Orbital-selective Mass Enhancements in Multi-band Ca$_{2-x}$Sr$_x$RuO$_4$ Systems Analyzed by the Extended Drude Model

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We investigated optical spectra of quasi-two-dimensional multi-band Ca$_{2-x}$Sr$_x$RuO$_4$ systems. The extended Drude model analysis on the ab-plane optical conductivity spectra indicates that the effective mass should be enhanced near $x=0.5$. Based on the sum rule argument, we showed that the orbital-selective Mott-gap opening for the $d_{x^2-y^2}$ bands, the widely investigated picture, could not be the origin of the mass enhancement. We exploited the multi-band effects in the extended Drude model analysis, and demonstrated that the intriguing heavy mass state near $x=0.5$ should come from the renormalization of the $d_{xy}$ band.

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The Mott metal-insulator transition has been one of major topics in condensed matter physics for many years. It is well known that the conduction electron mass could be enhanced in metals due to strong electron correlations, and that it should diverge toward a metal-insulator transition near $x=0.5$. Here $W$ for the Ru $t_{2g}$ states should keep decreasing due to the internal chemical pressure from substituting the Sr$^{2+}$ ion with the smaller Ca$^{2+}$ ion, which gives rise to a transition from a multi-band metal ($x=2.0$) to a Mott-insulator ($x=0.0$). Contrary to the simple expectation of the gradual change in $W$ with $x$, these compounds exhibit quite unusual behavior near $x=0.5$. While the metal-insulator transition occurs for $x=0.2$, anomalous enhancements of magnetic susceptibility $\chi(0)$ and specific heat coefficient $\gamma$, indicating a heavy mass state, were observed near $x=0.5$.

In order to explain these surprising experimental behaviors, numerous workers have paid attention to the directional dependences of the $d_{xy}$ behaviors near $x=0.5$. The extended Drude model analysis on the ab-plane optical conductivity spectra indicates that the effective mass should be enhanced near $x=0.5$. Based on the sum rule argument, we showed that the orbital-selective Mott-gap opening for the $d_{x^2-y^2}$ bands, the widely investigated picture, could not be the origin of the mass enhancement. We exploited the multi-band effects in the extended Drude model analysis, and demonstrated that the intriguing heavy mass state near $x=0.5$ should come from the renormalization of the $d_{xy}$ band.

Optical spectroscopy has been used as a powerful method to investigate metal-insulator transitions of strongly correlated electron systems. Especially, the extended Drude model (EDM) has been extensively used to explain the electrodynamic responses near metal-insulator transitions of numerous correlated electron systems, such as heavy fermion compounds and high transition temperature superconductors. This phenomenological theory describes optical conductivity spectra $\tilde{\sigma}(\omega) = \sigma_1(\omega) + i\sigma_2(\omega)$ of free carriers in terms of the frequency-$\omega$-dependent scattering rate $1/\tau(\omega)$ and the mass enhancement $\lambda(\omega)$ (effective mass $m^*(\omega)$ normalized by the band mass $m_0$), which is given as

$$\lambda(\omega) = \frac{\omega_p^2}{4\pi} \cdot \frac{\sigma_2(\omega)}{\omega|\tilde{\sigma}(\omega)|^2}. \tag{1}$$

Here, $\omega_p$ is the plasma frequency. However, it should be noted that the free carriers in this model were assumed to originate from a single band. Up to this point, there have been no efforts to extend this analysis to the multi-band metallic compounds.

