Optical conductivity in the CuO double chains of PrBa$_2$Cu$_4$O$_8$: Consequences of charge fluctuation

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(Dated: October 16, 2001)

We calculate the optical conductivity of the CuO double chains of PrBa$_2$Cu$_4$O$_8$ by the mean-field approximation for the coupled two-chain Hubbard model around quarter filling. We show that the $\sim 40$ meV peak structure, spectral shape, and small Drude weight observed in experiment are reproduced well by the present calculation provided that the stripe-type charge ordering presents. We argue that the observed anomalous optical response may be due to the presence of stripe-type fluctuations of charge carriers in the CuO double chains; the fast time scale of the optical measurement should enable one to detect slowly fluctuating order parameters as virtually a long-range order.

PACS numbers: 71.10.Fd, 73.90.+f, 71.27.+a, 78.20.Bh, 71.30.+h

I. INTRODUCTION

The CuO double chains of PrBa$_2$Cu$_4$O$_8$ (see Fig. 1(a)) show metallic conductivity along the chain direction down to 2 K [4, 5]. Because the system shows insulating conductivity along the directions perpendicular to the chains (except at low temperatures $\lesssim 140$ K), and also because the holes in the CuO$_2$ planes are localized in the O 2p-orbitals around Pr ions [3], PrBa$_2$Cu$_4$O$_8$ may be regarded as a model material of quasi-one-dimensional (1D) correlated conductors except at low temperatures. A possible Tomonaga-Luttinger-liquid description has then been tried to explain available experimental data [4, 5].

A number of anomalous behaviors have however been reported in the charge degrees of freedom of this system [4, 5]; a nuclear-quadrupole-resonance (NQR) experiment has demonstrated the presence of anomalous spin-lattice ($1/T_1$) and spin-spin ($1/T_2$) relaxations [4] and optical conductivity experiment clearly shows the presence of anomalous charge excitations at $\omega \gtrsim 40$ meV [4]. The CuO double-chain system therefore provides a good opportunity for studying the anomalous charge dynamics of strongly correlated 1D electron systems, such as charge ordering (CO) and charge fluctuations, just as the quasi-1D organic systems do [3, 6]. For example, a possible mechanism of metallization of this system due to charge frustration has recently been proposed [4].

Motivated by such development in the field, we in this paper focus on the optical conductivity spectra $\sigma(\omega)$ observed by Takenaka et al. [4] and consider the origin of the appearance of the characteristic peak structure at $\omega \sim 40$ meV as well as the presence of extremely small Drude weight. We use the mean-field approximation to the extended Hubbard model defined on a lattice of coupled two chains to obtain the ground-state phase diagram, where we find that there appear two types of CO phases, depending on the parameter values. We then calculate the optical conductivity spectrum $\sigma(\omega)$ for each of these two phases in the mean-field approximation. We find that the spectra calculated for the ground state of the stripe-type charge ordering reproduces the experimental peak structure including not only its peak position and spectral shape but also the observed very small Drude weight. We also find that the spectra calculated for the in-line--type CO phase completely fails to reproduce the experimental spectra.

It is known that the CuO double chains do not show the long-range CO but have the fluctuation of electronic charges with very slow time scale of the order of 0.1 – 10 MHz [6]. We then point out that the fast time scale of the optical measurement should enable one to detect slowly fluctuating order parameters as virtually a long-range order; i.e., the spectra observed should basically be the same as those for the long-range CO state. Therefore, we argue that the fact that our mean-field optical spectra agree well with experiment reasonably suggest that the stripe-type charge fluctuations should present in the system and should be the origin of the $\sim 40$ meV peak structure.
peak structure in the optical conductivity spectrum of the CuO double chains of PrBa$_2$Cu$_4$O$_8$.

This paper is organized as follows. In Sec. II, we define a model for describing the electronic states of the CuO double chains. In Sec. III, we derive the mean-field phase diagram of the Hamiltonian, and in Sec. IV, we calculate the optical conductivity of the model. The results are discussed in Sec. V by comparing with experimental data and their implication is considered. Summary is given in Sec. VI.

