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► **To cite this version:**

Giulio Dr. de Filippis, Vittorio Cataudella, Andrey Dr. Mishchenko, Carmine Dr. Antonio Perroni, Naoto Nagaosa. Optical conductivity of doped Mott insulator: the interplay between correlation and electron-phonon interaction. 2009. hal-00400458

**HAL Id: hal-00400458**

**<https://hal.science/hal-00400458v1>**

Preprint submitted on 30 Jun 2009

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# Optical conductivity of doped Mott insulator: the interplay between correlation and electron-phonon interaction

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(Dated: July 1, 2009)

The optical conductivity (OC) of cuprates is studied theoretically in the low density limit of the  $t$ - $t'$ - $J$ -Holstein model. By developing a limited phonon basis exact diagonalization (LPBED) method capable of treating the lattice of largest size  $4 \times 4$  ever considered, we are able to discern fine features of the mid-infrared (MIR) part of the OC revealing three-peak structure. The two lowest peaks are observed in experiments and the highest one is tacitly resolved in moderately doped cuprates. Comparison of OC with the results of semianalytic approaches and detailed analysis of the calculated isotope effect indicate that the middle-energy MIR peak is of mostly magnetic origin while the lowest MIR band originates from the scattering of holes by phonons.

PACS numbers: 71.10.Fd, 71.38.-k, 02.70.Ss, 75.50.Ee

The way to disclose the nature of the high temperature superconductors lies on the understanding of the dynamics of the holes doped into a Mott insulator [1]. It is recognized that the dynamics of holes is governed by the interaction with magnetic subsystem as it was proved by the angle resolved photoemission spectroscopy of underdoped compounds [2]. There is also a growing number of evidences that a considerable coupling to lattice contributes to the properties of the holes too [3]. These major interactions are expected to leave fingerprints in the the OC of cuprates. However, the interpretation of even the basic features of the OC is controversial.

Not to say about fine structure, there is no agreement on the issue of how many peaks are seen in the OC, both theoretically and experimentally. Initially, only the Drude term and MIR peak at around 0.5 eV have been considered as contributions coming from the dynamics of charged carriers [4]. Later, improved quality of the samples and experimental techniques gave an indication [5, 6, 7, 8, 9], and finally clearly showed [10], that there is at least one more band (MIR<sub>LOW</sub> band) induced by doping in the energy range,  $\sim 0.1$ eV, which is just above the phonons energy. Moreover the analysis of the experimental data suggests that also another contribution, peaked at 1.5eV, should be also taken into account [6, 7, 8, 9]. Although there is a temptation to explain the later contribution as reminiscent of the charge-transfer peak in doped system, this third high energy peak (MIR<sub>HIGH</sub> band) is observed in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  at 1.5 eV which is considerably smaller than the peak energy at 2 eV, observed in undoped compound [6].

Even if the existence of a peak structure is recognized, its nature has been debated. Inability of the prototypical  $t$ - $J$  model, where hole moves in an antiferromagnetic background, to explain the experimental structure of OC attracted a significant interest on this problem. One pos-

sible direction consists in considering the Hubbard model with moderate  $U$ . The interpretation of the mid-IR peak, based on a purely electronic effect (associated with the upper/lower-Hubbard bands and some in-gap states induced by doping), has been proposed [11]. Recently it was concluded [12] that the moderate  $U$  can reproduce the OC spectra in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  by  $U \cong 4eV$ , which is in sharp contrast to the  $t$ - $J$  picture in the strong correlation limit. The other possible direction is to consider the additional electron-phonon interaction (EPI), i.e.,  $t$ - $J$ -Holstein ( $t$ - $J$ -H) model, where hole interacts also with dispersionless phonons. The OC of the latter model was calculated by several methods: exact diagonalization (ED) on the small  $\sqrt{10} \times \sqrt{10}$  system [13], Self-Consistent Born Approximation (SCBA) with respect to both phonons and magnons [14], Dynamical Mean-Field Theory (DMFT) for infinite dimensions [15], Diagrammatic Monte Carlo (DMC) with SCBA for magnons [10], and ED within the Limited Functional Space (EDLFS) [16]. The fine structure of the OC in realistic 2D systems can be studied only by DMC [10] and EDLFS [16] whereas the rest of approaches encounter severe problems [17]. In the light of this statement it is extremely alarming that interpretation of the low energy peak of OC (POC<sub>LOW</sub>) in [10] and [16] is different. Magnetic origin of POC<sub>LOW</sub> is concluded in [16] while the statement about phononic origin in [10] might be an error originated from the SCBA in magnetic channel or spin wave approximation used in [10]. Therefore, the convincing evidence for the origin of the POC is an urgent and important issue towards the understanding of the basic interactions governing high temperature superconductors.

