Doping dependence of $d$-wave bond-charge excitations in electron-doped cuprates

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(Dated: August 13, 2018)

Abstract

Motivated by the recent experiments reporting the doping dependence of the short-range charge order (CO) in electron-doped cuprates, we study the resonant x-ray scattering spectrum from $d$-wave bond-charge fluctuations obtained in the two-dimensional $t$-$J$ model. We find that i) the CO is short-range, ii) the CO peak is pronounced at low temperature ($T$), although the peak structure itself survives even at high $T$, iii) the peak intensity increases with decreasing carrier doping $\delta$ down to $\delta \approx 0.10$ and is substantially suppressed below $\delta \approx 0.10$ due to strong damping, and iv) the momentum of the CO decreases monotonically down to $\delta \approx 0.10$ and goes up below $\delta \approx 0.10$. These results reasonably capture the major features of the experimental data, and the observed short-range CO can be consistently explained in terms of bond-charge fluctuations with an internal $d$-wave symmetry.

PACS numbers:
I. INTRODUCTION

Recently resonant x-ray scattering (RXS), resonant inelastic x-ray scattering (RIXS), and high-energy x-ray scattering revealed the presence of short-range charge order (CO) with modulation vector along the axial direction \((0, 0)-(\pi, 0)\) in various hole-doped cuprates (h-cuprates) such as \(Y_{1-5}\), Bi\(_{6-9}\), and Hg-based\(^{10,11}\) compounds, implying that the CO can be a universal phenomenon in h-cuprates. The understanding of the origin of those charge correlations, therefore, will likely yield an important clue to the origin of the pseudogap as well as high-\(T_c\) superconductivity\(^{12}\). In fact, a large number of theoretical studies were performed\(^{13-22}\), although a consensus has not been obtained.

On the other hand, a short-range CO was also observed in electron-doped cuprates\(^{23-25}\) (e-cuprates). Since the pseudogap features similar to those in h-cuprates are much weaker in e-cuprates, a theoretical study may be less complicated in e-cuprates. However, compared to theoretical studies of h-cuprates\(^{13-22}\), the CO in e-cuprates is much less studied. Ref.\(^{26}\) showed a comprehensive study of all possible COs in the two-dimensional (2D) \(t-J\) model and found a strong tendency to \(d\)-wave bond-charge order. Ref.\(^{27}\) then showed that \(d\)-wave bond-charge fluctuations can capture the charge excitation spectrum observed in experiments\(^{23}\). Although a theoretical framework is different from Refs.\(^{26}\) and \(^{27}\), similar \(d\)-wave bond-charge fluctuations were also proposed in Ref.\(^{28}\) to explain the experimental data.

The \(d\)-wave bond-charge order is different from a usual textbook-like charge-density-wave because the bond charge has an internal structure characterized by a \(d\)-wave symmetry. Therefore if the short-range CO observed in e-cuprates is indeed a \(d\)-wave bond-charge order, it can be interpreted as the first observation of unconventional CO in e-cuprates. Given that \(d\)-wave bond-charge order was discussed in h-cuprates\(^{29}\), it can be a universal phenomenon in the whole cuprate family. In addition, \(d\)-wave bond-charge order would be reduced to the electronic nematic order\(^{30}\), more precisely a \(d\)-wave Pomeranchuk instability\(^{31-33}\) when the momentum transfer approaches zero. In this sense, the nematic physics can also play a role in the charge dynamics in e-cuprates while the nematic physics is discussed only in h-cuprates so far\(^{34-37}\). Therefore it is very important to study whether recent experimental data in e-cuprates\(^{24}\), i.e., the doping dependence of the short-range CO, can be indeed captured in terms of \(d\)-wave bond charge, which will then provide a crucial step to establish the presence of \(d\)-wave bond-charge fluctuations in e-cuprates.
In this paper, we compute first the static charge susceptibility associated with \(d\)-wave bond-charge order and then the equal-time correlation function \(S(q)\), the quantity measured by RXS, in a large-\(N\) scheme of the \(t-J\) model. We show that our theoretical results capture the major features of the doping dependence of the recent RXS data\(^{24}\). The present calculations suggest the presence of unconventional charge fluctuations in \(e\)-cuprates, which are connected with the nematic order in the limit of momentum to zero.

