Close inspection of plasmon excitations in cuprate superconductors

Andrés Greco\textsuperscript{1,2}, Hiroyuki Yamase\textsuperscript{3}, and Matías Bejas\textsuperscript{1}

\textsuperscript{1}Facultad de Ciencias Exactas, Ingeniería y Agrimensura and Instituto de Física Rosario (UNR-CONICET), Av. Pellegrini 250, 2000 Rosario, Argentina

\textsuperscript{2}Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1, D-70569 Stuttgart, Germany

\textsuperscript{3}National Institute for Materials Science, Tsukuba 305-0047, Japan

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Abstract

Recently resonant inelastic x-ray scattering experiments reported fine details of the charge excitations around the in-plane momentum $\mathbf{q}_\parallel = (0,0)$ for various doping rates in electron-doped cuprates La$_{2-x}$Ce$_x$CuO$_4$. We find that those new experimental data are well captured by acoustic-like plasmon excitations in a microscopic study of the layered $t$-$J$ model with the long-range Coulomb interaction. The acoustic-like plasmon is not a usual plasmon typical to the two-dimensional system, but has a small gap proportional to the interlayer hopping $t_z$.

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**Introduction.** The x-ray scattering technique is nowadays commonly used in cuprate high-$T_c$ superconductors. X-ray diffraction and resonant x-ray scattering revealed a charge-order tendency around the in-plane momentum $\mathbf{q}_\parallel \approx (0.6\pi, 0)$ and $(0.5\pi, 0)$ in hole-doped cuprates (h-cuprates)\(^{1-3}\) and electron-doped cuprates (e-cuprates)\(^{4,5}\), respectively. Resonant inelastic x-ray scattering (RIXS) (Refs. 6–11) clarified a V-shaped dispersion of charge excitations around $\mathbf{q}_\parallel = (0, 0)$, which extends up to a few eV.

The coherent understanding of those experimental data has not been obtained. The charge-order tendency in h-cuprates was intensively studied theoretically\(^{12-18}\), but its origin is still under debate. For e-cuprates, a large-$N$ theory of the $t$-$J$ model\(^{19}\) found that charge excitations are characterized by a dual structure in energy space. In a low-energy region, typically with a scale less than the exchange interaction $J$, various bond-charge excitations are present. Among others, bond-charge excitations with a $d$-wave symmetry exhibit a softening around $\mathbf{q} = (0.5\pi, 0)$ (Refs. 19,21), consistent with the experiment data\(^{4,5}\). On the other hand, in a high-energy region, typically with a scale larger than $J$, plasmon excitations, i.e., collective on-site charge excitations, become dominant and explain the charge excitations observed around $\mathbf{q}_\parallel = (0, 0)$ (Refs. 19,22,23).

In the context of the $t$-$J$ model, both h- and e-cuprates are studied on an equal footing by changing a sign of the second nearest-neighbor hopping $t'$ (Refs. 24,25). However, the extension of the large-$N$ theory of the $t$-$J$ model to h-cuprates cannot capture the observed charge-order tendency around $\mathbf{q}_\parallel \approx (0.6\pi, 0)$ (Ref. 12). This may be because the charge-order tendency was observed inside the pseudogap state in h-cuprates, but the theoretical calculations\(^{19-21}\) were performed in a normal metallic state. Such calculations\(^{19-21}\) may be a reasonable description only for e-cuprates, where the pseudogap is absent or very weak\(^{26}\). On the other hand, as in the case of e-cuprates, the large-$N$ theory captures the high-energy charge excitations observed in h-cuprates\(^9\) in terms of plasmons\(^{23}\). The plasmon excitations, therefore, seem universal in both h- and e-cuprates. However, the origin of the high-energy charge excitations are under debate and mainly three different scenarios are proposed: (i) a certain collective mode near a quantum phase transition, which is specific to e-cuprates\(^7,27\), (ii) intraband particle-hole excitations\(^6,8,9\) present in both e- and h-cuprates, and (iii) plasmon excitations with finite out-of-plane momentum $q_z^{10,19,22,23}$, which should be present in both e- and h-cuprates\(^{19,22,23}\).