II. MODEL

The structure of the CuO double-chain system is illustrated in Fig. 1(a), where we show the two CuO chains connected with zigzag bonds. The electronic structure may be modeled as a single-band extended Hubbard-type Hamiltonian representing the doping holes in the antibonding band of the Cu 3$d_{x^2−y^2}$ and O 2$pσ$ orbitals. The filling of holes in the chains is reported to be $n ≃ 0.5$ (quarter filling) or; the angle-resolved photoemission spectroscopy experiment suggests the value $n ≃ 0.46 ± 0.06$ and optical conductivity measurement suggests the value $n ≃ 0.4$.

We therefore adopt in this paper the single-band Hubbard model around quarter filling defined on the lattice of two chains connected with zigzag bonds as shown in Fig. 1(b). The Hamiltonian may be of the following form:

$$H = −\sum_{<ij>\sigma} t_{ij} c_{iσ}^† c_{jσ} + \text{H.c.} + U \sum_{i} n_{i↑} n_{i↓} + \sum_{<ij>} V_{ij} n_{i} n_{j}$$

(1)

where $c_{iσ}^†$ ($c_{iσ}$) is the creation (annihilation) operator of a hole at site $i$ and spin $σ = \uparrow, \downarrow$, and $n_{i} = n_{i↑} + n_{i↓}$ is the number operator. We take into account the hopping parameters for the chain bond $t$ ($=t_2$) and for the zigzag bond $t'$ ($=t_1$) where we note $t' \ll t$ because of the bonding angle minimizing the overlap of the Cu 3$d_{x^2−y^2}$ and O 2$pσ$ wave functions for the hopping parameter $t'$; in the following, we use the value $t'/t = 0.1$ confirming that the sign of $t'$ does not change the results. We also take into account the intersite Coulomb interactions $V$ for the chain bonds and $V'$ for the zigzag bonds, as well as the on-site Coulomb interaction $U$; the values are varied to simulate various situation. We restrict ourselves to the case at and less than quarter-filling of holes $n = N/L ≤ 0.5$ where $N$ and $L$ are the total numbers of holes and lattice sites, respectively.

III. MEAN-FIELD PHASE DIAGRAM

We adopt the mean-field approximation

$$n_{i↑} n_{i↓} \simeq \langle n_{i↑} \rangle \langle n_{i↓} \rangle + \langle n_{i↑} \rangle \langle n_{i↓} \rangle - \langle n_{i↑} \rangle \langle n_{i↓} \rangle$$

$$n_{i} n_{j} \simeq \langle n_{i} \rangle \langle n_{j} \rangle + \langle n_{i} \rangle \langle n_{j} \rangle - \langle n_{i} \rangle \langle n_{j} \rangle$$

(2)

(3)

for the interaction terms of the Hamiltonian Eq. (1). We assume periodicity of the ordered states where each unit cell $r$ contains $M$ sites; the lattice site may thus be specified as $i = (r, µ)$ with the unit cell $r (=0, \cdots, L/M − 1)$ and site position $µ (=1, \cdots, M)$ therein. We have $2M$ order parameters

$$δ_µ = \langle n_{µ↑} \rangle + \langle n_{µ↓} \rangle − n$$

$$2S_µ^z = \langle n_{µ↑} \rangle − \langle n_{µ↓} \rangle$$

(4)

(5)

with the charge neutrality constraint $\sum_µ δ_µ = 0$. We define $c_{µκσ}$ as the Fourier transform of $c_{iσ}$:

$$c_{µκσ} = \frac{1}{\sqrt{L/M}} \sum_{r=1}^{L/M} e^{ikR r} c_{rµσ}$$

(6)

where $R r = r M a + µ a$ and $k$ is the wavevector taking $L/M$ values in the Brillouin zone $−π/Ma ≤ k < π/Ma$.

