Eur. Phys. J. B **5**, 413–417 (1998)

THE EUROPEAN PHYSICAL JOURNAL B

Orbital degrees of freedom and electron correlation in optical absorption of metallic $La_{1-x}Sr_xMnO_3$[⋆]

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Received: 13 February 1998 / Accepted: 17 March 1998

Abstract. The optical absorption in ferromagnetic metal $La_{1-x}Sr_xMnO_3$ is anomalous; it has a wide-range absorption up to about 1 eV even at zero temperature. Since 3d electrons in $La_{1-x}Sr_xMnO_3$ partially fill doubly degenerate e_g orbitals, the orbital degrees of freedom are crucial to understand this metallic system. We argue that the interband transition within e_g orbitals is important in the optical absorption. The optical spectrum is modified also by the inter-orbital Coulomb interaction. We have examined perturbatively the effect of the Coulomb interaction on the spectrum. Available experiments are discussed by comparing with the present results.

PACS. 71.27.+a Strongly correlated electron systems; heavy fermions – 71.30.+h Metal insulator transitions and other electronic transitions – 78.20.-e Optical properties of bulk materials and thin films

1 Introduction

$La_{1-x}Sr_xMnO_3$ are typical transition-metal oxides with the perovskite-type structure ABO_3 in which a systematic replacement of trivalent element A and transition element B in ABO_3 provides us with a variety of insulating and metallic phases [1–3]. The end member $LaMnO_3$ is an insulator, in which the so-called A-type antiferromagnetic order (*i.e.*, antiferromagnetically stacked ferromagnetic layers) is realized. It is believed that the e_g orbitals are also ordered to assist in stabilizing this magnetic order. As Sr is doped into $LaMnO_3$, the system becomes metallic and ferromagnetic beyond $x \sim 0.17$. No orbital order has been observed in the ferromagnetic metallic phase, suggesting that the orbital degrees of freedom are quenched.

A recent optical conductivity in $La_{1-x}Sr_xMnO_3$ ($x = 0.175$ and 0.3) [4] shows that excitations are present in a wide energy region from ~ 0.02 eV to ~ 1 eV. This is often called "incoherent absorption", the origin of which has been a matter of controversy. The main issue is the nature of orbital degrees of freedom in ferromagnetic metallic phase. The intensity of the "incoherent part" *increases* with the decrease of temperature, being finite at zero temperature. This has led some researchers to suggest that an orbital-charge separated non-Fermi liquid, which is entirely different from the Fermi liquid, is realized in this three-dimensional system [5]. In contrast to this view the present authors have proposed in a previous paper [6] that the excitations detected in optical conductivity must be related to interband transitions within e_g orbitals and that the orbital dependence of transfer integrals, whose importance in the perovskite-type transition metal oxides has been noted before in a different context [7], makes the transitions allowed. The purpose of this paper is to study this problem in more detail, *i.e.*, in particular, the effect of Coulomb interaction on the optical absorption, which is important in transition-metal oxides and to show that the anomalous "incoherent absorption" can be understood, at least qualitatively, within the standard Fermi liquid approach applied to the orbitally degenerate bands.

This paper is arranged as follows. In Section 2 our model is described and the magnitude of the parameters are estimated. The constraints from available experiments are critically discussed. The effect of Coulomb interaction on the optical absorption is presented in Section 3. Finally a comparison is made with experimental results and related theoretical problems are also discussed.

2 Model and magnitude of parameters

In this paper the simplest tight-binding model is used for the electrons in Mn e_g orbitals of $La_{1-x}Sr_xMnO_3$. The $2p$ orbitals of oxygen are not taken into account explicitly; therefore the optical spectrum we present is valid only below the charge-transfer energy. The e_g orbitals are Hund-coupled with localized electrons in t_{2g} orbitals. It is believed that the double exchange mechanism stabilizes the ferromagnetism in metallic $La_{1-x}Sr_xMnO_3$ [8–11]. We keep electrons with up spin only in e_g orbitals which are doubly degenerate, since our discussion is focussed on the low-temperature limit of the ferromagnetic state. (The temperature dependence has been discussed

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[⋆] Dedicated to J. Zittartz on the occasion of his 60th birthday

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in [12].) Then we have the following model:

