Dimensional Crossover driven by Magnetic Ordering in Optical Conductivity of Pr$_{1/2}$Sr$_{1/2}$MnO$_3$

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Abstract

We investigated optical properties of Pr$_{0.5}$Sr$_{0.5}$MnO$_3$, which has the A-type antiferromagnetic ordering at a low temperature. We found that $T$-dependence of spectral weight transfer shows a clear correlation with the magnetic phase transition. In comparison with the optical conductivity results of Nd$_{0.5}$Sr$_{0.5}$MnO$_3$, which has the CE-type antiferromagnetic charge ordering, we showed that optical properties of Pr$_{0.5}$Sr$_{0.5}$MnO$_3$ near the Néel temperature could be explained by a crossover from 3D to 2D metals. Details of spectral weight changes are consistent with the polaron picture.

75.50.Cc, 75.30.-m, 75.30.Kz, 78.20.Ci
Optical properties of colossal magnetoresistance manganites have been investigated extensively, since they showed drastic spectral weight changes with temperature variation [1–6]. Especially, in ferromagnetic (FM) metallic states, their optical conductivity spectra $\sigma(\omega)$ showed a large incoherent absorption peak near 0.5 eV. It has been argued that the anomalous incoherent absorption should be related to the Jahn-Teller polaron [2–7]. However, there are alternative scenarios which attribute the incoherent feature to orbital degree of freedom [1,8–10]. Recently, Mack and Horsch investigated a planar 2D model for FM phase of manganites which develops orbital order of $e_g$ electrons with $x^2 - y^2$ symmetry [11]. With the finite-temperature diagonalization method, they calculated dynamic structure factor of orbital excitations and $\sigma(\omega)$, and suggested that $\sigma(\omega)$ in the doped $x^2 - y^2$ ordered phase can allow one to distinguish between the orbital excitation and the Jahn-Teller polaron scenarios.

A recent neutron scattering study on Pr$_{0.5}$Sr$_{0.5}$MnO$_3$ (PSMO) provided a surprising result that the heavily doped 3D manganite has a ground state with an antiferromagnetic (AF) A-structure [12]. In this state, FM planes are coupled antiferromagnetically, and conduction should occur within the planes. And, the occupied orbitals were suggested to have $x^2 - y^2$ symmetry, so optical investigation could provide an answer to the test proposed by Mack and Horsch. Moreover, the compound has two interesting phase transitions: an AF-to-FM transition of the first order at $T_N \approx 150$ K, and a FM metal-to-paramagnetic insulator transition at $T_C \approx 270$ K [13].

Another half-doped Nd$_{0.5}$Sr$_{0.5}$MnO$_3$ (NSMO) compound was found to exhibit similar phase transitions. As $T$ increases, NSMO experiences an AF insulator-to-FM metal transition at $T_N \approx 160$ K and a FM metal-to-paramagnetic insulator transition at $T_C \approx 250$ K. In fact, the low temperature AF insulator phase of NSMO becomes metallic under a high magnetic field [14], and a similar transition was observed in PSMO [13]. Despite these similar features, the low temperature phase of NSMO accompanies the CE-type AF ordering with charge and orbital order [12].

With the CE-type ordering, small hopping energy of holes along the 1D FM chain is
dominated by the charge- or orbital-interaction (also combined with electron-phonon interaction), leading to a charge ordering. On the contrary, holes in the 2D FM layer of PSMO with the $A$-type ordering may have larger hopping energies, possibly leading to a 2D metallic state competing with the charge-ordering instability. Since the conduction in the FM metallic region is 3D, a dimensional crossover could occur at $T_N$. However, such a dimensional crossover due to the magnetic ordering has not been yet investigated in PSMO.

In this letter, we report the first results of the temperature dependent $\sigma(\omega)$ for PSMO. The variation of $\sigma(\omega)$ across $T_N$ in PSMO are compared with that in NSMO, and it can be well understood in terms of a dimensional crossover driven by the $A$-type AF ordering from a 3D FM metallic state above $T_N$ into a 2D metallic state below $T_N$. Details of spectral weight changes are consistent with the polaron picture.

Single crystals of PSMO and NSMO were grown by the floating zone method [15]. For optical measurements, both samples were polished up to 0.3 $\mu$m using diamond pastes. To release strain applied during the polishing procedure, they were annealed in an oxygen atmosphere [16]. Near normal incident reflectivity spectra $R(\omega)$ were measured from 8 meV to 6.0 eV at the temperature region between 15 K and 300 K, and $\sigma(\omega)$ were obtained using the Kramers-Kronig transformation. The results were also confirmed independently by spectroscopic ellipsometry techniques. Details of the optical measurements were published previously [17].