$a$ is the lattice constant of the topologically equivalent single-chain model with nearest-neighbor ($t', V'$) and next-nearest-neighbor ($t, V$) interactions. The mean-field Hamiltonian is then written as

$$H^{MF} = \sum_{kσ} \{c_{1kσ}^† c_{1kσ}, \cdots, c_{Mkσ}^† \} [H^{MF}_{kσ}] \begin{pmatrix} c_{1kσ} \\ c_{2kσ} \\ \vdots \\ c_{Mkσ} \end{pmatrix} + E_c$$

(7)

with

$$[H^{MF}_{kσ}] = [H_t] + [H'_t]$$

(8)

where

$$[H_t]_{µν} = \begin{cases} a_{±}^2 & \text{if } ν = µ ± 1 \\ a_{±}^2 & \text{if } ν = µ ± 2 \\ a_{±}^2 & \text{if } µ = 1, ν = M − 1 \\ a_{±}^2 & \text{if } µ = 1, ν = M \\ a_{±}^2 & \text{if } µ = 2, ν = M \\ a_{±}^2 & \text{if } µ = M − 1, ν = 1 \\ a_{±}^2 & \text{if } µ = M, ν = 1 \\ a_{±}^2 & \text{if } µ = M, ν = 2 \\ 0 & \text{otherwise} \end{cases}$$

(9)

and

$$[H'_t]_{µν} = U \langle n_{µ−σ} \rangle + V' \langle n_{µ−1,σ} + n_{µ+1,σ} \rangle + V \langle n_{µ−2} \rangle + \langle n_{µ+2} \rangle \rangle δ_{µν}.$$  

(10)

In Eq. (9), we define $a_{±}^± = −t_i e^{±iκ a}$ ($l = 1, 2$). Also, we have

$$E_c = −N \sum_{µ=1}^{M} \left[ U \langle n_{µ↑} \rangle \langle n_{µ↓} \rangle \right.$$  

$$\left. + \sum_{σ,σ'} \left( V'(n_{µσ}) \langle n_{µ+1,σ'} \rangle + V \langle n_{µσ'} \rangle \langle n_{µ+2,σ} \rangle \right) \right]$$

(11)
Diagonalizing the matrix (8) by the unitary transformation

$$[P_{k\sigma}] = (P_{k\sigma}^{(1)}, P_{k\sigma}^{(2)}, \ldots, P_{k\sigma}^{(M)})$$

(12)

with $P_{k\sigma}^{(n)} = (P_{1k\sigma}^{(n)}, P_{2k\sigma}^{(n)}, \ldots, P_{Mk\sigma}^{(n)})$, we have

$$H_{MF} = \sum_{nk\sigma} E_{k\sigma}^{(n)} \gamma_{nk\sigma} \gamma_{nk\sigma} + E_c$$

(13)

where $E_{k\sigma}^{(n)}$ is the energy of the $n$-th quasiparticle. The annihilation operator of the quasiparticle may be written as

$$\gamma_{nk\sigma} = \sum_{\mu=1}^{M} p_{\mu k\sigma}^{(n)*} c_{\mu k\sigma}$$

(14)

$$c_{\mu k\sigma} = \sum_{n=1}^{M} p_{\mu k\sigma}^{(n)} \gamma_{nk\sigma}.$$  

(15)

The mean-field ground state is then written as

$$|0\rangle = \prod_{nk\sigma}^{occ.} \gamma_{nk\sigma}^\dagger |\text{vacuum}\rangle.$$  

(16)

The order parameters are determined self-consistently so as to minimize the total energy of the system. The relation

$$\langle n_{\mu\sigma} \rangle = \frac{M}{T} \sum_{nk} \left| p_{\mu k\sigma}^{(n)} \right|^2 \theta(\varepsilon - E_{k\sigma}^{(n)})$$

(17)

is used to make iterations for self-consistency where $\theta(x)$ is the Fermi function at $T = 0$ K and $\varepsilon$ is the Fermi level.

The calculated ground-state phase diagram of the model is shown in Fig. 2, where we use $M = 8$. We find that there appear the following three types of phases: (i) The charge-ordered phase denoted as CO-1 where the in-line–type ordering of charges emerges (see inset of Fig. 2). The spins are aligned antiferromagnetically at sites on one of the two chains; there is no spin polarization on the other chain (and even charges vanish if we set $t' = 0$). (ii) The charge-ordered phase denoted as CO-2 where the stripe-type ordering of charges emerges (see inset of Fig. 2). The up and down spins on the neighboring two sites would form a spin-singlet pair if quantum fluctuation is introduced. (iii) The spin-density-wave (SDW) phase with a small modulation of the charge-density distribution, which we denote as SDW+CDW, as shown in Fig. 2.