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_1 \tag{2.1}$$

$$\mathcal{H}_0 = \sum_{ij\alpha\beta} t_{ij}^{\alpha\beta} c_{i\alpha}^{\dagger} c_{j\beta} \tag{2.2}$$

$$\mathcal{H}_1 = \tilde{U} \sum_{j} n_{j1} n_{j2}, \tag{2.3}$$

where α and β stand for one of the two e_g orbitals; $\alpha = 1$ and 2 represent $3z^2 - r^2$ and $x^2 - y^2$ orbitals, respectively. $c_{j\alpha}$ is the annihilation operator of Mn $3d$ electron on the α orbital of site j. $\tilde{U}(= U' - V)$ is an effective Coulomb interaction in which U' is the Coulomb interaction between two electrons on $3z^2 - r^2$ and $x^2 - y^2$ orbitals and V is the Hund coupling within the e_g orbitals [7]. The model (2.1) is similar to the Hubbard model; an important difference is, however, the orbital dependence of transfer integral $t_{ij}^{\alpha\beta}$ which comes from the wavefunction of $3z^2 - r^2$ and $x^2 - y^2$ orbitals relative to the vector $\boldsymbol{R}_{ij}$ connecting two neighboring Mn ions [7]. This point becomes crucial later. The crystal structure of La$_{1-x}$Sr$_x$MnO$_3$ for $x = 0.17 \sim 0.3$ is rhombohedral, but we assume the cubic symmetry for simplicity since the lattice is close to cubic.

The hopping term in (2.2) takes the following form:

$$\mathcal{H}_0 = \sum_{k\alpha\beta} \varepsilon^{\alpha\beta}(\mathbf{k}) c_{k\alpha}^{\dagger} c_{k\beta}, \tag{2.4}$$

where $\varepsilon^{\alpha\beta}$ is the Fourier transform of $t_{ij}^{\alpha\beta}$. $c_{k\alpha}$ is the annihilation operator of electron with wave vector $\mathbf{k}$ and orbital α. Using the e_g wave functions, one obtains from $t_{ij}^{\alpha\beta}$

$$\varepsilon^{\alpha\beta}(\mathbf{k}) = \varepsilon_0(\mathbf{k})\delta_{\alpha\beta} + \varepsilon_1(\mathbf{k})(\tau^z)_{\alpha\beta} + \varepsilon_2(\mathbf{k})(\tau^x)_{\alpha\beta}, \tag{2.5}$$

where τ^z and τ^x are Pauli matrices. $\varepsilon_0(\mathbf{k})$, $\varepsilon_1(\mathbf{k})$ and $\varepsilon_2(\mathbf{k})$ are given by

$$\varepsilon_0(\mathbf{k}) = t(\cos k_x + \cos k_y + \cos k_z), \tag{2.6}$$

$$\varepsilon_1(\mathbf{k}) = \frac{t}{2}(\cos k_x + \cos k_y - 2\cos k_z), \tag{2.7}$$

$$\varepsilon_2(\mathbf{k}) = -\frac{\sqrt{3}t}{2}(\cos k_x - \cos k_y). \tag{2.8}$$

Here and henceforth we take $a = \hbar = 1$, where a is the lattice constant. The off-diagonal matrix element $\varepsilon_2(\boldsymbol{k})$ originates from nonzero electron hopping between $3z^2 - r^2$ on a Mn ion and $x^2 - y^2$ on a neighboring Mn ion in the x-y plane. The eigenvalues of the 2×2 matrix $\varepsilon^{\alpha\beta}$, which are e_g bands in the tight-binding approximation, are as follows:

$$E_{\pm}(\mathbf{k}) = \varepsilon_0(\mathbf{k}) \pm \sqrt{\varepsilon_1^2(\mathbf{k}) + \varepsilon_2^2(\mathbf{k})}. \tag{2.9}$$

$E_{\pm}(\mathbf{k})$ has the cubic symmetry, as expected.

The model (2.1) has just one dimensionless parameter $\tilde{U}/t$. By comparing (2.9) with the e_g-band width in band-structure calculations [13] the value of t suitable for La$_{1-x}$Sr$_x$MnO$_3$ is estimated as $t = 0.3 \sim 0.4$ eV; the total band width $6t$ is $2 \sim 2.5$ eV. On the other hand $\tilde{U}$ is probably $2 \sim 2.5$ eV. This value is smaller than 3 eV used previously [17]; however, because of admixture of the e_g wave functions with oxygen $2p$ orbitals, one expects $\tilde{U}$ to be reduced to some extent. Summing up, we obtain

$$\frac{\tilde{U}}{6t} = 0.8 \sim 1.3. \tag{2.10}$$

This means the metallic La$_{1-x}$Sr$_x$MnO$_3$ ($x \sim 0.3$) should be regarded as an *intermediate coupling* system. This estimation is supported also from the following experimental facts:

(1) No orbital order is observed in the ferromagnetic metallic phase. In fact if the strong coupling ($\tilde{U}/6t \gg 1$) were appropriate as claimed by Ishihara *et al.* [5], one should expect an orbital order of some sort in the present cubic system.