Figure 1(a) shows $\sigma(\omega)$ of PSMO in the mid-infrared region. The sharp peaks below 0.1 eV represent optic phonon modes. Above $T_C$, there is a broad peak-like feature around 0.8 eV, followed by a rise above 2.0 eV. With cooling into the FM state, the spectral weight above 0.6 eV is transferred to a lower energy region. With further cooling below $T_N$, the spectral weight changes move in the opposite direction, i.e. from lower to higher energy regions.

Further insights on the spectral weight changes can be obtained from $\Delta\sigma(\omega) \equiv \sigma(\omega, T) - \sigma(\omega, T = 290 K)$, shown in Fig.1(b). Overall spectral weight changes in $\Delta\sigma(\omega)$ occur around 0.4 eV and 1.5 eV, which suggests that $\sigma(\omega)$ could be interpreted in terms of two-peak
As $T$ decreases, $\Delta \sigma(\omega)$ around 0.4 eV increases and reaches a maximum near $T_N$. Then, it decreases and remains finite at 15 K. As shown in the inset, a similar $T$-dependence was observed in $\Delta \sigma(\omega)$ of NSMO. However, $\Delta \sigma(\omega)$ around 0.4 eV becomes nearly zero at 15 K, which is different from the PSMO case.

Figure 2 displays $\sigma(\omega)$ of PSMO in the far-infrared (FIR) region. [For clarity, the $\sigma(\omega)$ curve at 290 K only were plotted by shifting upward by 800 $\Omega^{-1}$ cm$^{-1}$.] The peaks around 160 cm$^{-1}$, 330 cm$^{-1}$, and 580 cm$^{-1}$ represent infrared-active phonons which correspond to the external, bending, and stretching modes, respectively [19]. Around $T_C$, there are little changes in their characteristics. However, below $T_N$, the bending and the stretching modes are shifted by about 5 cm$^{-1}$ and 8 cm$^{-1}$, respectively, and their strength becomes increased. The phonon frequency changes agree with significant changes in lattice parameters accompanying the AF transition [12]. Below 200 cm$^{-1}$, Drude-like peaks in $\sigma(\omega)$ do appear for all temperatures. Even at 15 K, the Drude-like feature still can be seen.

The far-infrared $\sigma(\omega)$ of NSMO, displayed in the inset of Fig. 2, are different from those of PSMO. First, although the room temperature phonon spectrum of NSMO is similar to that of PSMO, the bending and the stretching phonon peaks are split strongly below $T_N$. These phonon splittings are very similar to those of other manganites in the CE-type charge ordered state [20]. Second, the Drude peak of NSMO vanishes completely at 15 K. The absence of the Drude peak is consistent with its high $dc$ resistivity value (i.e. about 0.43 $\Omega cm$), which arises from a strong carrier localization due to the charge ordering. The absence of the phonon splittings and the presence of the finite Drude component at $T \ll T_N$ in PSMO suggests that the electronic ground states are quite different in these two systems.

To get a better understanding on the $T$-dependent spectral weight changes, we decomposed the electronic contributions in $\sigma(\omega)$ into three parts: the FIR Drude component $\sigma_{\text{Drude}}(\omega)$, the midgap component $\sigma_{\text{ms}}(\omega)$, and the charge transfer component $\sigma_{\text{CT}}(\omega)$.

$$\sigma(\omega) = \sigma_{\text{Drude}}(\omega) + \sigma_{\text{ms}}(\omega) + \sigma_{\text{CT}}(\omega).$$

The gradual rise at $\omega > 2$ eV in Fig.1(a) corresponds to an onset of $\sigma_{\text{CT}}(\omega)$, and we used the
Lorentz oscillator model to fit this energy region. Since the significant changes of spectral weight occur around 0.4 eV and 1.5 eV, as shown in Fig. 1(b), we used two Gaussian peaks for fitting. We set the peak positions to be 0.5 eV (for Peak I) and 1.5 eV (for Peak II). Note that similar analyses were performed on La$_{1-x}$Ca$_x$MnO$_3$ quite successfully [5].