Even for a multi-band system, $\tilde{\sigma}(\omega)$ is an averaged response for all $k$-space. It contains all the contributions from each independent band, so it should be expressed as a linear addition of such contributions. As a result, the optical sum rule should be valid for the multi-band system, and the total spectral weight of the coherent peak $\omega_p^2$ should be $\omega_p^2 = \sum_i \omega_{p,i}^2$. By expressing the contribution from each band in terms of EDM and comparing the sum with Eq. (1), we obtained

$$\lambda(\omega) = \sum_i \omega_{p,i}^2 \sum_i \omega_{p,i}^2 \tau_i^2(\omega) \lambda_i(\omega) / \left( \sum_i \omega_{p,i}^2 \tau_i(\omega) \right)^2. \tag{2}$$

When all of $\tau_i$ are nearly the same, $\lambda(\omega)$ obtained from Eq. (1) corresponds approximately to the averaged value of contributions from each band weighted by $\omega_{p,i}^2$, i.e.,

$$\lambda(\omega) \approx \sum_i \omega_{p,i}^2 \lambda_i(\omega) / \sum_i \omega_{p,i}^2. \tag{2}$$

For the CSRO system, superscripts $\alpha$ and $\beta$ are commonly used to indicate two bands formed by the $d_{yz}$
analysis, between 5 meV and 30 eV. Using the Kramers-Kronig activity spectra were measured in a photon energy range ≤ of CSRO (0.06 in-plane. Explain the frequency limit. While the simple Drude model cannot explain the EDM would be useful to get information about the changes of electrodynamic parameters.

Before going into a detailed discussion about the mass state should be contributed to by a strong renormalization of the in-plane orbital states. And let us assign the $d_{xz}$, $d_{yz}$, and $d_{xy}$ bands as $i = 1$, 2, and 3, respectively.

In this Letter, we investigated the doping-dependent in-plane $\sigma_1(\omega)$ taking into account of the multi-band effects. Based on the sum rule, we could argue that the controversial $d_{yz/zx}$ bands would not experience the OSMT. Instead, we could demonstrate that the heavy mass state should be contributed to by a strong renormalization of the $d_{xy}$ band. We will provide additional indirect evidence for this intriguing picture from the $x$-dependent changes of $p-d$ charge transfer excitations.

High quality CSRO single crystals were grown by the floating zone method. Near-normal incident reflectivity spectra were measured in a photon energy range between 5 meV and 30 eV. Using the Kramers-Kronig analysis, $\sigma_1(\omega)$ were obtained.

Figure 1 shows the doping-dependent $\sigma_1(\omega)$ of CSRO (0.06 ≤ $x$ ≤ 2.0) at room temperature. All of the $\sigma_1(\omega)$ curves show strong coherent peaks in the zero frequency limit. While the simple Drude model cannot explain the $\omega$-dependence of these $\sigma_1(\omega)$, application of the EDM would be useful to get information about the changes of electrodynamic parameters.

Before going into a detailed discussion about the mass enhancement changes in CSRO using Eq. (1), it is worthwhile to mention how we obtained $\omega_p$ values. First, we estimated $\omega_p$ by integrating the spectral weight of the coherent peak using $\omega_p^2 = 8 \int_0^{\omega_p} \sigma_1(\omega) d\omega$. Here, $\omega_c$ is the highest cutoff frequency for the free carrier response. As shown in Fig. 1(a), an isosbestic point exists around 1.5 eV: as $x$ decreases, the spectral weight below 1.5 eV decreases, but that of the higher energy region increases. So, we chose $\omega_c$ as 1.5 eV. The $x$-dependent values of $\omega_p^2$ are displayed as solid triangles in Fig. 1(b), where $\omega_p^2$ keeps decreasing down to $x=0.6$. Second, we determined $\omega_p^2$ from $\varepsilon_1(\omega) = \varepsilon_\infty - 4\pi \sigma_1(\omega)/\omega$ using the relation $\varepsilon_1(\omega_p/\sqrt{\varepsilon_\infty}) = 0$. $\varepsilon_1(\omega)$ for two representative metallic compounds, i.e., $x=2.0$ and 0.20, are shown in the inset of Fig. 1(a). They show monotonic $\omega$-dependent increases crossing a zero level around 1.3 eV. By adopting $\varepsilon_\infty = 5.8$, we estimated $\omega_p^2$ and displayed them as open circles in Fig. 1(b), which show behavior quite similar to that of the solid triangles. For further discussion in this paper, we will adopt the $\omega_p^2$ values estimated by the first method.

