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Frequency-dependent conductivity from carriers in Mott insulators

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The unusual broad absorption band in the infrared observed in the high-T_c superconductors $YBa_2Cu_3O_{7-x}$ is explained as a consequence of the diffusive character of the higher energy states of a carrier moving in a Mott insulator. A good fit is obtained to the experimental spectrum of Thomas *et al.* with reasonable values of the input parameters.

I. INTRODUCTION

The discovery of high-temperature superconductivity in the Cu oxides[1] and Anderson's proposal[2] that the essential elements, the Cu-O_2 planes, are described by a strongly correlated band near to half filling has given new impulses to the theoretical study of the properties of carriers in Mott insulator. The continual improvement of experimental results in terms of accuracy and reliability has reached the point where direct confrontations of experiment and the theoretical models can be undertaken. As an example, a series of infrared and optical experiments on single crystals of $YBa_2Cu_3O_{7-\delta}$ by Schlesinger, Collins, Kaiser, and Holtzberg[3] and by Thomas and coworkers[4] has found an unusual frequency dependence of the conductivity in the energy range $0<\omega\leq 2$ eV. In the present work we show that this behavior finds a natural explanation within the strongly correlated model. In this model the charge carriers are holes (formal charge Cu^{3+}, spin 0) moving in a background of localized spins on the sites with formal charge Cu^{2+}. The localized spins are coupled with a Heisenberg exchange coupling. A number of years ago Brinkman and Rice[5] showed that the majority of the hole states in a Mott insulator have a diffusive character which arises from the strong coupling between the motion of the holes and the spin configurations. In this work we show that this diffusive character causes an essential broadening of the optical conductivity due to these holes. The result is a broad continuum absorption stretching up to quite high energies whose form can quantitatively fit the experiments.

II. FREQUENCY-DEPENDENT CONDUCTIVITY

Our starting point is the Kubo formula for the real part of conductivity

$$\sigma(\omega)=\frac{1-e^{-\beta\omega}}{\Omega}\left(\frac{\pi}{Z}\right)\sum_{n,m}e^{-\beta E_n}\delta(E_m-E_n-\omega)\times|\langle n|J_x|m\rangle|^2, \quad (1)$$

where the summation is over eigenstates $|n\rangle$ with energies E_n and Z is the partition function, J_x the current operator in x direction, $\beta=1/k_BT$, Ω the volume (in d dimensions). If the current operator J_x commutes with the Hamiltonian, the current matrix element $\langle n|J_x|m\rangle\propto\delta_{n,m}$ and $\sigma(\omega)$ is simply a δ function at $\omega=0$. For a normal metal, umklapp scattering between electrons, and between electron and other particles causes a finite relaxation time and the conductivity is usually of the Drude form. However, the situation can be very different for systems with strongly diffusive motion. In the latter case, $|\langle n|J_x|m\rangle|^2$ is roughly constant, relatively independent of $|n\rangle$ and $|m\rangle$. Applying Eq. (1) to such systems, we have at low temperatures ($\beta\omega\gg 1$),

$$\sigma(\omega)\approx\frac{1}{\omega}\sum_m\delta(E_m-E_0-\omega)\times\text{const}, \quad (2)$$

where E_0 is the ground state energy of the system. If the density of states is relatively flat, then $\sigma(\omega)\propto 1/\omega$, very different from the frequency dependence of carriers with propagating motion.

In the following we show explicitly that such a frequency-dependent conductivity is realized in doped Mott insulators at frequencies $\omega\gg J$, where J is the spin-spin interaction coupling. We argue further that $\sigma(\omega)$ should show additional structure at low frequencies, $\omega\lesssim J$.

Let us consider the effective Hamiltonian of the strongly correlated Hubbard model in a d-dimensional cubic lattice. Note this form can also be derived more generally.[6]

$$H=H_t+H_s,$$
$$H_t=t\sum_{\langle ij\rangle s}Pc_{is}^{\dagger}c_{js}P+\text{H.c.},$$
$$H_s=J\sum_{\langle ij\rangle}\mathbf{S}_i\cdot\mathbf{S}_j. \quad (3)$$

In Eq. (3), $\langle ij\rangle$ represents a nearest-neighbor pair, P is the operator which projects out all doubly occupied sites, and H_t and H_s describe the kinetic and spin-spin interaction parts, respectively. We shall be interested only in the case $J/t\ll 1$, and a low density of holes, which can be treated independently.