We first note that the $U/t$ dependence of the phase boundary is rather small, that the CO phases are maintained up to $\sim 10\%$ doping levels, and that the doping dependence of the location of the phase boundary is also very small. Because we assume the unit cell of 8 sites with the commensurate CO, the system becomes metallic upon doping where the small Fermi surface emerges.

We also note that the phase boundary between CO-1 and CO-2 is discontinuous; i.e., the transition is of the first order. The transition between the phases CO-2 and SDW+CDW is however continuous; when $t'$ is smaller than a small critical value, a pure SDW phase is realized in this ‘SDW+CDW’ region and the transition to the CO-2 phase is of the second order. When $t'$ exceeds the critical value, the phase boundary becomes obscure and the transition changes into a crossover; when the value of $U/t$ decreases, the crossover becomes broader.

The spin degrees of freedom of the fluctuating CO phases seems intriguing; the CO-1 phase, if it is a long-range CO, should behave like a 1D Heisenberg antiferromagnet, and in the CO-2 phase, the spin-singlet formation should occur. Experimentally, it has been found that the temperature-independent Pauli paramagnetic contribution and Curie-Weiss contribution coexist with a small anomaly due to the ordering of Pr spins at low temperatures. It would be interesting if any influence of the fluctuating charges on the spin states can be detected in experiment.
IV. OPTICAL CONDUCTIVITY

We now calculate the optical conductivity spectra for the states derived in the previous section. Using the eigenstates \( |f \rangle \) and energies \( E_f \) (where \( f = 0 \) denotes the ground state) of the many-body Hamiltonian Eq. (1), we generally define the optical conductivity \( \sigma_{\alpha\alpha}(\omega) \) \((\alpha = x, y, z)\) as

\[
\sigma_{\alpha\alpha}(\omega) = D_{\alpha\alpha} \delta(\omega) + \frac{\pi e^2}{L} \sum_{f \neq 0} \frac{1}{\omega} |\langle f | j_\alpha | 0 \rangle|^2 \delta(\omega - (E_f - E_0)) \tag{18}
\]

with the current operator

\[
j_\alpha = i \sum_{ij\sigma} t_{ij} (r_i - r_j) c_i^\dagger \gamma_{\alpha\sigma} c_j \tag{19}
\]

where \( D_{\alpha\alpha} \) is the Drude weight. In 2D, the sum rule holds with the kinetic energy operator

\[
T = -\sum_{ij\sigma} t_{ij} |r_i - r_j|^2 \gamma_{\alpha\sigma} c_i^\dagger c_j. \tag{21}
\]

The relation (20) may be used to calculate the Drude weight \( D \) as

\[
\frac{D}{\pi e^2} = -\frac{1}{L} \langle 0 | T | 0 \rangle + \frac{2}{L} \sum_{f \neq 0} \frac{1}{E_f - E_0} \sum_{f \neq 0} \frac{1}{E_f - E_0}. \tag{22}
\]

Note that because the present system is finite along the y-direction the Drude contribution vanishes for the electric field perpendicular to the chains; \( D \) is thus associated only with \( E \parallel x \).

We use the mean-field results to calculate the optical conductivity and Drude weight. The interband optical transition from the mean-field ground state may be written

\[
|s \leftrightarrow t\rangle_{k\sigma} = \gamma_{s\kappa\sigma}^\dagger \gamma_{t\kappa\sigma} |0\rangle \tag{23}
\]

where the momentum is conserved. We may then rewrite the terms in Eq. (22) as

\[
\sum_{f \neq 0} \frac{|\langle f | j_{\alpha} | 0 \rangle|^2}{E_f - E_0} = \sum_{s \neq t} \sum_{k\sigma} \frac{|\gamma_{s\kappa\sigma}^\dagger \gamma_{t\kappa\sigma} |0\rangle|^2}{E_{s\kappa\sigma} - E_{t\kappa\sigma}} \tag{24}
\]