Recently, Cu $L_3$-edge RIXS experiments reported details of the high-energy charge exci-
tations for e-cuprates La$_{2-x}$Ce$_x$CuO$_4$ (LCCO). By using doping-concentration-gradient films, the authors in Ref. [11] reported fine details of the charge excitations as a function of doping, $q_z$, and $q_{∥}$, which offer a stringent test of the plasmon scenario advocated in Refs. [10,22,23]. We find that those detailed data are well understood in terms of acoustic-like plasmons obtained in the large-$N$ theory of the layered $t$-$J$ model.

**Model.** It is well known that cuprates are correlated electron systems and a minimal model of the CuO$_2$ planes is the $t$-$J$ model. To understand the high-energy charge excitations around $q_{∥} = (0, 0)$, the coupling between the adjacent planes is important as shown in a theoretical study, where a layered $t$-$J$-$V$ model was employed,

$$
H = -\sum_{i,j,\sigma} t_{ij} \tilde{c}_{i\sigma}^{\dagger} \tilde{c}_{j\sigma} + J \sum_{\langle i,j \rangle} \left( \vec{S}_i \cdot \vec{S}_j - \frac{1}{4} n_i n_j \right) + \frac{1}{2} \sum_{i,j} V_{ij} n_i n_j ,
$$

(1)

Here $\tilde{c}_{i\sigma}^{\dagger}$ and $\tilde{c}_{i\sigma}$ are the creation and annihilation operators, respectively, of electrons with spin $\sigma(=\uparrow, \downarrow)$ in the Fock space without any double occupancy, $n_i$ the electron density operator, and $\vec{S}_i$ the spin operator. The indices $i$ and $j$ run over the sites of a three-dimensional lattice. The hopping $t_{ij}$ takes a value $t$ ($t'$) between the first (second) nearest-neighbors sites on a square lattice, and $t_z$ between the adjacent planes. $\langle i,j \rangle$ indicates a pair of nearest-neighbor sites on the square lattice and the exchange interaction $J$ is considered only inside the plane because the out-of-plane exchange term is much smaller than $J$ (Ref. [29]). $V_{ij}$ is the Coulomb repulsion.

Treating the non-double occupancy constraint within a large-$N$ approximation, the electronic quasiparticles disperse in momentum space as

$$
\varepsilon_{k} = \varepsilon_{k}^{∥} + \varepsilon_{k}^{\perp}
$$

(2)

where the in-plane dispersion $\varepsilon_{k}^{∥}$ and the out-of-plane dispersion $\varepsilon_{k}^{\perp}$ are given by, respectively,

$$
\varepsilon_{k}^{∥} = -2 \left( \frac{t \delta}{2} + \Delta \right) \cos k_x \cos k_y - 4t' \frac{\delta}{2} \cos k_x \cos k_y - \mu ,
$$

(3)

$$
\varepsilon_{k}^{\perp} = 2t_z \frac{\delta}{2} \left( \cos k_x - \cos k_y \right)^2 \cos k_z .
$$

(4)

The functional form $(\cos k_x - \cos k_y)^2$ in $\varepsilon_{k}^{\perp}$ is frequently invoked for cuprates. Other forms for $\varepsilon_{k}^{\perp}$, however, do not change the qualitative features of our results. Although the
electronic dispersion looks like that in a free electron system, the hopping integrals $t$, $t'$, and $t_z$ are renormalized by doping $\delta$ because of electron correlation effects. In addition, the term $\Delta$ in Eq. (3), which is proportional to $J$, is the mean-field value of the bond variables introduced to decouple the exchange term through a Hubbard-Stratonovich transformation. The value of $\Delta$ is computed self-consistently together with the chemical potential $\mu$ for a given $\delta$.

The term $V_{ij}$ in the Hamiltonian (11) describes the long-range Coulomb interaction, which is given in momentum space by

$$V(q) = \frac{V_c}{A(q_x, q_y) - \cos q_z},$$

where $V_c = e^2 d (2\epsilon_\perp a^2)^{-1}$ and $A(q_x, q_y) = \alpha (2 - \cos q_x - \cos q_y) + 1$ with $\alpha = \frac{\bar{\epsilon}}{(a/d)^2}$ and $\bar{\epsilon} = \epsilon_\parallel/\epsilon_\perp$; $\epsilon_\parallel$ and $\epsilon_\perp$ are the dielectric constants parallel and perpendicular to the planes, respectively; $a$ and $d$ are the lattice constants in the planes and between the planes, respectively; $e$ is the electric charge of electrons.