In the present Letter we study theoretically the OC of  $t$ - $t'$ - $J$ -H model directly compared with the experimental observations supporting the vital role of EPI in cuprates. In addition to the inclusion of the realistic next near-

est neighbor hopping  $t'$  to reproduce the observed Fermi surfaces in cuprates [1], we avoid the spin wave and self-consistent Born approximations for the coupling to the spin system. By developing a LPBED method, we can calculate the OC of the largest ever considered  $4 \times 4$  system. Due to the exponential growth of the basis with size of the system, the  $4 \times 4$  lattice has considerably denser quantum states than the  $\sqrt{10} \times \sqrt{10}$  system so that it is possible to resolve fine structure of the OC. For the first time we observe, in different ranges of EPI, three peaks in OC, the highest one being seen because the spin-wave approximation is avoided. Calculating the isotope effect, which induces changes both in phonon frequency and exchange constant  $J$ , we show that, in the weak coupling regime, the low energy  $\text{POC}_{\text{LOW}}$  and the middle energy POC are of phononic and magnetic origin, respectively. Furthermore, in the intermediate coupling regime the low energy  $\text{POC}_{\text{LOW}}$  is still of purely phononic origin while the middle energy POC is a mixture of the lattice and magnetic excitations. Finally, comparison with the results of different approximate schemes shows that the highest energy  $\text{POC}_{\text{HIGH}}$  peak is due to incoherent transitions into the states unaffected by lattice deformation associated with the hole.

The Hamiltonian for t-t'-J-Holstein model is a sum of t-t'-J Hamiltonian

$$H_{tt'J} = -t \sum_{i,\delta,\sigma} c_{i+\delta,\sigma}^\dagger c_{i,\sigma} - t' \sum_{i,\delta',\sigma} c_{i+\delta',\sigma}^\dagger c_{i,\sigma} + \frac{J}{2} \sum_{i,\delta} S_{i+\delta} S_i - \frac{J}{8} \sum_{i,\delta} n_{i+\delta} n_i, \quad (1)$$

EPI Hamiltonian  $H_{h-ph} = \omega_0 g \sum_i (1 - n_i) (a_i^\dagger + a_i)$ , and dispersionless phonons Hamiltonian  $H_{ph} = \omega_0 \sum_i a_i^\dagger a_i$  ( $a_i^\dagger$  is the creation operator of a phonon at site  $i$  with frequency  $\omega_0$ ). Here  $t$  represents the hopping amplitude of the site  $i$  to nearest neighbors  $i + \delta$ ,  $t'$  is the diagonal hopping amplitude to next nearest neighbors  $i + \delta'$ ,  $J$  is the exchange constant of the spin-spin interaction,  $c_{i,\sigma}$  is the fermionic operator with excluded double occupancy,  $S_i$  is the  $\frac{1}{2}$ -spin operator at site  $i$ , and  $n_i$  is the site  $i$  number operator. We introduce the EPI dimensionless coupling constant,  $\lambda = g^2 \omega_0 / 4t$ , with the value  $\lambda = 1$  dividing the weak and strong coupling regimes of the Holstein model in the adiabatic limit. Below we set  $\hbar = 1$ ,  $t = 1$ ,  $J = 0.3$ ,  $t' = -0.25$ ,  $\omega = 0.15$  and OC is in units of  $2\pi e^2$ . The one-hole ground state of the t-J model on  $4 \times 4$  lattice is sixfold degenerate. This degeneracy between  $(\pm\pi/2, \pm\pi/2)$ ,  $(0, \pi)$  and  $(\pi, 0)$  is partially removed by  $t'$  [19] providing a four-fold degenerate ground state at momentum  $(\pm\pi/2, \pm\pi/2)$ . Hence, one can naively expect that the OC should be sensitive to the value of  $t'$ .