(2) The γ value of the specific heat is as modest as $\gamma = 3 \sim 4$ mJ/K^{2}mole [14,15]. This magnitude can be naturally interpreted within the band-structure calculation [14].

(3) From the Hall coefficient the carrier concentration is estimated as $n \sim 1$ hole/Mn-site [16], suggesting a "large Fermi surface". On the other hand, the orbital-charge separated non-Fermi liquid [5] predicts a "small Fermi surface", which contradicts with the observed Hall coefficient.

3 Doubly degenerate e$_g$ bands and optical absorption

The frequency-dependent conductivity $\sigma_{\mu\mu}(\omega + i\delta)$ ($\delta \to +0$) can be most conveniently calculated from the Kubo formula

$$\sigma_{\mu\mu}(\omega + i\delta) = \frac{-i}{\omega + i\delta}\Big[K_{\mu\mu}(\omega + i\delta) - K_{\mu\mu}(0)\Big], \tag{3.1}$$

where $K_{\mu\mu}(\omega + i\delta)$ is obtained from the thermal Green function

$$K_{\mu\mu}(i\omega_m) = \int_0^{\beta} d\tau e^{i\omega_m \tau} \langle T[J_\mu(\tau) J_\mu(0)] \rangle \tag{3.2}$$

via analytic continuation $K_{\mu\mu}(i\omega_m) \to K_{\mu\mu}(\omega + i\delta)$. ω_m is the Matsubara frequency, $\omega_m = 2m\pi k_{\mathrm{B}}T$ (m: integer).

The current operator along the μ direction, $J_\mu = (-e)\dot{R}_\mu$, is given by

$$J_\mu = \sum_{k\alpha\beta} j_\mu^{\alpha\beta}(\mathbf{k}) c_{k\alpha}^{\dagger} c_{k\beta}, \tag{3.3}$$

where $j_\mu^{\alpha\beta}(\mathbf{k}) = (-e)\partial\varepsilon^{\alpha\beta}(\mathbf{k})/\partial k_\mu$. The same result is also obtained by introducing the Peierls phase factor to $t_{ij}^{\alpha\beta}$ as $t_{ij}^{\alpha\beta} \exp(i(-e)\mathbf{A} \cdot (\mathbf{r}_i - \mathbf{r}_j)/c)$ and expanding it in terms of the vector potential $\mathbf{A}$. From (2.5) $j_\mu^{\alpha\beta}(\mathbf{k})$ has the following form:

$$j_\mu^{\alpha\beta}(\mathbf{k}) = j_{\mu 0}(\mathbf{k})\delta_{\alpha\beta} + j_{\mu 1}(\mathbf{k})(\tau^z)_{\alpha\beta} + j_{\mu 2}(\mathbf{k})(\tau^x)_{\alpha\beta}. \tag{3.4}$$

The expression for $j_{\mu 0}$, $j_{\mu 1}$ and $j_{\mu 2}$ is evident from $j_\mu^{\alpha\beta}(\mathbf{k}) = (-e)\partial\varepsilon^{\alpha\beta}(\mathbf{k})/\partial k_\mu$. Note that the 2×2 matrices $\varepsilon^{\alpha\beta}(\boldsymbol{k})$ and $j_\mu^{\alpha\beta}(\boldsymbol{k})$ do not commute with each other, unless $j_{\mu 1}\varepsilon_2 - j_{\mu 2}\varepsilon_1 = 0$ holds.