At frequencies $\omega\gg J$, the hole does not see the spin fluctuations on the time scale J^{-1} so that the high-frequency part of conductivity is essentially independent of H_s. Therefore, we can replace H by H_t in calculating $\sigma(\omega)$.

Almost two decades ago, Brinkman and Rice[5] investigated the motion of a single hole in various spin con-

figurations in the limit $J/t \rightarrow 0$. In that work, the problem was formulated in terms of the number of paths which return to the origin leaving the spin configuration unchanged. It was found that the dominant contribution is from the class of paths in which the hole completely retraces all steps and that such paths contribute independent of the spin configuration. This class has also been referred to as paths with no closed loops or paths on a Bethe lattice. The density of states of a single hole was calculated with this retraceable path approximation (rpa). The density of states $D(\omega)$ is given by

$$ztD(\omega)=\frac{1}{\pi}(z-1-\omega^2/4t^2)^{1/2}/[1-(\omega/zt)^2]\,,$$

where z is the coordination number of the lattice. In Fig. 1 we plot the density of states for a square lattice. Note, $D(\omega)$ is relatively flat except near the edges. Assuming the current matrix elements to be constant, we would expect $\sigma(\omega) \propto 1/\omega$ from Eq. (2).

Very recently Joynt,[7] and Lederer and Takahashi[8] have calculated density of states for a single hole in a quantum antiferromagnetic (AF) configuration and in a resonant valence-bond state, including also some closed loops. Their results show that the hole density of states in these different configurations is similar and so the deviations from a $1/\omega$ law should not be large.

Given the above qualitative discussions, we now are in a position to calculate $\sigma(\omega)$ quantitatively. The Kubo formula Eq. (1) can be rewritten as

$$\sigma_0(\omega)=-\frac{1-e^{-\beta\omega}}{\omega 4\pi Z\Omega}\int_{-\infty}^{+\infty}dv\,e^{-\beta v}F(v,v+\omega)\,, \tag{4}$$

where we use notation $\sigma_0(\omega)$ for single-hole conductivity, and

$$F(\omega_1,\omega_2)=\sum_{s_1,s_2=\pm 1}s_1s_2\mathcal{J}(\omega_1+is_1\delta,\omega_2+is_2\delta)\,, \tag{5}$$

$$\mathcal{J}(\omega_1,\omega_2)=\mathrm{Tr}[(\omega_1-H_t)^{-1}J_x(\omega_2-H_t)^{-1}J_x]\,. \tag{6}$$

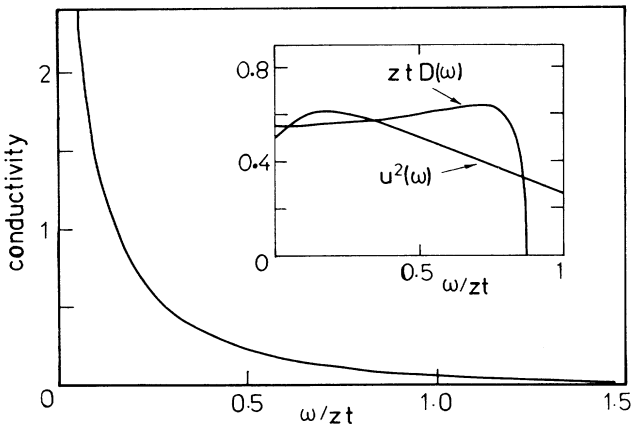

FIG. 1. Conductivity of a charge carrier in a Mott insulator, in units of d^2a^2/Ω, for $\omega \gg J,T$. The inset shows the density of states $D(\omega)$ and the function $u^2(\omega)$ of Eq. (10). All quantities are calculated for a square lattice, and within the retraceable path approximation.

In Eq. (6), we have replaced H by H_t for the reasons given above. The current operator J_x for systems in (3) is given by

$$J_x=eati\sum_{js}(c^\dagger_{j+x,s}c_{js}-c^\dagger_{j-x,s}c_{js})\,, \tag{7}$$

with a the lattice constant, and $(j+x)$ the lattice site next to j in the positive x direction. The dc conductivity formula given in Ref. 5 is a limiting case of Eqs. (4)–(7) as $\omega \rightarrow 0$.