II. MODEL AND FORMALISM

The \(d\)-wave bond charge is obtained in a non-bias study of the 2D \(t-J\) model by employing a large-\(N\) scheme\(^{13}\). We follow such a theoretical scheme and focus on the excitation spectrum of the \(d\)-wave bond charge. The 2D \(t-J\) model on a square lattice reads

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

where \(t_{ij} = t(t')\) is the hopping between the first (second) nearest-neighbors sites; \(J\) and \(V\) are the magnetic exchange and Coulomb interaction between the nearest-neighbors sites as denoted by \(\langle i,j \rangle\), respectively. \(\tilde{c}_{i\sigma}^{\dagger} (\tilde{c}_{i\sigma})\) is the creation (annihilation) operators of electrons with spin \(\sigma (=\uparrow, \downarrow)\) in the Fock space without any double occupancy. \(n_i = \sum_\sigma \tilde{c}_{i\sigma}^{\dagger} \tilde{c}_{i\sigma}\) is the electron density operator and \(\vec{S}_i\) is the spin operator.

Various approximations to the \(t-J\) model\(^{26,38,39}\) found a strong tendency toward phase separation especially for band parameters appropriate for \(e\)-cuprates. The phase separation, however, can be an artifact caused by discarding the Coulomb repulsion\(^{40}\). Therefore we have included the nearest-neighbor Coulomb interaction in the Hamiltonian (1) to suppress the tendency of the phase separation. From a more realistic point of view, we would include the interlayer hopping as well as the long-range Coulomb interaction. This is actually important when studying the high-energy charge excitation spectrum, which is dominated by plasmon excitations\(^{41,42}\). However, three dimensionality and the precise form of the Coulomb interaction are not important to low-energy charge excitations\(^{42}\) addressed in the present work.

It is not straightforward to analyze the Hamiltonian (1) because it is defined in the Fock space without double occupancy of electrons. Here we employ a large-\(N\) technique
in a path integral representation of the Hubbard $X$ operators, where the leading order approximation become exact in the limit of large $N$. With this formalism, all possible charge instabilities included in the Hamiltonian are treated on equal footing and were studied at leading order. In particular, for band parameters appropriate for $e$-cuprates, $d$-wave bond-charge instability is leading around the doping rate $\delta = 0.15$ (Ref. 26). Furthermore, its excitation spectrum captures the essential features of the experimental observation in $e$-cuprates at $\delta = 0.14$ and 0.15 (Ref. 23). Therefore to study the recent experimental data performed at different doping rates in $e$-cuprates, we focus on the charge excitations coming from $d$-wave bond-charge fluctuations. Since we deal with the $t$-$J$ model, which is derived from the three-band Hubbard model, bond-charge order can be interpreted as charge density wave at the oxygen sites because bond charge is defined on a bond between the nearest-neighbor Cu sites.

RXS measures the equal-time correlation function, which is defined by

$$S(q) = \frac{1}{\pi} \int_{-\infty}^{\infty} d\omega \text{Im}\chi_d(q, \omega) [n_B(\omega) + 1], \quad (2)$$

where $n_B(\omega) = 1/(e^{\omega/T} - 1)$ is the Bose factor and $T$ is temperature. The $d$-wave bond-charge susceptibility $\chi_d(q, \omega)$ is obtained in the large-$N$ expansion at leading order as

$$\chi_d(q, \omega) = \frac{(8J\Delta^2)^{-1}}{1 - 2J\Pi_d(q, \omega)}, \quad (3)$$

which becomes exact in the limit of large $N$. Here $\Delta$ is the mean-field value of a bond field and is given by $\Delta = \frac{1}{4N_s} \sum_{k}(\cos k_x + \cos k_y)f(\epsilon_k)$; the value of $\Delta$ is determined self-consistently. This bond field $\Delta$ naturally appears in our path integral formalism as a Hubbard-Stratonovich field. $N_s$ is the total number of lattice sites and $f(x) = 1/(e^{x/T} + 1)$ is the Fermi-Dirac distribution function. The electron dispersion $\epsilon_k$ is renormalized already at leading order in the large-$N$ expansion

$$\epsilon_k = -2\left(\frac{t}{2} + J\Delta\right)(\cos k_x + \cos k_y) - 4t'\frac{\delta}{2}\cos k_x \cos k_y - \mu, \quad (4)$$

where $t$ and $t'$ are reduced by a factor of $\delta/2$ and $\mu$ is the chemical potential. The $d$-wave polarization $\Pi_d(q, \omega)$ in Eq. 3 reads

$$\Pi_d(q, \omega) = \frac{1}{N_s} \sum_{k} \gamma^2(k)\frac{f(\epsilon_{k+q/2}) - f(\epsilon_{k-q/2})}{\epsilon_{k+q/2} - \epsilon_{k-q/2} - \omega - i\Gamma}, \quad (5)$$
where the $d$-wave form factor $\gamma(k) = (\cos k_x - \cos k_y)/2$ describes a $d$-wave symmetry of the bond-charge order and $\Gamma$ is infinitesimal small. In the limit of $q = 0$, $\chi_d(q, \omega)$ would be reduced to the electronic nematic susceptibility\textsuperscript{45} associated with a $d$-wave Pomeranchuk instability\textsuperscript{31–33}. In the following, we measure all quantities with the dimension of energy in units of $t$.