Let us start from the noninteracting case [6], in which $K_{\mu\mu}(i\omega_m)$ in (3.2) is simply given by

$$K_{\mu\mu}(i\omega_m) = -\frac{1}{\beta}\sum_n \sum_{\alpha\beta\alpha'\beta'} j_\mu^{\alpha\beta}(\mathbf{k}) j_\mu^{\beta'\alpha'}(\mathbf{k}) \times G_k^{\beta\beta'}(i\omega_m + i\varepsilon_n) G_k^{\alpha'\alpha}(i\varepsilon_n), \tag{3.5}$$

where $G_k^{\alpha\beta}(i\varepsilon)$ is the Fourier transform of the one-particle Green function

$$G_k^{\alpha\beta}(\tau) = -\langle T[c_{k\alpha}(\tau)c_{k\beta}^\dagger(0)]\rangle. \tag{3.6}$$

From this relation, $G_k^{\alpha\beta}(i\varepsilon)$ is obtained as

$$G_k^{\alpha\beta}(i\varepsilon) = \frac{A_+^{\alpha\beta}(\mathbf{k})}{i\varepsilon - E_+(\mathbf{k})} + \frac{A_-^{\alpha\beta}(\mathbf{k})}{i\varepsilon - E_-(\mathbf{k})}, \tag{3.7}$$

where the spectral weights for the two bands are given by

$$A_+^{\alpha\beta}(\mathbf{k}) = \frac{1}{2}\Big(\delta_{\alpha\beta} + \frac{\varepsilon_1(\mathbf{k})(\tau^z)_{\alpha\beta} + \varepsilon_2(\mathbf{k})(\tau^x)_{\alpha\beta}}{\sqrt{\varepsilon_1^2 + \varepsilon_2^2}}\Big), \tag{3.8}$$

$$A_-^{\alpha\beta}(\mathbf{k}) = \frac{1}{2}\Big(\delta_{\alpha\beta} - \frac{\varepsilon_1(\mathbf{k})(\tau^z)_{\alpha\beta} + \varepsilon_2(\mathbf{k})(\tau^x)_{\alpha\beta}}{\sqrt{\varepsilon_1^2 + \varepsilon_2^2}}\Big). \tag{3.9}$$

After substituting this expression into (3.5), the real part of the conductivity is easily obtained as

$$\Re\, \sigma_{\mu\mu}(\omega + i\delta) = \Re\, \sigma_{\mu\mu}^{intraband}(\omega + i\delta) + \Re\, \sigma_{\mu\mu}^{interband}(\omega + i\delta), \tag{3.10}$$

$$\Re\, \sigma_{\mu\mu}^{intraband}(\omega + i\delta) = \int_{-\infty}^{\infty} d\varepsilon \frac{f(\varepsilon) - f(\varepsilon + \omega)}{\omega} \times \sum_k \Big[\Big(j_0 + \frac{j_1\varepsilon_1 + j_2\varepsilon_2}{\sqrt{\varepsilon_1^2 + \varepsilon_2^2}}\Big)^2 \tilde{D}_k^{(+)}(\varepsilon)\tilde{D}_k^{(+)}(\varepsilon + \omega) + \Big(j_0 - \frac{j_1\varepsilon_1 + j_2\varepsilon_2}{\sqrt{\varepsilon_1^2 + \varepsilon_2^2}}\Big)^2 \tilde{D}_k^{(-)}(\varepsilon)\tilde{D}_k^{(-)}(\varepsilon + \omega)\Big] \tag{3.11}$$

$$\Re\, \sigma_{\mu\mu}^{interband}(\omega + i\delta) = \int_{-\infty}^{\infty} d\varepsilon \frac{f(\varepsilon) - f(\varepsilon + \omega)}{\omega} \sum_k \Big(\frac{j_1\varepsilon_2 - j_2\varepsilon_1}{\sqrt{\varepsilon_1^2 + \varepsilon_2^2}}\Big)^2 \times \Big[\tilde{D}_k^{(-)}(\varepsilon)\tilde{D}_k^{(+)}(\varepsilon + \omega) + \tilde{D}_k^{(+)}(\varepsilon)\tilde{D}_k^{(-)}(\varepsilon + \omega)\Big] \tag{3.12}$$

where $\sigma^{intraband}$ and $\sigma^{interband}$ are intraband and interband contributions, respectively. $\tilde{D}_k^{(\pm)}(\varepsilon)$ is defined by

$$\tilde{D}_k^{(\pm)}(\varepsilon) = -\frac{1}{\pi}\Im\,\Big(\frac{1}{\varepsilon + i\delta - E_\pm(\mathbf{k})}\Big). \tag{3.13}$$

Because of the cubic symmetry $\sigma_{\mu\mu}(\omega + i\delta)$ for $\mu = x, y$ and z are identical.