In a large-$N$ scheme, the density-density correlation function is renormalized already at leading order and can describe collective charge excitations; see Ref. 22 for a full formalism of the correlation function. We compute the imaginary part of the density-density correlation function as a function of $q$ and $\omega$ for the parameters $t'/t = 0.30$, $t_z/t = 0.03$, $J/t = 0.3$, $V_c/t = 8$, and $\alpha = 3.2$. We consider 30 planes along the $z$ direction to get a reasonable resolution of the out-of-plane momentum transfer $q_z$, that is, our $q_z$ is given by $q_z = 2\pi n_z/30$ with $n_z$ being integer. The temperature is set zero.

Our results are compared with mainly the data in Ref. 11, where momentum is given in units of $(2\pi/a, 2\pi/b, 2\pi/c)$; $a$, $b$, and $c$ are lattice constants. In the present theory, on the other hand, momentum is measured in units of $a^{-1} = b^{-1} = d^{-1} = 1$. Since LCCO contains two planes in the unit cell, the distance between the adjacent planes is $d = c/2$. For instance, the momentum $q = (0.06, 0, 1.68)$ in Ref. 11 corresponds to $q = (0.12, 0, 0.32)\pi$ in the first Brillouin zone in our theory. We set $t = 750$ meV (Ref. 23) to describe energy in units of eV.

In the present theory, the long-range Coulomb repulsion and the in-plane electronic correlations are treated on an equal footing. Hence our theoretical scheme is different from that in Ref. 10. In Ref. 10, the in-plane charge susceptibility was calculated first by determinant quantum Monte Carlo in the three-band Hubbard model and after that the effect of the
FIG. 1: Plasmon dispersion as a function of in-plane momentum along \((\pi, \pi)-(0,0)-(\pi, 0)\) direction for (a) \(q_z = 0\), (b) \(5\pi/15\), and (c) \(\pi\). The doping rate is \(\delta = 0.17\). The broadening is taken as \(\Gamma = 10^{-2} t\) to make the dispersion sharper. Faint spectral weight below 0.8 eV corresponds to the particle-hole continuum and becomes broader with increasing \(q\). The insets in (b) and (c) magnify a very low-energy region around \(q_{||} = (0, 0)\). Plasmon dispersion as a function of \(q_z\) for \(q_{||} = (0.05, 0)\pi\) (d), \((0.2, 0)\pi\) (e), and \((0.8, 0)\pi\) (f).

Results and comparison with experiments. In Figs. 1 (a), (b), and (c), we show the plasmon dispersion along \((\pi, \pi)-(0,0)-(\pi, 0)\) direction for \(q_z = 0\), \(5\pi/15\) (close to an experimental value reported in Ref. 11), and \(\pi\), respectively. The result for \(q_z = 0\) (upper panel) describes the optical plasmon mode, which is in good agreement with the plasmon frequency observed in cuprates. Early experiments reported that the optical plasmon frequency increases with increasing doping, which is well reproduced in the \(t-J\) model. For finite values of \(q_z\) (middle and lower panels) the plasmon energy at \(q_{||} = (0, 0)\) suddenly drops. A close look at the results reveals the presence of a gap at \(q_{||} = (0, 0)\); see the insets in Figs. 1 (b) and (c). Although the gapless excitations at \(q_{||} = (0, 0)\), namely acoustic plasmons were discussed for finite values of \(q_z\) in Refs. 10 and 11, the inter-layer hopping \(t_z\) yields a finite gap at \(q_{||} = (0, 0)\) for a finite \(q_z\) (Ref. 22). From Figs. 1 (b) and (c), we predict a gap around 70 meV for LCCO, i.e., much smaller than approximately 300 meV reported for \(\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_4\) (NCCO). This smaller gap originates mainly from a small value of \(t_z/t = 0.03\) in the present theory for LCCO and seems compatible with the experimental data. Hence we consider...
that the experimental data is reasonably interpreted as *acoustic-like* plasmons, instead of acoustic plasmons.

Given that the crystal structures of LCCO and NCCO are the same, the reason why the value of $t_z/t$ in LCCO can become smaller than NCCO needs to be studied further, although a new data for NCCO ([Ref. 10](#)) suggests a gap smaller than 300 meV; see a supplemental material in Ref. [10](#). It is also interesting to explore how the plasmon energy with finite $q_z$ changes by controlling the interlayer distance, e.g., by uniaxial pressure or intercalation of some elements between the layers, because the present theory predicts that the acoustic-like plasmon energy is proportional to $t_z$ in a small $t_z$ region\(^{22}\).