In the temperature region $\beta\omega \gg 1$, Eq. (5) becomes

$$\sigma_0(\omega)=-\frac{1}{4\pi\omega\Omega}\lim_{v\rightarrow v_0}F(v,v+\omega)/D(v)\,, \tag{8}$$

where $-v_0$ is the lower band-edge position and $v_0=2\sqrt{z-1}\,t$ within rpa.

The function $\mathcal{J}(\omega_1,\omega_2)$ was evaluated in Ref. 5 within the rpa, and using the result given there, we obtain for $\omega \gg J$ and T,

$$\sigma_0(\omega)=\frac{1}{\omega\Omega}2\pi(eat)^2D(-v_0+\omega)u^2(\omega)\,, \tag{9}$$

where

$$u^2(\omega)=\frac{(z-2)^3[(z-2)^2+4(z-1)\tilde{\omega}(2-\tilde{\omega})]}{z[(z-2)^2+2\tilde{\omega}(z-1)]^2}\,, \tag{10}$$

with $\tilde{\omega}=\omega/v_0$.

If $u^2(\omega)$ were independent of ω, Eq. (9) would reduce to Eq. (2), so that the ω dependence of $u^2(\omega)$ given in Eq. (10) may be regarded as a correction to Eq. (2). The form of $u^2(\omega)$ in a square lattice ($z=4$) is plotted in Fig. 1, and it is apparent that $u^2(\omega)$ depends only weakly on ω.

Note, in a one-dimensional (1D) system, in which $z=2$, we obtain $u^2(\omega)=0$. Therefore $\sigma_0(\omega)$ vanishes everywhere except at $\omega=0$. Note in one dimension the current operator J_x commutes with H_t but not in higher dimensions.[5] The f-sum rule then requires the dc conductivity to be infinite, in accordance with the infinite mobility in one dimension obtained in Ref. 5.

We are primarily interested in a square lattice ($z=4$), and $\sigma_0(\omega)$ as a function of ω is plotted in Fig. 1. In a wide region of ω, $\sigma_0(\omega)$ is close to $1/\omega$. As a test for the rpa, we have compared our results against the exact f-sum rule. For the model Hamiltonian (3), all contributions to conductivity are intraband transitions and the f-sum rule in this case was derived in Ref. 9,

$$\int_0^\infty d\omega\,\sigma_{\mathrm{exact}}(\omega)=-\frac{\pi e^2a^2}{2\Omega}T_x\,,$$

where T_x is the kinetic energy in the x direction. Integrating $\sigma_0(\omega)$ of Eq. (9), we find

$$\int_0^\infty d\omega\,\sigma_0(\omega)\Big/\int_0^\infty d\omega\,\sigma_{\mathrm{exact}}(\omega)\simeq 0.87\,,$$

which shows that the rpa is quite good.

III. COMPARISON WITH EXPERIMENT

We now wish to compare our results with available experiments. Within this independent hole approximation,

the conductivity contributed from all $Cu\text{-}O_2$ planes in bulk superconducting Cu oxides is

$$\sigma(\omega) = n_h \Omega \sigma_0(\omega) , \tag{11}$$

where n_h is the number of in-plane holes per cm^3, Ω here is the area of a $Cu\text{-}O_2$ plane. For $YBa_2Cu_3O_{7-\delta}$ compounds there are also holes on chains which contribute to $\sigma(\omega)$. It is far from clear how to separate the chain and plane contributions. Since in the insulating regime (e.g., $\delta = 0.8$ in Fig. 2) only the chains contribute to $\sigma(\omega)$ for $\omega \lesssim 2$ eV and since $\sigma(\omega)$ is approximately constant here we make the, admittedly ad hoc, assumption that a constant form continues to hold also for all values of δ. The form of $\sigma(\omega)$ is compared in Fig. 2 to experimental values obtained from the reflectivity measurements by Thomas and co-workers.[4] The experimental $\sigma(\omega)$ are replotted here from Ref. 4. In the theoretical calculations, we use the hopping integral $|t| = 0.5$ eV, Cu-Cu lattice constant $a = 3.8$ Å, and the density of in-plane holes are $n_h = 2.5 \times 10^{21}$ cm^{-3} [in Fig. 2(a)] and $n_h = 3.4 \times 10^{21}$ cm^{-3} [in Fig. 2(b)]. These values correspond to formal valence estimates of +2.21 and +2.29, respectively, for the Cu ions on the planes. The calculated $\sigma(\omega)$ are almost unchanged if we change the value of t (for example, $t = 0.3$ eV) while keeping $n_h t$ fixed. At high frequencies $\omega > 2000$ cm^{-1}, the theoretical results of $\sigma(\omega)$ agree well with the experiments.