Figures 2(a) and 2(b) show the results for $\lambda(\omega)$ obtained from Eq. (1). As $x$ decreases, the $\lambda(\omega)$ curves show a small upward shift down to $x=0.5$ [Fig. 2(a)], and then they exhibit a rather significant downward shift below $x=0.5$ [Fig. 2(b)]. Figure 2(c) shows the $x$-dependent changes of $\lambda(\omega)$ at the low frequency limit, i.e., 200 cm$^{-1}$, which displays a peak near $x=0.5$. This $x$-dependent change is reminiscent of the enhancement of $\gamma$ shown as a solid line in Fig. 2(c) [4]. These unusual behaviors suggest that the Fermi surface topologies must experience anomalies near $x=0.5$.

The de Haas-van Alphen (dHvA) measurement could...
be a very powerful tool to investigate Fermi surface topologies of very pure samples. Mackenzie et al. performed the dHvA measurements on Sr$_2$RuO$_4$. They observed quantum oscillations coming from three $t_{2g}$ bands, two from the $d_{yz/zx}$ bands and one from $d_{xy}$ band. They found that $n_1:n_2:n_3 \approx 1:1:1$ and $m_{b1}:m_{b2}:m_{b3} \approx 1:2:3$, where $n_i$ is the free carrier density of the $i$-th band. Since $\omega_{p,i}^2 = 4\pi\epsilon^2/m_i$, $\omega_{p,1}^2:\omega_{p,2}^2:\omega_{p,3}^2 \approx 6:3:2$. And, they also reported that $\lambda_1(0):\lambda_2(0):\lambda_3(0) \approx 3:3:3$. From these values, we can estimated $\lambda(0) = 3.3$ from Eq. (2). This value is well consistent with the optically observed value ($\sim 3.5$), shown as the solid circle on the y-axis of Fig. 2(a), demonstrating the validity of our EDM analysis for the multi-band system.

On the other hand, angle-resolved photoemission spectroscopy (ARPES) studies could be another way to look into the Fermi surface. Recently, Wang et al. performed ARPES measurements on an $x=0.5$ sample, and showed that its Fermi surface topology remains nearly the same as that of an $x=2.0$ sample. They claimed that the OSMT should not occur in the CSRO system. The photoemission studies are generally sensitive to the surface (not to the bulk), and the ARPES study could not explain why the heavy mass state does exist near the $x=0.5$ sample.

Optical spectroscopy could provide new insights on OSMT of the CSRO system. We will test the model of the OSMT in the $d_{yz/zx}$ bands, which is a seemingly fascinating and the most widely investigated scenario 6,8 to explain the anomalous heavy mass state around $x=0.5$. Using the dHvA measurement results 17, the ratio between $\omega_{p,2}^2$ and $(\omega_{p,1}^2+\omega_{p,2}^2)$ for $x=2.0$ could be estimated to be about $1/4.5$. If the $d_{yz/zx}$ bands open a Mott gap, the optical sum rule states that the reduction in $\omega_{p}^2$ should reach around $82\%$ of $\omega_{p}^2$. However, as shown in Fig. 1(b), $\omega_{p}^2$ for $x=0.5$ decreases only by $\sim 20\%$. Such a difference in the reduction of $\omega_{p}^2$ clearly demonstrates that the Mott-gap opening could not occur in the narrow $d_{yz/zx}$ bands.