with

\[
k\sigma \langle s \leftrightarrow t | j_{\alpha} | 0 \rangle = ia \cos \phi \left( J_{st,k\sigma}^{(1x)} + J_{st,k\sigma}^{(2x)} \right) \theta_{k\sigma}(1 - \theta_{k\sigma})
\]

\[
k\sigma \langle s \leftrightarrow t | j_{\alpha} | 0 \rangle = ia \sin \phi \left( J_{st,k\sigma}^{(1y)} \right) \theta_{k\sigma}(1 - \theta_{k\sigma}) \tag{25}
\]

where

\[
J_{st,k\sigma}^{(1x)} = \sum_{\mu=1}^{M} \left( F_{1} p_{\mu+1,k\sigma}^{(s)*} p_{\mu,k\sigma} - F_{1} p_{\mu,k\sigma}^{(s)*} p_{\mu+1,k\sigma} \right)
\]

\[
J_{st,k\sigma}^{(1y)} = \sum_{\mu=1}^{M} (-1)^{\mu} \left( F_{1} p_{\mu+1,k\sigma}^{(s)*} p_{\mu,k\sigma} - F_{1} p_{\mu,k\sigma}^{(s)*} p_{\mu+1,k\sigma} \right)
\]

and

\[
\theta_{k\sigma}(t) = \theta(\varepsilon_{F} - E_{k\sigma})
\]

\[
F_{1} = \frac{1}{4} t t e^{i k \xi} \quad (l = 1, 2). \tag{27}
\]

Here, \( \phi \) is the angle between the chain and zigzag bonds defined in Fig. 1. Also, the kinetic energy term may be written

\[
\langle 0 | T | 0 \rangle = -a^2 \sum_{nk\sigma} \left( T_{nk\sigma}^{(1)} + 2 \cos^2 \phi T_{nk\sigma}^{(2)} \right) \theta_{k\sigma}^{(n)} \tag{28}
\]

with

\[
T_{nk\sigma}^{(1)} = \sum_{\mu=1}^{M} \left( F_{1} p_{\mu+1,k\sigma}^{(n)*} p_{\mu,k\sigma} + F_{1} p_{\mu,k\sigma}^{(n)*} p_{\mu+1,k\sigma} \right). \tag{29}
\]

Note that in the paramagnetic state the spectral weight at \( \omega > 0 \) completely vanishes, leaving only the Drude weight, because the present system has only a single band and therefore has no interband transitions.

Calculated results for the parameter and doping dependences of the optical conductivity spectra at \( \omega > 0 \) for \( E \parallel x \) are shown in Fig. 3. We note that the spectral weight for \( E \parallel y \) is two orders of magnitude smaller than that for \( E \parallel x \) (and therefore not shown here) because the spectra for \( E \parallel x \) have the weight roughly proportional to \( t^2 \) while the spectra for \( E \parallel y \) have the weight proportional to \( t^2 \), and hence we assume the value \( t'/t = 0.1 \). This is consistent with the fact that the experimental spectral weight for \( E \parallel a \) is more than two orders of magnitude smaller than the weight for \( E \parallel b \).

Conversely, this agreement with experiment demonstrates that the value of \( t' \) is indeed much smaller than the value of \( t \) as we have assumed in this paper.

We find in Fig. 3 that the spectra for the CO-2 ground state locate in the energy around \( \omega/t = 0.2 \) whereas the spectra for the CO-1 ground state locate in a much higher energy range \( \omega/t = 3 - 6 \). Because the gaps of the quasiparticle band structure of the two CO states have a similar value, this difference in the location of the spectral weight is purely the matrix-element effects, i.e., the selection rule for these two types of CO states is quite different. We may have the following real-space interpretation for the obtained spectra: In the CO-1 ground
The results are at quarter filling (\(n = 0.5\)) for the ground states of CO-2 (in (a) and (b)) and CO-1 (in (c) and (d)). Drude peak (\(\omega = 0\)) is not included. The results are at quarter filling (\(n = 0.5\)) in (a) and (c), and at a 2% doping level (\(n = 0.49\)) in (b) and (d). Parameter values used for calculation are: \(V'/t = 0.707\), \(V/t = 0.5\), and \(t'/t = 0.1\) in (a) and (b), and \(V/t = 1\) and \(t'/t = 0.1\) in (c) and (d).