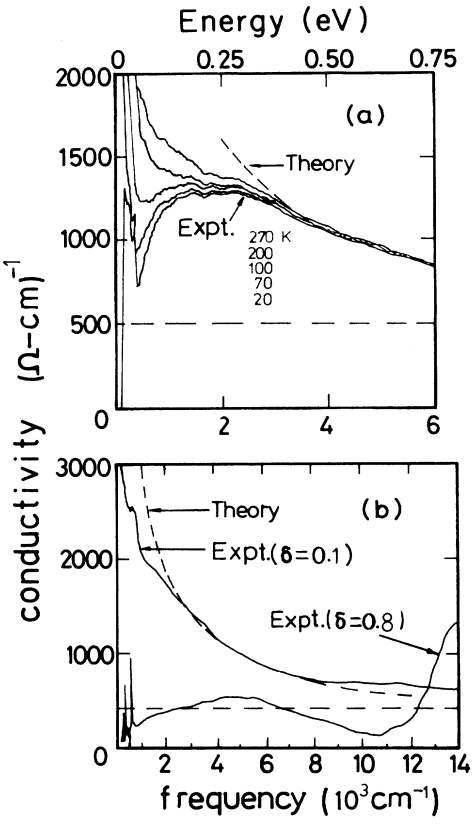

FIG. 2. Comparison between theory and experiments on the frequency-dependent conductivity. The solid curves are the observed conductivity (replotted from Ref. 4) obtained by Kramers-Kronig transformation of the reflectivity of $YBa_2Cu_3O_{7-\delta}$ samples. (a) from Ref. 4, with $T_c = 50$ K, at five different temperatures as listed, and (b) from Ref. 4, with $\delta = 0.1$ and $\delta = 0.8$ (AF). The dashed curves are the results of the theory of Brinkman and Rice, given by Eqs. (9) and (11). In the calculations, the hopping integral $|t| = 0.5$ eV, and the density of the in-plane holes $n_h = 2.5 \times 10^{21}$ cm^{-3} in (a) and $n_h = 3.4 \times 10^{21}$ cm^{-3} in (b). A flat background has been included in $\sigma(\omega)$ to approximately include the effects of the chains as indicated by the dashed horizontal lines.

At low frequencies, $\omega \lesssim 2000^{-1}$, the experimental $\sigma(\omega)$ show strong temperature dependence [this can be seen from Fig. 2(a), also from Ref. 4], indicating strong interaction between the moving holes and the background. As discussed above, Eq. (9) for the conductivity is evaluated under the assumption $\omega \gg J$. If we were to simply continue to $\omega = 0$, then the theory of Ohata and Kubo[10] and Ref. 5 for the temperature dependence of the mobility of independent holes gives $\mu(T) \propto T^{-1}$ (or the resistivity $\rho \propto T$) but only for higher temperatures ($T \gtrsim czt$ where c is a numerical constant ≈ 0.1). At lower T, there is a crossover to $T^{-1/2}$ behavior. However, this theory is not applicable to real $Cu\text{-}O_2$ planes because of the large value of J and the finite density of holes. Note, however, that the experimental result that $\rho(T) \propto T$, as $T \to 0$ requires additional structure in $\sigma(\omega, T)$ to preserve the sum rule. A detailed discussion of this low (ω, T) behavior is beyond the scope of this work. There are two approaches in the literature. One concentrates on charge carriers with a broad bandwidth ($\sim 2zt$) which are strongly coupled to spin excitations. This is the approach taken by Anderson and Zou,[11] who describe the transport properties with a strongly coupled holon-spinon model. A related interpretation in terms of carriers strongly coupled to excitations is discussed in the paper of Thomas and co-workers.[4] In this approach our calculation can be viewed as a way of treating the optical matrix elements in the very strong coupling limit in which the optical transition is accompanied by the emission of many spin excitations. The second approach discusses the existence of coherent states within an energy J of the bottom of the hole band.[12–17] Kane, Lee, and Read[17] have recently given an extensive discussion of these coherent states and their influence on $\sigma(\omega)$ for $\omega \lesssim J$. Our calculation is complementary to theirs and describes optical transitions to the strongly diffusive states above the coherent band. Finally, it is worth noting that the existence of these higher energy optical transitions will strongly screen the Coulomb repulsion between holes moving in the coherent band and may thereby aid pairing.