What should be responsible for the unusual behavior near $x=0.5$? It should be noted that the enhancement of $\gamma$ between the $x=2.0$ and the $x=0.5$ samples is about 6.0, significantly higher than the corresponding value (i.e. 1.4) of $\lambda$(200 cm$^{-1}$) [See Fig. 2(c) ]. This difference comes from how the mass enhancement of each band can contribute to the total responses of $\gamma$ and $\lambda$ in the multi-band CSRO system. The specific heat coefficient $\gamma$ can be written as a linear summation of contributions from three $t_{2g}$ bands, i.e., $\gamma = \sum_1^3 \gamma_i$. On the other hand, $\lambda(\omega)$ obtained from Eq. (1) should be the averaged value of mass enhancement contributions, weighted by $\omega_{p,i}^{-2}$, as shown in Eq. (2). As an illustration, let us consider a simple multi-band system with three bands, whose plasma frequencies and masses are exactly the same. If one of the bands experiences a mass enhancement of 16 times, we can estimate that the total specific heat can be enhanced by 6 times. And from Eq. (2), assuming $m_b = m^*$, that the average $\lambda$ can be enhanced by only about 1.5 times. In a real system, $\omega_{p,2}^2$ and $\lambda(\omega)$ of each band cannot be the same. However, using a similar argument with proper guidance, we will be able to unveil the nature of the heavy mass state near the $x=0.5$ CSRO sample.

First, let us assume one limiting case (Case I), where $\omega_{p,1}^2$ would decrease with $x$, while $\omega_{p,2}^2$ and $\omega_{p,3}^2$ remain the same as those at $x=2.0$. This limiting case is illustrated in Fig. 3(a). Using Eq. (2), $\omega_{p,i}^2 = 4\pi\epsilon^2/m_{b,i}$, and $\lambda(0) = m^*_i(0)/m_{b,i}$, we could evaluate $m^*_i(0)$ 18, assuming the carrier densities $n_i$ of each band would not change with $x$ 10. As shown in Fig. 3(c), $m^*_i(0)$ cannot be significantly enhanced, failing to explain the specific heat data. The reduction of $\omega_{p,2}^2$ gives a similar result with that of Case I. Second, let us consider the other limiting case (Case II), where the $d_{xy}$ band would experience a significant change with $x$ 10, which is illustrated in Fig. 3(b). Using a similar process, we could evaluate $m^*_i(0)$. As shown with the solid squares in Fig. 3(c), $m^*_i(0)$ enhances from 21 at $x=2.0$ to around 200 at $x=0.5$, and then decreases for $x<0.5$. This change of $m^*_i(0)$ can explain that of $\gamma$ quite well. Our EDM analysis based on the multi-band extension indicates that the heavy mass state around $x=0.5$ should be contributed to by the renormalization of the $d_{xy}$ band. It is quite interesting that the $d_{xy}$ band, having a larger $W$ than the $d_{yz/zx}$ bands, could be strongly renormalized near $x=0.5$.

Finally, let us discuss how the $d_{xy}$ band could be renor-
1.5 eV to 6 eV. For the 

d

unoccupied Ru

peak structures around 3.5 and 4.7 eV. For the metallic 

case II agree quite well with the experimental 

shift, indicating that the unoccupied band having the 

dxy character must be shifted to a higher energy away 

from the Fermi energy. These changes could result in 

smaller contribution of dxy band to the metallic response 

and lead to the enhancement of its m∗(0) near x=0.5. 

From the neutron scattering experiments [3], it is well 

known that a rotation of the RuO6 octahedra along the 

c-axis increases from around x=1.5 and saturates below 

x=0.5. In this respect, the observed evolution of the dxy band 

could be attributed to the RuO6 rotation [21]. The 

detailed mechanism should be elucidated in future work 

[22].

In summary, we addressed the evolution of electronic 

structures in multi-band Ca2−xSr2RuO4 by investigating 

the optical spectra. The argument based on the optical 

sum rule revealed that dyz/zx bands should remain itinerant 

near x=0.5. Using the extended Drude model analysis, we found that the Fermi surface topologies should experience anomalies near x=0.5, and we proposed that the heavy mass state near x=0.5 should come from a strong renormalization of the dxy band.

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