The LPBED method is based on the modified Lanczos algorithm [20], where magnetic degrees of freedom

are treated exactly whereas the phonon variables are efficiently limited to a set which, as it is shown below, gives better results than both the Momentum Average (MA) approximation [21] and SCBA. We use the translational symmetry associated to periodic boundary conditions, requiring that the states have a definite momentum, and work in the one-hole subspace with  $\sum_i S_i^z = \frac{1}{2}$ . Each basis vector is a linear superposition with appropriate phases of the 16 translational copies of a state having a given hole location with assigned locations of the phonon quanta and spin flips (hole, spin and lattice configurations are together rigidly translated). All 6435 spin configurations of  $4 \times 4$  lattice are included.

The real bottleneck comes from Hilbert space required by the phonons basis. For instance, if all the phonon configurations up to  $M=15$  phonon states are included the size of the system is strongly limited ( $\sqrt{10} \times \sqrt{10}$  [13]). To circumvent this difficulty, LPBED keeps only two groups of phonon states. The first group are the lattice configurations involving only *single site* deformations, all the others being undeformed  $|ph\rangle_j^{(n)} = (a_j^\dagger)^n |0\rangle_j [\sqrt{n!}]^{-1} \prod_{i \neq j} |0\rangle_i$ . Here  $j = 1, \dots, N$  denotes the lattice sites,  $|0\rangle_i$  is the  $i$ -site phonon vacuum state, and all possible  $n = 0, 1, \dots, M$  values are limited by  $M = 20$ , which is shown to be enough for convergence even in the strong coupling regime [13]. Restriction to such basis makes the method equivalent [22] to the MA approximation [21]. This is already a reasonable approximation, which satisfies exactly the first six spectral weight sum rules for the Lehmann spectral function [21]. To improve the MA scheme, LPBED method includes additional phonon states with up to three ( $n_1 + n_2 + n_3 \leq 3$ ) phonons on *different lattice* sites  $|ph\rangle_{j_1, j_2, j_3}^{(n_1, n_2, n_3)} = \prod_{h=1}^3 (a_{j_h}^\dagger)^{n_h} [\sqrt{n_h!}]^{-1} |0\rangle_{j_h} \prod_{i \neq j_1, j_2, j_3} |0\rangle_i$ . Such limit is chosen since three-phonon basis is able to recover the SCBA contribution (up to three phonons) and it goes beyond including all other processes of the same order which are not present in the SCBA approach [23]. Hence, LPBED approach is better than MA and SCBA methods and the only domain where it can fail is the case when there is a strong non-local deformation. However, strong deformations are realised only in the strong coupling regime where deformation in the case of local Holstein interaction is just restricted to the hole position. Our approach, able to improve MA, is related to that introduced in Ref. [24], with the advantage that in LPBED method it is possible to calculate not only self energy of the quasiparticle but any correlation function.