III. RESULTS

We choose $J = 0.3$ and $t' = 0.3$ in our Hamiltonian (1) as typical parameters for e-cuprates\textsuperscript{26}; the precise value of $V$ is not important as long as it suppresses phase separation. As a value of $\Gamma$ in Eq. (5) we take $\Gamma = 10^{-4}$, which should be reasonably small. In Fig. (a), we present the static part of the $d$-wave bond-charge susceptibility $\chi_d(q) = \chi_d(q, \omega = 0)$ as a function of $q$ for several choices of temperatures at $\delta = 0.13$. Note that $\chi_d(q)$ has $4\pi$ periodicity because of the presence of the $d$-wave form factor [see Eq. (5)] and thus the $q$ region is in $0 \leq q_x \leq 2\pi$ in Fig. (a). With decreasing $T$, a peak is pronounced at $q = (\pm 2\pi Q_{co}, 0)$ and $(0, \pm 2\pi Q_{co})$ with $Q_{co} \approx 0.25$, indicating a tendency toward a charge ordered phase. However, the static susceptibility does not diverge and the charge order remains a short range.

We show in Fig. (b) the equal-time correlation function $S(q)$ for the same parameters as in Fig. (a). While $S(q)$ has a peak at almost the same position of $\chi_d(q)$, the peak intensity is slightly suppressed with decreasing $T$ even though $\chi_d(q)$ shows a pronounced peak at low $T$ [Fig. (a)] (Ref. 46). This counter-intuitive feature comes from the presence of the Bose factor $n_B(\omega)$ in Eq. (2). In fact, if $n_B(\omega)$ were omitted in Eq. (2), $S(q)$ would show a peak, which is enhanced with decreasing $T$.

The suppression of $S(q)$ with decreasing $T$ in Fig. (b) can be an artifact of the present leading order theory of large-$N$ expansion, in the sense that we completely neglect the damping of quasiparticles, that is, we assume $\Gamma = 10^{-4}$ at any temperature. In addition, the damping effect should be pronounced at low doping because of the strong antiferromagnetic fluctuations. Therefore, we invoke a finite value of $\Gamma$ in Eq. (5) to simulate phenomenologically the damping of quasiparticles as a broadening of the spectrum. A value of $\Gamma$ may in principle depend on $T$, $\delta$, $q$, $\omega$, and others. Since our major interest is a study of temperature and doping dependences of $S(q)$, we consider only possible $T$ and $\delta$ dependences of $\Gamma$. 


As a function of $T$, a leading correction may be given by a linear term in $T$, i.e.,

$$\Gamma(\delta, T) = \Gamma(\delta) + \alpha T.$$  \hfill (6)

Concerning the doping dependence, we recall that neutron scattering experiments\(^\text{17}\) revealed that the antiferromagnetic correlation length starts to increase substantially below $\delta \approx 0.10$ in the normal metallic phase around $T \sim 300$ K. Concomitantly, quasiparticles may be damped heavily below $\delta \approx 0.10$. To mimic this phenomenology in a simple way, we assume a $\delta$ dependence of $\Gamma(\delta)$ as shown in Fig. 2, where $\Gamma$ increases rapidly below $\delta \approx 0.10$. An explicit expression is given by

$$\Gamma(\delta) = 0.001 + 0.05 \left[ 1 - \tanh \left( \frac{\delta - 0.09}{0.02} \right) \right].$$  \hfill (7)

In Fig. 2 we also plot the boundary of the $d$-wave bond-charge phase at $T = 0$. When $\Gamma$ is infinitesimally small and independent of doping, the model would exhibit the $d$-wave bond-charge instability at $\delta_c \approx 0.125$ (Refs. 26 and 48). With increasing $\Gamma$, the $d$-wave bond-charge phase shrinks. As a result, we have only charge fluctuations associated with the $d$-wave bond-charge order for doping above the dashed line in Fig. 2.

While the choice of the absolute value of $\Gamma$ is rather arbitrary in Eq. (7), we choose it to have no charge instabilities even at low doping rate at $T = 0$ (solid line in Fig. 2), so that our calculations are performed in the paramagnetic phase in the entire doping region. For a finite $T$, we choose $\alpha = 9$ in Eq. (6) after checking that our conclusions are not modified for other choices of $\alpha = 3$ and 6.
FIG. 2: Doping dependence of the damping $\Gamma$ (solid line) and the phase boundary of the $d$-wave bond-charge order at $T = 0$ (dashed line).