state, the electric field along the chain takes out a hole and put it on the neighboring site to make either a doubly occupied site via \(t\) or a singly occupied site via \(t'\), the former of which costs the energy \(U\) and the latter of which costs the energy \(V'.\) Since we assume \(t \gg t'\), the main spectral weight appears at the position \(\omega \sim U\). We find in Figs. 3 (c) and (d) that the position of the main peak indeed depends strongly on \(U\) but not on \(V'.\) In the CO-2 ground state, the excitation by the electric field along the chain does not create doubly occupied sites in the strong coupling limit, the energy cost of which is at most \(V\). We thus have the main peak much lower in energy. We find some fine structures of the spectra which reflects the fairly complicated quasiparticle multi-band structure.

Integrated spectral weight of the Drude (\(\omega = 0\)) and interband (\(\omega > 0\)) contributions are shown separately in Fig. 4. We find that the Drude weight grows very rapidly by doping away from quarter filling at which the Drude weight vanishes. We also find that the \(U/t\) dependence of the integrated spectral weight is not very strong.

Let us first summarize the experimental aspects of the optical conductivity of \(\text{PrBa}_2\text{Cu}_4\text{O}_8\) observed at room temperature [2]. The spectra have the following three features: (i) a broad peak structure with large spectral weight (98% of the total weight) located around \(\omega \approx 40\, \text{meV}\), (ii) very small Drude weight (2% of the total weight), and (iii) the spectra for \(E \parallel a\) have the weight which is two orders of magnitude smaller the weight for \(E \parallel b\). Another important aspect of the charge dynamics of this material is given by the NQR experiment [1]; (iv) the CuO double chains have a very slow fluctuation of the electric-field gradient caused by the spatial fluctuation of electronic charge carriers. The system may thus be in the vicinity of the long-range CO state.

Then, in Fig. 5, we compare our calculated optical conductivity with the experimental spectrum [2] for \(\text{PrBa}_2\text{Cu}_4\text{O}_8\). We find that the experimental spectrum is well reproduced: (i) The calculated spectrum, which is in reasonable agreement with experiment, is for the CO-2 ground state near the crossover region to the SDW+CDW phase (see Fig. 2). The spectra calculated for the CO-1 states completely fail to agree with experiment as already pointed out in Sec. IV. (ii) The spectra for the electric field perpendicular to the chains have the weight two orders of magnitude smaller than those for the electric field parallel to the chains, which is consistent with experiment (see Sec. IV). (iii) The calculated peak structure is found to locate at rather higher energies than experiment. Although this is partly due to insufficiency of the mean-field approximation, fine-tuning of the parameter values would improve the agreement with experiment. (iv) The observed very small Drude weight

FIG. 3: Optical conductivity spectra \(\sigma(\omega)\) with \(E \parallel x\) calculated for the ground states of CO-2 (in (a) and (b)) and CO-1 (in (c) and (d)). Drude peak \((\omega = 0)\) is not included. The results are at quarter filling \((n = 0.5)\) in (a) and (c), and at a 2% doping level \((n = 0.49)\) in (b) and (d). Parameter values used for calculation are: \(V'/t = 0.707\), \(V/t = 0.5\), and \(t'/t = 0.1\) in (a) and (b), and \(V/t = 1\) and \(t'/t = 0.1\) in (c) and (d).

FIG. 4: Filling dependence (left panel) and \(U/t\) dependence (right panel) of the spectral weight at \(\omega = 0\) (Drude weight, filled circles) and integrated weight over \(\omega > 0\) excluding the Drude contribution (open circles). The CO-2 phase is assumed. Parameter values used for calculation are: \(V/t = 0.5\), \(V'/t = 0.707\), and \(t'/t = 0.1\).