At  $T = 0$  the OC at nonzero frequency is calculated using the Kubo expression of the OC in terms of the current-current correlation function  $\sigma_{xx}(\omega) = -(N\omega)^{-1} \Im (\Pi(\omega + i\eta) + \Pi(\omega - i\eta))$  with  $\Pi(\omega + i\eta) = \langle \psi_0 | j_x [\omega + i\eta - H + E_0]^{-1} j_x | \psi_0 \rangle$ . Here  $|\psi_0\rangle$  is the ground state ( $\vec{k} = (\frac{\pi}{2}, \frac{\pi}{2})$ ) with energy  $E_0$ ,  $N$  indicates the number of lattice sites,  $\eta$  is a broadening

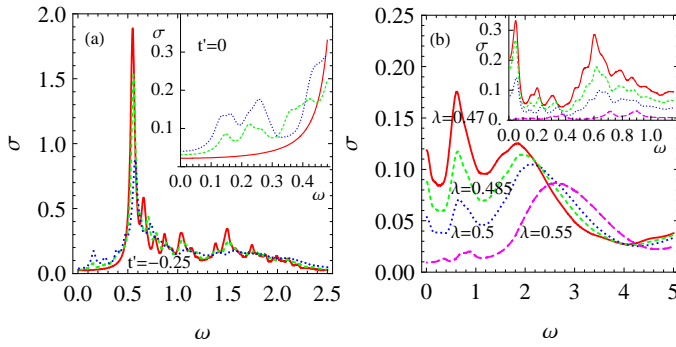


FIG. 1: (Color online) OC in (a) weak [ $\eta = 0.025$ ] coupling regime for  $t' = -0.25$  and (inset) for  $t' = 0$ . OC at  $t' = -0.25$  (b) from intermediate to strong coupling regime for  $\eta = 0.1$  and (inset)  $\eta = 0.025$ .

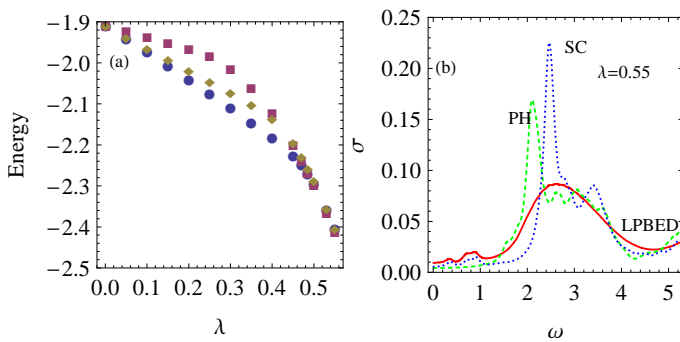


FIG. 2: (Color online) (a) Binding energy obtained within LPBED method (circles), MA method (diamonds) and SC scheme (squares). (b) OC in the strong coupling regime ( $\eta = 0.1$ ).

factor that shifts the poles of  $\sigma_{xx}(\omega)$  in the complex plane by replacing the  $\delta$  functions by Lorentians, and  $j_x = iet \sum_{i,\delta,\sigma} c_{i+\delta,\sigma}^\dagger c_{i,\sigma} (\vec{\delta})_x + iet' \sum_{i,\delta',\sigma} c_{i+\delta',\sigma}^\dagger c_{i,\sigma} (\vec{\delta}')_x$ .

For  $\lambda = 0$  we duly observe the well known POC in the t-J model at the energy  $2J$ : the presence of  $t'$  has little influence on the main feature of OC of t-t'-J model (Fig. 1a). At weak EPI, in agreement with [10], we detect  $\text{POC}_{\text{LOW}}$  just above the phonon frequency. We stress that the existence of  $\text{POC}_{\text{LOW}}$  is not related to  $t'$  hopping term, since the  $\text{POC}_{\text{LOW}}$  is also observed in the t-J-H model (inset in Fig. 1a). This low energy peak appears only at nonzero EPI and persists up to the strong coupling regime (inset in Fig. 1b). By increasing  $\lambda$ , the POC peak, which is around  $2J$  at  $\lambda = 0$ , shifts to higher energies and its weight is gradually transferred to the high energy  $\text{POC}_{\text{HIGH}}$  above  $\omega \geq 2t$ . Hence, the EPI changes the spectrum of the t-J model: OC exhibits three peaks. The nature of these peaks is either unknown or under dispute (cf. [10] and [16]). In the following we present several results unambiguously revealing genesis of these

peaks in the weak and strong coupling regimes.