For the noninteracting case, after integrating over ε in (3.11), the intraband part has the following form

$$\Re\, \sigma_{\mu\mu}^{intraband}(\omega + i\delta) = A_{Drude}\delta(\omega), \tag{3.14}$$

where the Drude weight A_{Drude} is given by

$$A_{Drude} = (et)^2\pi\sum_k \sin^2 k_z \Big[\Big(1 - \varepsilon_1/\sqrt{\varepsilon_1^2 + \varepsilon_2^2}\Big)^2 \times \Big\{-f'(E_+(\mathbf{k}))\Big\} + \Big(1 + \varepsilon_1/\sqrt{\varepsilon_1^2 + \varepsilon_2^2}\Big)^2 \times \Big\{-f'(E_-(\mathbf{k}))\Big\}\Big]. \tag{3.15}$$

ε_1 and ε_2 are defined in (2.7) and (2.8), respectively. $E_\pm(\mathbf{k})$ are two e_g bands in (2.9). The Drude weight is a sum of contributions from two bands with different weights, as expected. For $T = 0$ this result for the Drude weight can be obtained also from Kohn's formula by introducing the vector potential with the Peierls phase factor [18].

The interband contribution for the noninteracting case is obtained from (3.12) by integration over ε:

$$\begin{aligned}\Re\, \sigma_{\mu\mu}^{interband}&(\omega + i\delta) \\ &= \frac{4\pi}{\omega}\sum_k (j_{\mu 1}\varepsilon_2 - j_{\mu 2}\varepsilon_1)^2 \frac{f(E_-(\mathbf{k})) - f(E_+(\mathbf{k}))}{(E_+(\mathbf{k}) - E_-(\mathbf{k}))^2} \\ &\quad \times \delta(\omega - E_+(\mathbf{k}) + E_-(\mathbf{k})) &(3.16) \\ &= e^2t^4\frac{\pi}{\omega^3}\sum_k \Big[\sin^2 k_z(\cos k_x - \cos k_y)^2 \\ &\quad + \sin^2 k_x(\cos k_y - \cos k_z)^2 + \sin^2 k_y(\cos k_z - \cos k_x)^2\Big] \\ &\quad \times \Big[f(E_-(\mathbf{k})) - f(E_+(\mathbf{k}))\Big]\delta(\omega - E_+(\mathbf{k}) + E_-(\mathbf{k})). &(3.17)\end{aligned}$$

Clearly the nonvanishing of the interband contribution is due to $j_{\mu 1}\varepsilon_2 - j_{\mu 2}\varepsilon_1 \neq 0$. Therefore, if the transfer integral has no off-diagonal component (*i.e.* $t_{ij}^{\alpha\beta} \propto \delta_{\alpha\beta}$), the interband contribution vanishes. Actually the importance of the off-diagonal component is recognized in connection with the magnetic structure realized in the insulating case [7,17]. In Figure 1 the intraband and interband processes are shown together with the tight-binding energy bands. Notice the interband transition starts from zero energy because of the degeneracy of the two bands along the Γ-R line.

The integrated intensities for the Drude part and the interband transtion are defined by

$$I^{Drude} = \int_0^\infty d\omega \Re\sigma_{\mu\mu}^{Drude}(\omega + i\delta) = \frac{1}{2}A_{Drude}, \tag{3.18}$$

$$I^{interband} = \int_0^\infty d\omega \Re\sigma_{\mu\mu}^{interband}(\omega + i\delta). \tag{3.19}$$

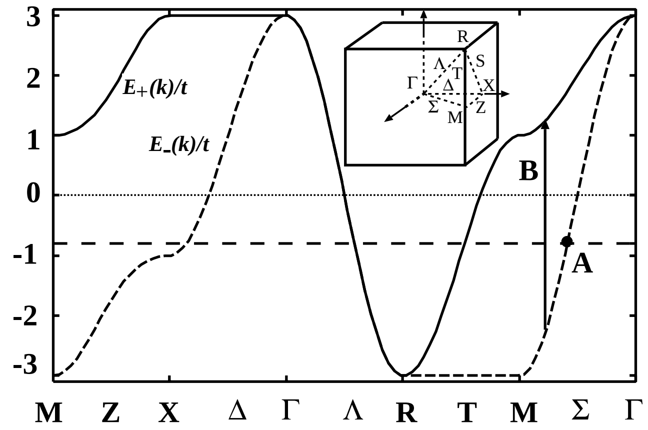


Fig. 1. The intraband (A) and interband (B) processes within the e_g bands. The broken line represents the Fermi energy. The inset shows the Brillouin zone and the symmetry points of the simple cubic lattice.