As clarified in Ref. [23](#), a characteristic feature of the plasmon excitations appears in its substantial $q_z$ dependence for a small in-plane momentum, which sharply distinguishes from the usual intraband particle-hole excitations. Figures [1](#) (d), (e), and (f) show the plasmon dispersion as a function of $q_z$ for several choices of $q_{∥}$. As seen in Figs. [1](#) (a)-(c), the plasmon intensity becomes very weak for a small $q_{∥}$, but it is discernible in Fig. [1](#) (d) that the plasmon energy and its intensity show a clear dependence on $q_z$. This $q_z$ dependence was reported in the experiment\(^{10}\). On the other hand, both the energy and the intensity become less sensitive to $q_z$ for larger values of $q_{∥}$ as shown in Figs. [1](#) (e) and (f). Currently no experimental data is available for a larger value of $q_{∥}$.

To make closer comparisons with the experimental data in Ref. [11](#), we show in Figs. [2](#) (a) and (b) charge excitation spectra as a function of $ω$ at $q_{∥} = (0.12,0)π$ and $(0.22,0)π$ for $δ = 0.17$; experimental values of $q_z$ are finite and the charge excitation spectrum in $q_{∥}$-$ω$ space corresponds to Fig. [1](#) (b), where the acoustic-like plasmons are realized. A broadening $Γ = 0.12t$ is introduced phenomenologically in the calculations of the charge response. The peak energy at both momenta in (a) and (b) agrees very well with the experimental results (c) and (d), respectively. For a given value of $q_{∥}$ the experiment shows that the peak energy shifts upwards and, at the same time, the peak intensity decreases with decreasing $q_z$. As seen in (a) and (b) these two features are reproduced in the present theory. In addition, the difference of the peak energy between the two different values of $q_z$ decreases with increasing $q_{∥}$ [(c) and (d)], which is also captured in (a) and (b).

In Fig. [3](#) (a) we show the doping dependence of the peak energy at $q_{∥} = (0.12,0)π$. With increasing doping the peak energy increases monotonically in a way very similar to the experimental results [Fig. [3](#) (b)]. In the experimental results of Fig. [3](#) (b) the peak
FIG. 2: Charge excitation spectra as a function of $\omega$ at $q_\parallel = (0.12, 0)\pi$ (a) and $(0.22, 0)\pi$ (b) for $\delta = 0.17$ and two different momenta $q_z$. (c) and (d) are the corresponding experimental data from Figs. 2 (c) and (d) in Ref. 11. The arrow in (d) indicates excitations of charge origin. The peaks at $\omega \approx 0.3$ eV in (d) correspond to paramagnon excitations and the peaks at $\omega = 0$ in (c) and (d) are elastic; these features are out of a scope of the present work. While the in-plane momenta are set exactly the same as the experimental values, the values of $q_z$ are slightly different between our values and the experimental ones because we have a finite number of planes. For instance, $l = 1.68$ in the experiment corresponds to $q_z = 0.32\pi$ which is close to $q_z = 5\pi/15$ in the present calculations for 30 planes.

width decreases with increasing doping. This implies the suppression of incoherent features with doping. To reproduce this feature within the present theory, we have introduced a broadening $\Gamma$ (Ref. 23), which decreases with increasing doping. This $\Gamma$ mimics a broadening of the spectrum due to electron correlations obtained in a numerical study of the $t$-$J$ model.

Figure 4 (a) shows results at $q_\parallel = (0.12, 0)\pi$ and $(0.08, 0.08)\pi$ for doping rate $\delta = 0.18$, where the peak position practically coincides at those two momenta. The peak energy agrees with the experimental data shown in Fig. 4 (b) and the spectral shape of the peak would become essentially the same as the experimental data if the background is subtracted properly. In Fig. 4 (c) we show the doping dependence of the peak energy for those two momenta (solid lines), which well reproduces the experimental results (see symbols). While a different doping dependence was discussed in Ref. 11 for a large $q_\parallel$ such as $q_\parallel = (0.22, 0)\pi$, the present theory predicts essentially the same behavior also for $q_\parallel = (0.22, 0)\pi$ [black line
FIG. 3: (a) Charge excitation spectra as a function of $\omega$ at $q = (0.12, 0, 5/15)\pi$ for different doping rates. The broadening $\Gamma$ is assumed to decrease with increasing doping. (b) The corresponding experimental results from Fig. 4 (a) in Ref. [11]. Although $q_z$ changes a little by changing doping in the experiment [see Fig. 4 (g) in Ref. [11]], such a change is very small and our value of $q_z = 5\pi/15$ is appropriate for all doping.