To study the origin of the three peaks for large EPI, we introduce strong coupling (SC) adiabatic approach in which wave function is factorized into a product of normalized variational functions  $|\psi(\mathbf{r})\rangle$  and  $|\phi(\mathbf{R})\rangle$  depending on electron  $\mathbf{r}$  and phonon  $\mathbf{R}$  coordinates, respectively. The expectation value of the Hamiltonian on the state  $|\phi\rangle$  provides a Hamiltonian,  $H_{el}$ , depending only on the electronic degrees of freedom. It describes the t-t'-J model in a potential well. In Fig. 2a we compared results of SC approach for ground state energies of t-t'-J model with data obtained by LPBED method. For  $\lambda$  above the critical  $\lambda_c \approx 0.5$  the results of SC, MA, and LPBED approaches are in good agreement and, thus, the system is in the strong coupling regime. In the SC limit, according to Franck-Condon principle, the lattice is frozen in the ground state during the hole optical excitations and the OC can be calculated considering excitations of the hole in the static potential well formed by the lattice deformation. Comparison of SC result for OC with that obtained by LPBED for  $\lambda = 0.55$  (Fig. 2b) shows that SC approach reproduces all three peaks. We note that in the SC approach both initial and final electronic states are calculated in the lattice potential associated with the ground state wave function of the hole. On the other hand, if one assumes that the hole in the final state releases the deformation, we obtain another curve for OC with only the high energy  $\text{POC}_{\text{HIGH}}$ . Such approach is often called "photoemission" (PH) process because the lattice deformation is only present in one of the counterparts of the states linked by current operator. Comparing results from LPBED, SC, and PH approaches we conclude that  $\text{POC}_{\text{LOW}}$  and POC in the strong coupling regime represents hole transitions between states within the self-consistent potential well generated by the phonons. To the contrary,  $\text{POC}_{\text{HIGH}}$  is associated with transitions into states which are intact by the EPI driven lattice deformation. Here we note that the association of the theoretical  $\text{POC}_{\text{HIGH}}$  with experimental  $\text{MIR}_{\text{HIGH}}$  band should be done with care, since both structures are located at energies which are near the limit of applicability of the t-J model, where one is required to use methods which can handle the initial unreduced three-band Hubbard model [25]. We stress that the above discussed scenario is valid also in the more simple t-J-Holstein model.

Another way to establish the nature of the peaks of OC is to study changes of OC with small variations of the phonon frequency  $\omega_0$  and/or exchange constant  $J$ . Such study does not serve solely as Gedanken experiment but establishes how the spectra will be altered by the isotope substitution (IS). First of all, changes of the oxygen from  $^{16}\text{O}$  to  $^{18}\text{O}$  induces modifications in the values of coupling constant  $g$  and phonon frequency  $\omega_0$ . Phonon frequency  $\omega_0 = \sqrt{k/M}$  is expressed in terms of  $k$ , which is the restoring force for length unit of the local oscillators, and  $M$ , which is the mass of oxygen atoms surrounding the