V. DISCUSSION
is well reproduced by our calculation. (v) The asymmetric spectral shape of the main peak structure, i.e., sharp in the lower energy side and broad in the higher energy side, is well reproduced by our calculation. The width of the peak is however rather narrow compared with experiment. This may be due to the effect of electron correlations neglected in the mean-field approximation, i.e., the scattering of the quasiparticles may well broaden the spectra. (vi) As for the amplitude of the order parameters to be \( \delta = 0.013 \text{ (per site)} \) and \( 2S^2 = 0.21 \text{ (per site)} \) for the parameter values used in the calculation of the optical conductivity of Fig. 5. This value of \( \delta \) is consistent with the experimental estimation where it has been pointed out that less than 0.02 holes per \( \text{Cu} \) are frozen.

Finally, let us discuss the optical conductivity spectra for fluctuating charge carriers. We should first note that there is no experimental indication of the long-range CO in \( \text{PrBa}_2\text{Cu}_4\text{O}_8 \) but rather there is only the slow fluctuation of electronic charge degrees of freedom. Then, in this situation, we find that the spectra calculated for the \textit{ordered} state agree reasonably well with experimental spectra. The time-scale argument may readily resolve this apparent contradiction; the fast time scale of the optical measurement should enable one to detect slowly fluctuating order parameters as virtually a long-range order. This situation reminds us of the optical conductivity spectra of the well-known CO material \( \alpha^\prime\text{-NaV}_2\text{O}_5 \). The spectra for the CO phase of this system differ very little from the spectra of the uniform phase at temperatures above \( T_{CO} \); there exists slow fluctuation of the charge degrees of freedom even far above the transition temperature. Similar situation has also been noticed for the quasi-1D organic materials.

In the present \( \text{CuO} \) double-chain systems, if the system were exactly at quarter filling, the long-range CO might occur with vanishing metallic conductivity. This means that if a fine filling-control is made on the \( \text{CuO} \) double chains one may be able to obtain the insulating double chains. We therefore would suggest that the experimental filling control, e.g., by applying pressure or elemental substitution (if it purely changes the filling of the \( \text{CuO} \) double chains), seems to be quite interesting.

VI. SUMMARY

We have considered the anomalous charge dynamics of the \( \text{CuO} \) double chains of \( \text{PrBa}_2\text{Cu}_4\text{O}_8 \) by focusing on the optical conductivity spectra recently observed in experiment.

Our study presented in this paper may be summarized as follows: (i) We have experimental evidence of the presence of strong charge fluctuations, i.e., the system may be in the vicinity of the charge-ordered state. Then, one may naturally assume the presence of strong intersite Coulomb repulsions in the system. (ii) We have therefore adopted an extended Hubbard-type model with the intersite Coulomb repulsion near quarter filling and treated this in the mean-field approximation. We thus have found that the possible charge-ordered state may be either of the in-line-type or of the stripe-type charge modulations. (iii) We have calculated the optical conductivity spectra for the model in the same approximation and found that the spectra calculated for the stripe-type charge ordering reproduce the experimental features of the \( \sim 40 \text{meV} \) peak and small Drude weight reasonably well but that the spectra for the in-line-type charge ordering does not agree with experiment. (iv) Then, we have argued that the stripe-type fluctuations of charge carriers should present in the system and this is the origin of the anomalous charge response of the system. The \( \text{CuO} \) double chains have no \textit{long-range} charge ordering but the fast time-scale of the optical measurement should detect the very slow fluctuation of charges as virtually a long-range charge ordered state.

The \( \text{CuO} \) double chains in \( \text{PrBa}_2\text{Cu}_4\text{O}_8 \) thus provide a good opportunity for studying the anomalous charge dynamics in the 1D strongly correlated electron systems. We hope that further experimental and theoretical studies will be made to clarify various aspects of this intriguing system.

Acknowledgments

We thank K. Takenaka for sending us their unpublished experimental data and enlightening discussion. We also acknowledge useful discussions with S. Horii, T.
Ohama, H. Seo, and H. Yoshioka. This work was supported in part by Grants-in-Aid for Scientific Research (Nos. 11640335 and 12046216) from the Ministry of Education, Culture, Sports, Science, and Technology of Japan. Computations were carried out at the computer centers of the Institute for Molecular Science, Okazaki, and the Institute for Solid State Physics, University of Tokyo.

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