The ratio $I^{Drude}/I^{interband}$ for the noninteracting case is ~ 1.2 for the hole concentration $x = 0.3$ and insensitive to x in $0.175 < x < 0.3$.

Now we turn to the interaction effects on $\sigma_{\mu\mu}(\omega + i\delta)$. As mentioned in Section 2, there are several reasons to believe that the effective Coulomb interaction $\tilde U$ is not extremely large in metallic La$_{1-x}$Sr$_x$MnO$_3$ ($\tilde U/6t = 0.8 \sim 1.3$) so that the perturbational treatment can capture the essential point. This is equivalent to say that the Fermi liquid approach is valid for the present case. Led by this reasoning, the simplest second-order perturbation in $\tilde U$ is used for the self-energy part $\Sigma(\varepsilon + i\delta)$ ignoring the k dependence. This means that only the local correlation effect is taken into account in the same spirit as the dynamical mean-field theory [19]. Then one can easily show that the self-energy part is diagonal and independent of the orbitals. Within this local approximation the vertex correction due to $\tilde U$ is absent. Thus the correlation effect is included into (3.10)∼(3.12) by replacing (3.13) with

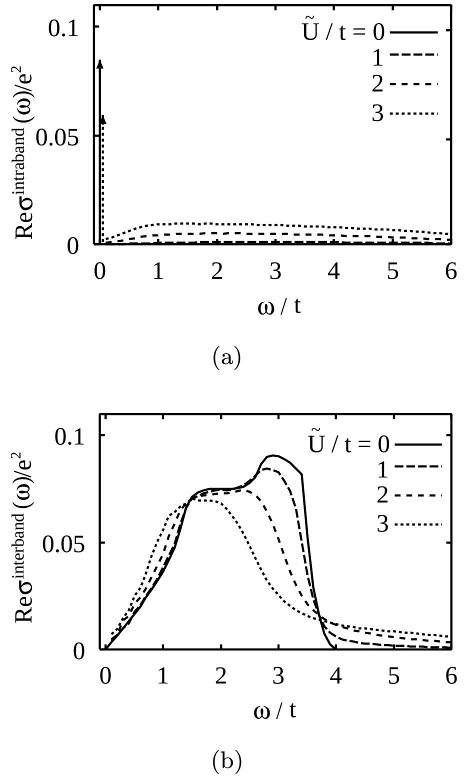
$$\tilde D_k^{(\pm)}(\varepsilon) = -\frac{1}{\pi}\Im\left(\frac{1}{\varepsilon - \Sigma(\varepsilon + i\delta) - E_\pm(\mathbf{k})}\right)\cdot \qquad (3.20)$$

$\Sigma(0)$ giving a shift of the chemical potential is taken into account in μ; therefore $\Sigma(\varepsilon + i\delta) - \Sigma(0)$ is used actually for $\Sigma(\varepsilon + i\delta)$.

The intraband and interband conductivities calculated in this way are presented in Figures 2a and 2b, respectively. First, the Coulomb interaction produces incoherent absorption in the intraband part at the expense of the reduction of the Drude weight. It is easy to show from (3.11) that the Drude weight A_{Drude} is given by

$$\frac{A_{Drude}}{A^{(0)}_{Drude}} = \frac{1}{1 - \left.\frac{\partial\Sigma(\varepsilon+i\delta)}{\partial\varepsilon}\right|_{\varepsilon\to 0}}, \qquad (3.21)$$

where $A^{(0)}_{Drude}$ is the Drude weight for $\tilde U = 0$, *i.e.*, (3.15). Second, the interband conductivity is modified by $\tilde U$ in such a way that the peak position is shifted to lower frequency as $\tilde U$ increases and that $\tilde U$ causes the high-energy tail due to inelastic scattering.



(a)

(b)

Fig. 2. The intraband (a) and interband (b) absorption for various values of $\tilde U/t$. The chemical potential is assumed as $\mu/t = -1.0$, which corresponds to $x \sim 0.3$.

