The characteristic asymmetric shape of $I(\omega)$ and the strong damping of the plasmon peak, emerging from our analysis, appears to be consistent with these experiments

The determination of the loss function $I(\omega)$ from optical $q\to 0$ data turns out to be also difficult, in particular because of the limited spectral range of the reflectivity data which makes the Kramers-Kronig transformation less reliable. However the use of ellipsometric data appears to provide highly accurate results, which are fully consistent with the existing EELS data.[21,22] $I(\omega)$ determined for a number of high-T_c superconductors show a single broad peak in the energy range below 2 eV. A characteristic measure for the width of the peak, which can be easily compared with experimental data, is the half-width Γ taken at the low-energy side of the peak at x_0. For $\tilde{\lambda}=1.0$ we obtain $\Gamma/x_0\sim 0.31$ (see inset of Fig. 2). Typical values extracted from experimental loss function data are 0.35 for BSCCO, 0.40 for $YBa_2Cu_3O_7$, and 0.45 for $Tl_2Ba_2Ca_2Cu_3O_{10}$, i.e., consistent with the theoretical values for Γ/x_0. Furthermore it has been noted that the low-energy part of the loss function of all systems is quadratic in frequency, i.e., $I(\omega)\propto\omega^2$, nearly up to the plasma frequency. It has been conjectured that this quadratic frequency dependence is universally obeyed in all high-T_c superconductors,[21,22] although at lowest frequencies ($\omega < 0.2$ eV) the dependence seems to be linear.[23] We note that $I(\omega<\omega_p)$, as given by Eq. (15), shows indeed an approximative quadratic dependence, which is due to the anomalous damping [Eq. (10)], while at low frequency the ω dependence becomes linear, because of the T scale in Eq. (10).

As we have assumed equidistant planes, we shall focus here on the single-layer material LSCO for a quantitative comparison between theory and experiment. For moderate doping the plasmon peak in the loss function is found at 0.80 (0.83) eV for $x=0.10$ (0.20), respectively.[5] These values and $\Gamma/x_0\sim 0.3$ are in reasonable agreement with our numerical results. It is surprising, however, that the experimental plasma frequency is not monotonous in its doping dependence. For $x=0.34$ Uchida *et al.*[5] report $\omega_p=0.77$ eV. This discrepancy may be related to the interplay with interband excitations in the highly doped material, which are not contained in the t-J model. Yet it is remarkable that in a model where the physics scales with the hole concentration (which is rather small) the correct scale of the plasma oscillation can be obtained.[24]

In summary we have shown that the anomalous damping of the optical conductivity, as obtained within the t-J model, explains the universal line shape of the loss function observed in the optimal doping range.[21] In this regime the width of the plasmon peak is of the order of the plasmon energy itself. Model results give, besides the qualitative, also a good quantitative description of EEL and the plasmon resonance in cuprates, at least in the intermediate doping regime.

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