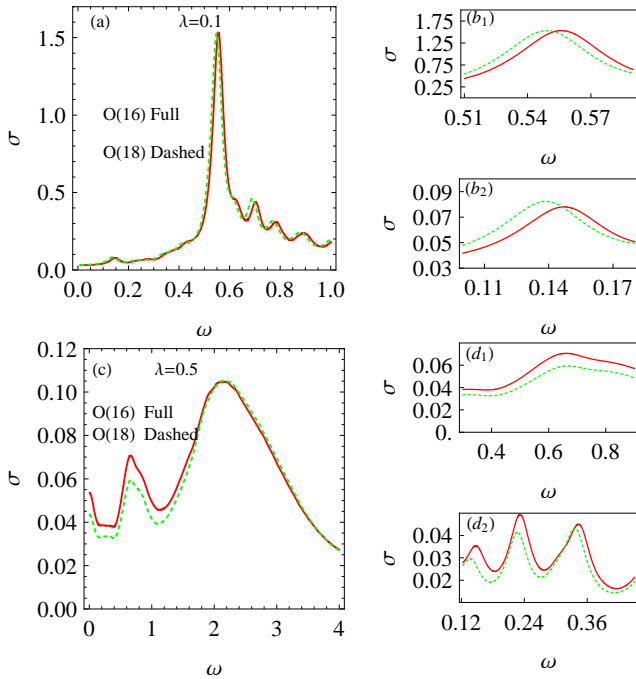


FIG. 3: (Color online) Effect of the O IS on OC in the weak [(a), (b<sub>1</sub>), and (b<sub>2</sub>)], and the intermediate coupling regime [(c), (d<sub>1</sub>), and (d<sub>2</sub>)].

Cu ion in the CuO<sub>2</sub> plain. IS changes the values of  $\omega_0$  and  $g$  to  $\omega_0^* = \omega_0 \sqrt{M/M^*}$  and  $g^* = g(M^*/M)^{1/4}$  with value of  $\lambda$  independent on isotope. In particular, the relative shift of  $\omega_0$  is about 6%. The second possible effect of IS is the decrease of the antiferromagnetic exchange constant  $J$  in compounds with the apical oxygen [26, 27], driven by its vibrations out of plane. With the IS the value of  $J$  is reduced by about 1% and in the following we assume  $\Delta J/J = -0.01$ .

In Fig.3 we present the changes of OC induced both by change of the in-plane oxygen mass and exchange constant  $J$ . The contribution of these changes are well distinct with only one exception. In the weak coupling regime (Fig. 3a and Fig. 3b<sub>1</sub>) POC at frequencies around  $2J$  is shifted down by about 1% indicating the clear magnetic origin of this peak in the weak coupling regime. To the contrary, POC<sub>LOW</sub>, with frequency around  $\omega_0$ , is shifted down about 6% (Fig. 3b<sub>2</sub>), that is just softening of  $\omega_0$  induced by IS. Thus, we get one more confirmation of the phononic origin of theoretical POC<sub>LOW</sub> and experimental MIR<sub>LOW</sub> band, in agreement with Ref. [10].

One gets the same conclusion about POC<sub>LOW</sub> from the IS in the intermediate coupling regime (Fig. 3c and Fig. 3d). Indeed the POC<sub>LOW</sub> is shifted down by 6% again. On the contrary, behavior of the middle energy POC differs from that in the weak coupling regime since it reduces its intensity and almost does not move with the IS. It is known that the middle POC energy increases by increasing  $\lambda/\lambda_c$  [10]. Moreover,  $\lambda_c$  decreases by de-

creasing  $\omega_0$  [28]. Thus, the decrease of  $\omega_0$  increases the energy of POC. To the contrary, decrease of  $J$  tends to soften the POC. Hence, joint influence of both effects leaves the peak at the same position. To confirm this conclusion we repeated the calculations setting to zero the change of  $J$  and found (not shown) the increase of POC energy. Hence, we conclude that in the intermediate coupling regime the middle POC is of mixed origin.

In conclusion, we developed a novel method capable of studying the fine structure of the OC of the t-t'-J-Holstein model and found that the influence of the diagonal hopping  $t'$  on OC is surprisingly little. For the first time we revealed a 3-peak structure and established the origin of these peaks in the whole range of hole-phonon couplings. We also predicted the influence of the IS on the OC which can be compared with experiment after the problem of residual <sup>16</sup>O in the matrix of <sup>18</sup>O, strongly influencing low energy part of OC [29], will be solved.

A.S.M. was supported by RFBR 07-02-00067a, N.N. by Grant-in-Aids No. 15104006, No. 16076205, No. 17105002, No. 19048015, and NAREGI Japan; G.D.F., V.C. and C.A.P. received financial support from Research Program MIUR-PRIN 2007.

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