FIG. 3: (Color online) (a) $q$ dependence of $S(q)$ for several choices of temperatures at doping $\delta = 0.13$. (b) $\Delta S^{\text{peak}}$ as a function of temperature for various doping rates.

Figure 3(a) shows $S(q)$ along the direction $(0,0)-(2\pi,0)$ in a temperature range $0 < T < 0.035$ for $\delta = 0.13$. $S(q)$ forms a peak structure at $q \sim (0.5\pi,0)$ even at high $T$. To clarify how the peak of $S(q)$ develops, we define $\Delta S(q) = S(q; T) - S(q; T = 0.035)$. Since a realistic value of $t/2$ (Ref. 49) in cuprates is around 400 meV (Ref. 50), $T = 0.035$ corresponds to room temperature and thus may be regarded as a background. Hence $\Delta S(q)$ is regarded as $S(q)$ after background subtraction, as often seen in the experimental data analysis. We plot in Fig. 3(b) the temperature dependence of the peak intensity of $\Delta S(q)$.
for several choices of doping rates (> 0.10). The peak intensity $\Delta S^{\text{peak}}$ is pronounced upon decreasing temperature and doping rate, which is qualitatively the same as the experimental observation (see Fig. 2 G in Ref. 24).

With decreasing $\delta$ beyond $\delta \approx 0.10$, $S(\mathbf{q})$ is suppressed substantially as shown in Fig. 4 which is actually observed in experiments 24. This rapid suppression comes from the pronounced increase of the damping $\Gamma$ below $\delta \approx 0.10$ as shown in Fig. 2. If $\Gamma$ is assumed to be constant, the peak intensity of $S(\mathbf{q})$ would continue to increase with decreasing $\delta$.

As shown in Fig. 3(a), $S(\mathbf{q})$ exhibits a peak at $\mathbf{q} = (\pm 2\pi Q_{\text{co}}, 0)$ and $(0, \pm 2\pi Q_{\text{co}})$. This peak position is plotted in Fig. 4 as a function of doping rate at $T = 0$ together with $Q_{\text{edge}}$, the distance between the Fermi surface edges across $\mathbf{k} = (\pi, 0)$ and its equivalent wavevectors (see the inset in Fig. 4). As already pointed out in Ref. 27, the peak structure is formed by particle-hole scattering processes characterized by $Q_{\text{edge}}$. Hence $Q_{\text{co}}$ corresponds to such scattering wavevector $Q_{\text{edge}}$ at least down to $\delta \approx 0.10$, although it becomes slightly larger than $Q_{\text{edge}}$, since $S(\mathbf{q})$ is an energy-integrated quantity [see Eq. (2)]. Below $\delta \approx 0.10$, $Q_{\text{co}}$ goes up and deviates substantially from $Q_{\text{edge}}$. This is because the damping $\Gamma$ increases rapidly and the structure coming from the underlying Fermi surface is blurred. We also plot the experimental data of $Q_{co}$ (Ref. 24) in Fig. 4. The agreement with the experiment is very good in $\delta > 0.10$. While the experimental data seem to follow $Q_{\text{edge}}$ in $\delta < 0.10$, the present theory cannot reproduce such a tendency (see also Sec. IV).

![Graph](image.png)

FIG. 4: (Color online) Peak intensity of $S(\mathbf{q})$ and the momenta $Q_{\text{co}}$ and $Q_{\text{edge}}$ as a function of $\delta$ at $T = 0$. $Q_{\text{edge}}$ is defined in the inset. Solid circles are the experimental data from Ref. 24.
IV. DISCUSSIONS

The equal-time correlation function $S(q)$ can be measured directly by RXS. In particular, we have computed $S(q)$ associated with the $d$-wave bond-charge order, which is reduced to the nematic order in the limit of $q = 0$ (Refs. 31–33). A possibility of $d$-wave bond-charge order as well as the nematic order is already discussed in h-cuprates. Therefore it is a crucial step toward the understanding of the charge dynamics in cuprates to clarify to what extent our results capture recent RXS data in e-cuprates. Since the previous data of Ref. 23 was already discussed in Ref. 27, we focus on the recent experimental data given in Refs. 24 and 25.

$S(q)$ shows a peak structure at any temperature in Fig. 3(a) and the peak intensity is enhanced with decreasing temperature [Fig. 3(b)], but the CO remains short-range without instability toward the $d$-wave bond-charge phase (see also Fig. 2). These features are consistent with the experimental observation.