In Fig. 4 (c) at least in a doping region between 0.11 and 0.18. In Fig. 4 (d) we plot the full width at the half-maximum extracted from Fig. 3 (a) as a function of doping, which agrees with the experimental data.

In Figs. 3 (a) and 4 (c) the plasmon energy decreases with decreasing doping. In particular, the plasmon energy goes to zero at half filling (see Fig. 7 in Ref. [22]) because of the underlying strong electron correlations inherent in the $t$-$J$ model. Similar behavior was discussed for the optical plasmon mode, namely for $q_z = 0$, in Ref. [36] in the context of the $t$-$J$ model. On the other hand, the conventional theory of metals predicts a finite plasmon energy at half-filling. Therefore, it is important to measure the doping dependence of the plasmon energy upon approaching half-filling to test the present theory.

Outlook. We have shown that the acoustic-like plasmons obtained in the layered $t$-$J$ model explain well the high-energy charge excitation around $q_{||} = (0, 0)$ for e-cuprates. For h-cuprates, however, it is not clear whether plasmons can be indeed present. The recent momentum-resolved electron energy-loss spectroscopy [37, 38] cast doubt on the presence of plasmons in the h-cuprate Bi$_2$Sr$_{1.9}$CaCu$_2$O$_{8+x}$. In those papers the authors claimed that the dynamical charge response is characterized by featureless and momentum-independent excitations, showing the absence of plasmons in h-cuprates. On the other hand, Ref. [9] re-
FIG. 4: (a) Excitation spectra at $q = (0.12, 0, 5/15)\pi$ and $(0.08, 0.08, 5/15)\pi$ for $\delta = 0.18$. (b) The corresponding experimental data from Fig. 4 (d) in Ref. 11. (c) Doping dependence of the peak energy at $q = (0.08, 0.08, 5/15)\pi$ (blue line), $(0.12, 0, 5/15)\pi$ (red line), and $(0.22, 0, 5/15)\pi$ (black line). (d) The peak width at $q = (0.12, 0, 5/15)\pi$ as a function of doping. The symbols (triangles and circles) in (c) and (d) are the experimental data from Figs. 4 (e) and (f) in Ref. 11, respectively.

ported dispersive high-energy charge excitations in h-cuprates La$_{2-x}$(Br,Sr)$_x$CuO$_4$ by RIXS measurements. While the $q_z$ dependence, which is crucial to the plasmon scenario$^{23}$, was not measured and the obtained dispersion was interpreted as incoherent charge excitations in Ref. 9, the observed dispersion as a function of in-plane momentum was explained in terms of the acoustic-like plasmons$^{23}$ similar to the present work. Our theory therefore implies that plasmons exist in both h- and e-cuprates in a rather symmetric way$^{23}$, although many properties such as the pseudogap, superconductivity, and antiferromagnetism exhibit a pronounced asymmetry$^{26}$ between those systems. Further RIXS experiments in h-cuprates are important to clarify whether the high-energy charge excitations are indeed plasmons.

It is natural to ask a possible connection between plasmons and the pseudogap mainly observed in h-cuprates. In the present theory, plasmon excitations practically decouple from the low-energy excitations$^{29}$. Hence if low-energy charge fluctuations are related to
the pseudogap in h-cuprates as frequently discussed in the literature\textsuperscript{39,40}, we expect that in contrast to Refs. 37 and 38, high-energy charge excitations may not be the major source of the strange metallic properties. In fact, high-energy charge excitations extend to higher doping and have a doping dependence different from the pseudogap, superconductivity, and the strange metallic behavior.

Conclusions. Recalling that cuprate high-$T_c$ superconductivity is realized by charge carrier doping into the Mott insulator, the solid understanding of the charge dynamics is definitely indispensable to the cuprate physics. We have found that the acoustic-like plasmons obtained in the layered $t$-$J$ model with the long-range Coulomb interaction explain even the fine details of charge excitations observed recently as a function of doping, in-plane and out-of-plane momenta for Ce-doped La$_2$CuO$_4$.

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