Figure 3 shows the experimental data and the fitting results of $\sigma(\omega) - \sigma_{CT}(\omega) - \sigma_{Drude}(\omega)$. Agreement between the experimental and the fitting results is quite good. A strength of each peak was obtained by integrating the corresponding spectral weight. The solid circles in Fig. 4(a) and (b) denote $S_I$ and $S_{II}$, which are strengths of Peak I and Peak II, respectively. As $T$ decreases, $S_I$ increases slightly around $T_C$ and then begins to decrease near $T_N$. The $T$-dependence of $S_{II}$ is roughly the opposite. Fig. 4(c) shows the $T$-dependence of Drude peak strength, $S_D$, which is similar to that of $S_I$. For comparisons, peak strengths for NSMO were also plotted with solid squares in Fig. 4(a), (b), and (c). In NSMO, the $T$-dependences of $S_I$, $S_{II}$, and $S_D$ are much stronger and $S_D$ becomes nearly zero below $T_N$.

Mack and Horsch investigated a planar model for the FM phase which develops orbital order of $e_g$ electrons with $x^2 - y^2$ symmetry [11]. Based on the orbital $t$-$J$ model, they provided a theoretical prediction for $\sigma(\omega)$ in the 2D FM state: $\sigma(\omega)$ should show both a Drude peak and a gapped incoherent absorption. The incoherent absorption due to the orbital excitation is expected to be peaked at $2t$, where $t$ is the hopping energy. With $t \sim 0.25$ eV, Peak I should correspond to the incoherent absorption, and the decrease of $S_I$ below $T_N$ is consistent with their prediction. However, for the coherent peak, $S_D$ is predicted to increase as $T$ is lowered, opposite to our observation in Fig.4(c). Therefore, our data cannot be explained in terms of the orbital excitation only.

Another interesting scenario is the Jahn-Teller polaron picture [3–5,7], where the Drude peak and Peak I can be interpreted as coherent and incoherent absorption peaks due to polaron motion, respectively. [Namely, Peak I can be assigned as the inter-atomic $e_g^1$(Mn$^{3+}$) $\rightarrow$ $e_g$(Mn$^{4+}$) transition, where $e_g^1$ represents the lower energy orbital of the Jahn-Teller split levels.] The increase of $S_I$ at $T < T_C$ can be interpreted in terms of the FM alignment of the Mn moments, since number of available hopping sites will increase. In the FM state, there
will be a crossover from small to large polaron states \[4\]. Coherent motion of carriers and far-IR Drude absorption will increase, which is in good agreement with Fig. 4(c). Below $T_N$, the AF ordering will prevent polaron hopping between neighboring Mn sites with opposite spins, so $S_I$ and $S_{Drude}$ should decrease, consistent with Fig. 4(a) and (c).

In earlier works \[2,5\], Peak II was assigned as a transition between the Jahn-Teller split levels: i.e. $e_g^1$(Mn$^{3+}$) $\rightarrow$ $e_g^2$(Mn$^{3+}$). Assuming that this transition is intra-atomic \[21\], the temperature dependence of $S_{II}$ can be explained. The intra-atomic transition becomes possible due to the Mn $e_g$–O 2$p$ hybridization and the local distortion of the MnO$_6$ octahedron. In the metallic states, the Jahn-Teller distortion becomes weakened due to increased screening, so the MnO$_6$ octahedron becomes more close to cubic and $S_{II}$ will be decreased. Below $T_N$, the local symmetry becomes lowered again \[12\], and $S_{II}$ becomes enhanced, consistent with Fig.4(b).

Note that the $T$-dependences of $S_I$, $S_{Drude}$ and $S_{II}$ of PSMO are weaker than those of NSMO. This implies that metallic screening is more effective, particularly for $T < T_N$. The prevalence of the screening effects indicate that in the $A$-type AF phase of PSMO, the FM layers can form 2D metallic layers with c-axis conduction forbidden. The existence of the Drude-like feature at 15 K, displayed in Fig. 2, and the finite value of $S_{Drude}$ below $T_N$, displayed in Fig. 4(c), also supports that PSMO should remain in a metallic state with the AF ordering. Then, the resistivity change of PSMO at $T_N$ should be interpreted as a dimensional crossover from 3D to 2D metallic states driven by the $A$-type AF ordering.

To further investigate this intriguing phenomenon, we looked into total absorption strength due to the polaron motion, i.e., $S_{tot} = S_{Drude} + S_I$, which is shown in Fig. 4(d). As $T$ decreases, $S_{tot}$ starts to increase around $T_C$ and then begins to decrease at $T_N$. In (La,Pr)$_{0.7}$Ca$_{0.3}$MnO$_3$, whose ground state remains a 3D FM metal at a low $T$, it was found that $S_{tot}(T)$ could be scaled with the double exchange bandwidth $\gamma_{DE}(T) = \langle \cos(\theta_{ij}/2) \rangle$ \[3\], where $\theta_{ij}$ is the relative angle of neighboring spins and $\langle \rangle$ represents thermal average in the double exchange model \[22\]. This scaling behavior was explained in a model by Röder et al. \[23\], where the double exchange and the Jahn-Teller polaron Hamiltonians were taken into
account. The dotted line in Fig. 4(d) shows $\gamma_{DE}(T)$ for the 3D metal. Above $T_N$, agreement between $S_{tot}(T)$ and $\gamma_{DE}(T)$ is quite good.