The peak position of $Q_{co}$ tends to be larger with increasing $T$ as shown in Fig. 3(a). In the experiments, however, the peak position is almost independent of $T$. This difference suggests additional corrections at high $T$ beyond the present leading order theory. On the other hand, $Q_{co}$ decreases with decreasing doping in the experiments. In particular, it tends to be constant in $\delta \lesssim 0.10$ if we do not consider seriously the data at $\delta = 0.059$, where the existence of a peak at $Q_{co}$ in Fig. 2(B) of Ref. 24 is unclear. We think that Fig. 4 reasonably captures the major feature of $Q_{co}$ observed in the experiments.

As shown in Fig. 4, the peak intensity of $S(q)$ gradually increases and sharply drops in $\delta \lesssim 0.10$. Therefore the CO signal is expected to become difficult to be detected in $\delta \lesssim 0.10$. Conversely, we expect that the CO signal is observed more clearly above $\delta \approx 0.10$, as is indeed the case in the experiments. We should note that we cannot define the onset temperature of the CO peak because the peak structure itself survives even at high $T$ [Fig. 3(a)].

Recently RIXS measurements showed that the energy range of charge dynamics coincides with that of magnetic excitations. This observation is natural from a view of $d$-wave bond-charge order. As shown in Refs. 13 and 42, $d$-wave bond-charge order comes from the exchange interaction, i.e., the $J$-term in the $t$-$J$ model. This is also clear from Eq. (3) where the interaction strength $J$ enters. If the charge dynamics originates from usual on-site charge excitations, its major contribution would appear above the particle-hole excitations.
as plasmons. On the other hand, needless to say, magnetic excitations are controlled by the $J$-term. Therefore both bond-charge and spin excitations naturally appear in the same energy scale of $J$. Obviously more theoretical studies are necessary to clarify the reason why the spin excitations are enhanced at the same wavevector as that of the bond-charge excitations, which is beyond the scope of the present work.

V. SUMMARY

Motivated by the recent RXS measurements of the doping dependence of the short-range CO observed in e-cuprates, we have studied the equal-time correlation function $S(q)$ associated with the $d$-wave bond-charge order by using a large-$N$ technique in the two-dimensional $t$-$J$ model. We have extended our previous work for a single doping rate by introducing the doping and temperature dependences of the damping $\Gamma$ and showed the consistency of our proposed scenario of the $d$-wave bond-charge order. The short-range CO is present even at high temperature and is pronounced at low temperature (Fig. 3). The peak intensity develops gradually down to $\delta \approx 0.10$ and is substantially suppressed below that (Fig. 4), due to the strong damping effect presumably coming from antiferromagnetic fluctuations. A recent experiment on a h-cuprate also shows that the CO competes with antiferromagnetic fluctuations. The momentum of the short-range CO decreases with lowering doping and goes up below $\delta \approx 0.10$ (Fig. 4). All these features reasonably capture the essential features of the recent experimental data. This agreement suggests three important implications for the physics in e-cuprates. i) The origin of the CO lies in the magnetic exchange interaction, i.e., $J$-term in the $t$-$J$ model. ii) The CO is not generated by antiferromagnetic fluctuations. Rather they seem to contribute to the enhancement of the quasiparticle damping and to suppress the charge ordering tendency. iii) The CO is not a usual charge-density-wave, but a bond-charge order. In particular, it is characterized by $d$-wave symmetry and is connected to the nematic order in the limit of $q = 0$. In this sense, the nematic physics plays a role also in e-cuprates, although so far it has been discussed only in h-cuprates.
Acknowledgments

The authors thank K. Ishii, B. Keimer, M. Minola, T. Tohyama, and R. Zeyher for very fruitful discussions. H.Y. acknowledges support by JSPS KAKENHI Grant No. 15K05189. A.G. acknowledges the Japan Society for the Promotion of Science for a Short-term Invitational Fellowship program (S17027) under which this work was initiated.

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temperature of the charge instability. This peculiar feature is addressed in detail in Ref. 27. Since a large momentum near \( q \approx (\pi, \pi) \) is not accessible by RXS and furthermore the peak structure around \( q_1 \) is not realized in the presence of a large \( \Gamma \), we focus on the peak structure around \( q_2 \), which is relevant to RXS as well as to recent experimental data\(^{24,25}\).

A factor of 1/2 comes from a large-\( N \) formalism where \( t \) is scaled by \( t/N \) and the physical situation corresponds to \( N = 2 \).

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