IV. CONCLUSIONS

We find a good fit to the unusual form of optical conductivity in the optical and infrared region of the spectrum ($\omega \lesssim 2$ eV) of the high-T_c superconductors, $YBa_2Cu_3O_{7-\delta}$, using a one-band effective Hamiltonian with holes moving in a background of localized spins. The parameters used in the fit give very reasonable values for the number of carriers and hopping matrix element.

Note added in proof. Since the submission of this work we have become aware of related theoretical treatments of the frequency-dependent conductivity from carriers in Mott insulators by M. M. Mohan (unpublished) and earlier by H. G. Reik [in *Conduction in Low-Mobility Materials*, edited by N. Klein, D. S. Tannhauser, and M. Pollak (Taylor and Francis, London, 1971), p. 134]. We are grateful to Dr. Mohan for drawing our attention to these works.

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[1]J. G. Bednorz and K. A. Müller, Z. Phys. B **64**, 189 (1986).

[2]P. W. Anderson, Science **235**, 1196 (1987); P. W. Anderson, in *Frontiers and Borderlines in Many Particle Physics,* Proceedings of the Enrico Fermi International Summer School of Physics, Varenna, Italy, 1987, edited by R. A. Bvoglia and J. R. Schrieffer (North-Holland, Amsterdam, in press).

[3]Z. Schlesinger, R. T. Collins, D. L. Kaiser, and F. Holtzberg, Phys. Rev. Lett. **59**, 1958 (1987); and (unpublished).

[4]G. A. Thomas, J. Orenstein, D. H. Rapkine, M. Capizzi, A. J. Millis, R. N. Bhatt, L. F. Schneemeyer, and J. V. Waszczak, Phys. Rev. Lett. **61**, 1313 (1988); J. Orenstein, G. A. Thomas, D. H. Rapkine, A. J. Millis, L. F. Schneemeyer, and J. V. Waszczak, in *Proceedings of the International Conference on High-Temperature Superconductors and Materials and Mechanisms of Superconductivity, Interlaken, Switzerland, 1988,* edited by J Müller and J. L. Olsen [Physica C **153–155**, 1740 (1988)].

[5]W. F. Brinkman and T. M. Rice, Phys. Rev. B **2**, 1324 (1970).

[6]F. C. Zhang and T. M. Rice, Phys. Rev. B **37**, 3759 (1988).

[7]R. Joynt, Phys. Rev. B **37**, 7979 (1988).

[8]P. Lederer and Y. Takahashi, Z. Phys. B **71**, 415 (1988).

[9]D. Baeriswyl, C. Gros, and T. M. Rice, Phys. Rev. B **35**, 8391 (1987).

[10]N. Ohata and R. Kubo, J. Phys. Soc. Jpn. **28**, 1402 (1970).

[11]P. W. Anderson and Z. Zou, Phys. Rev. Lett. **60**, 132 (1988); *ibid.* **60**, 2557 (1988). See also, C. Kallin and A. J. Berlinsky, Phys. Rev. Lett. **60**, 2556 (1988).

[12]E. L. Nageev, Zh. Eksp. Teor. Fiz. **58**, 1269 (1970) [Sov. Phys. JETP **31**, 682 (1970)].

[13]S. A. Trugmann, Phys. Rev. B **37**, 1579 (1988).

[14]S. Schmitt-Rink, C. M. Varma, and A. E. Ruckenstein, Phys. Rev. Lett. **60**, 2793 (1988).

[15]S. Sachdev (unpublished).

[16]B. I. Shraiman and E. D. Siggia, Phys. Rev. Lett. **61**, 467 (1988).

[17]C. L. Kane, P. A. Lee, and N. Read (unpublished).