However, $S_{tot}(T)$ deviates from $\gamma_{DE}(T)$ below $T_N$. This deviation can be explained by the dimensional crossover from 3D to 2D due to the AF ordering. In the 3D FM state, around a Mn ion, there are six neighboring Mn sites with parallel spins, so they are all available for polaron hopping. In the 2D AF state, four of them have parallel spins, but the rest two have opposite spins. If we neglect the dependence of hopping on orbital directions, $S_{tot}$ in 2D should be $2/3$ of $S_{tot}$ in 3D. [Note that, due to the multi-domain nature of the crystal, we are not probing the in-plane and the c-axis conductivities separately but a statistical average of the two.] The ratio between the experimental value of $S_{tot}(15 \text{ K})$ and its theoretical value predicted from $\gamma_{DE}(15 \text{ K})$ is found to be $(0.72\pm0.06)$, which is close to $2/3=0.67$. The small deviation might come from the hopping dependence on orbital directions and/or contribution of the orbital excitation.

We want to address some interesting points for future studies. First, $S_{Drude}$ is much smaller than $S_I$. Second, the $T$-dependence of $S_{Drude}$ is stronger than that of $S_{tot}(T)$ [or $S_I$]. As $T$ decreases from $T_N$ to 15 K, $S_{Drude}$ decreases by a factor of about 3. A detailed model which includes both the orbital excitation and the polaron is highly desirable to get further understanding on $\sigma(\omega)$. A recent calculation by Kilian et al. [10] suggested that $\sigma(\omega)$ of (La,Sr)MnO$_3$ could be explained better by including the orbital and the lattice degrees of freedom together. Finally, it would be highly desirable to investigate the anisotropic nature of the 2D metallic state using single domain PSMO samples. Similar studies were done very recently on Nd$_{0.45}$Sr$_{0.55}$MnO$_3$ [24].

In summary, we investigated optical properties of Pr$_{0.5}$Sr$_{0.5}$MnO$_3$, which has the $A$-type antiferromagnetic ordering at a low temperature. Its temperature dependent spectral weight change shows a clear correlation with the magnetic phase transition. Its spectral weight changes below the Néel temperature could be explained by a crossover driven by the magnetic ordering from 3D to 2D metal.

We acknowledge Professor J.-G. Park for discussion. This work was supported by Min-
istry of Science and Technology through grant No. I-3-061, by Seoul National University Research Fund, and by the Korea Science & Engineering Foundation through RCDAMP of Pusan National University. The work by Y.M. was supported by a Grant-In-Aid for Scientific Research from the Ministry of Education, Science, Sports and Culture, and from PRESTO, JST.
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FIGURES

FIG. 1. (a) $T$-dependent optical conductivity $\sigma(\omega)$, and (b) $\Delta \sigma(\omega) \equiv [\sigma(\omega,T) - \sigma(\omega,290K)]$ of PSMO. In the inset, $\sigma(\omega)$ of NSMO are shown.

FIG. 2. $T$-dependent $\sigma(\omega)$ of PSMO in the far-infrared region. For clarity, $\sigma(\omega)$ at 290 K are plotted by moving upward by 800 $\Omega^{-1}\text{cm}^{-1}$. In the inset, $\sigma(\omega)$ of NSMO are shown. The dotted lines represent the phonon frequencies at 290 K.

FIG. 3. Midgap state optical conductivity $\sigma_{\text{ms}}(\omega)$ of PSMO. The solid circles, the dotted lines, and the solid lines are represent the experimental data, the Gaussian functions, and the sums of two Gaussian functions, respectively.

FIG. 4. Optical strengths of (a) $S_I$, (b) $S_{II}$, (c) $S_{\text{Drude}}$, and (d) $S_I + S_{\text{Drude}}$. In (d), the dotted line represents the prediction from the $T$-dependent double exchange bandwidth. The deviation of $(S_I + S_{\text{Drude}})$ near $T_N$ can be attributed to the dimensional crossover driven by magnetic ordering.