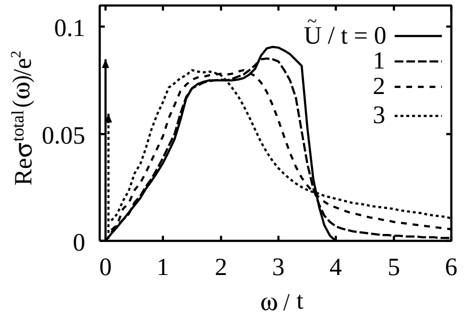


Fig. 3. The total optical conductivity for various $\tilde U/t$. The chemical potential is the same as in Figure 2.

The total conductivity is shown in Figure 3. As far as the finite-energy region is concerned, the interband contribution is clearly dominant within the magnitude of $\tilde U$ shown here. As mentioned in Section 2, the $\tilde U$ value realistic for La$_{1-x}$Sr$_x$MnO$_3$ is estimated as $\tilde U/t = 4.8 \sim 7.8$. Since the simplest second-order perturbation in $\tilde U$ is used, we took modest values ($\tilde U/t \le 3$) within the applicability limit. However, the tendency in the interaction effect on the optical absorption is evident at this stage.

4 Discussion and conclusion

Let us compare our results with available experiments. Looking at the experimental spectrum in [4], we notice that the broad "incoherent absorption" from 0.02 eV to 1 eV which is observed in the fully spin-polarized state at $T = 0$ K is qualitatively well reproduced in our result, *i.e.*, Figure 3. The frequency dependence of $\Re\,\sigma_{\mu\mu}(\omega + i\delta)$ shows that the peak due to the interband absorption shifts to lower frequency as $\tilde{U}$ increases. The spectrum for larger $\tilde{U}$ shows a better agreement with the experimental spectrum. Therefore we conclude that the electron correlation is important. According to our result the broad "incoherent absorption" is due to the interband absorption within the e_g bands which is modified by the electron correlation. The frequency region is consistent with the interband absorption, while the frequency dependence requires the electron correlation. To achieve quantitative agreement with experiments the theory has to be improved by going beyond the simplest second-order perturbation and the local approximation. It is left for a future study.

Our study is essentially a Fermi-liquid approach starting from the weak-coupling side. According to widely accepted wisdom the Fermi statistics usually dominates low-energy physics in *three-dimensional itinerant* systems. We believe most experimental results on metallic $La_{1-x}Sr_xMnO_3$ are consistent with this view. In our treatment a possible coupling with lattice distortions is not included. The reason is, first, there is no experimental evidence suggesting a strong electron-lattice coupling in the *metallic phase*, which is nearly cubic. This is in contrast to the insulating phase where the Jahn-Teller distortion is actually observed. Second, we are interested in the frequency region up to 1 eV, which is so high that the lattice displacement cannot follow the oscillating electric polarization.

Finally, let us consider what happens if one starts from the strong-coupling limit ($\tilde{U}/t \to \infty$) as Ishihara *et al.* did [5]. First, if such a limit is appropriate, the ground state should have an orbital long-range order of some sort in three-dimensional systems like $La_{1-x}Sr_xMnO_3$ ($x \sim 0.3$), where no orbital order is observed experimentally. This means that any state *without* orbital order is higher in energy for $\tilde{U}/t \to \infty$. Thus, if one develops a theory from a state without orbital order, the starting point is inevitably unstable. Second, under the local constraint due to $\tilde{U}/t \to \infty$, Ishihara *et al.* express $c_{i\alpha}$ as

$$c_{i\alpha} = h_i^\dagger b_{i\alpha} \tag{4.1}$$

where h_i is "orbital-less" *fermion* (holon) and $b_{i\alpha}$ is a *boson* for orbital. Here the local constraint $h_i^\dagger h_i + \sum_\alpha b_{i\alpha}^\dagger b_{i\alpha} = 1$ should be satified for *any site i*. Let us recall that one can equally well assume that h_i is a *boson* (holon) and $b_{i\alpha}$ is a *fermion*. If the local constraint is exactly taken into account, both representations give identical results. However, if the local constraint is replaced by a "grobal' one (*i.e.*, so-called "mean-field" treatment), the results depend on which statistics one chooses. This is clearly unphysical. For these two reasons we think the weak coupling approach is more suitable than the strong-coupling approach, as far as the metallic phase of $La_{1-x}Sr_xMnO_3$ is concerned.

We thank Y. Tokura for showing us his group's data prior to publication. We are indebted to N. Hamada and R. Shiina for helpful discussions. This work is supported by Grant-in-Aid for Scientific Research from the Ministry of Education, Science and Culture.

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