Magnetic Field Dependent Optical Studies of a Layered Antiferromagnet

Pr$_{1/2}$Sr$_{1/2}$MnO$_3$

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Magnetic field (H)-dependent optical conductivity spectra $\sigma(\omega)$ and dielectric constant spectra $\varepsilon(\omega)$ of a layered antiferromagnet (A-type) Pr$_{1/2}$Sr$_{1/2}$MnO$_3$ were presented. At 0.0 T in 4.2 K, the $\sigma(\omega)$ showed a small Drude-like peak in the far-infrared region and a broad absorption peak in the mid-infrared region. With increasing $H$, large spectral weights were transferred from high to low energy regions with increase of the Drude-like peak. We found that a polaron picture could explain most of the $H$-dependent spectral changes. By comparing with the $H$-dependent spectral weight changes and $\varepsilon(\omega)$ of Nd$_{1/2}$Sr$_{1/2}$MnO$_3$, which has the CE-type spin ordering, we showed that a dimensional crossover from a 2-dimensional to a 3-dimensional metal occurs in Pr$_{1/2}$Sr$_{1/2}$MnO$_3$.

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Doped manganites with chemical formula $R_{1-x}A_x$MnO$_3$ ($R$ = La, Pr, Nd and $A$ = Ca, Sr, Ba) have attracted much attention due to their exotic electrical and magnetic properties. Basic physics of doped manganites have been explained by the double exchange model based on the strong Hund coupling between itinerant $e_g$ and localized $t_{2g}$ electrons. However, additional degrees of freedom, such as a polaron due to the Jahn-Teller distortion of the Mn$^{3+}$ ion, and an orbital fluctuation have been suggested to explain colossal magnetoresistance and charge/orbital ordering phenomena.

In addition to ferromagnetic metallic ($x \sim 1/3$) and charge ordered insulating ($x \sim 1/2$) ground states, another intriguing ground states were reported in a high doping region ($i.e.$, $x \geq 1/2$). It was proposed that some doped manganites, such as Nd$_{0.45}$Sr$_{0.55}$MnO$_3$ and Pr$_{1/2}$Sr$_{1/2}$MnO$_3$, have metallic ground states where spins in a layer are ferromagnetically ordered but the coupling between the layers is antiferromagnetically ordered. This spin configuration is called the "A-type" antiferromagnetic ordering. The realization of the layered antiferromagnetic metallic ground states was explained by a kinetic energy gain when the $d_{3x^2-r^2}$ orbitals form a 2-dimensional (2D) band. This intriguing ground state was experimentally demonstrated in a single domain Nd$_{0.45}$Sr$_{0.55}$MnO$_3$ crystal by measuring anisotropic electric properties: a metallic conduction within the ferromagnetic layers and an insulating behavior along the antiferromagnetic directions below the Néel temperature.

Under a high magnetic field ($H$), these manganites in the A-type antiferromagnetic ground state experience large resistivity changes with a strong hysteresis.

In spite of these fascinating physical phenomena, optical investigations on the layered antiferromagnetic materials have been rare. Recently, we reported temperature dependent optical conductivity spectra $\sigma(\omega)$ of Pr$_{1/2}$Sr$_{1/2}$MnO$_3$. We demonstrated that the polaron scenario should be better to explain the temperature dependent optical spectra of Pr$_{1/2}$Sr$_{1/2}$MnO$_3$ than the orbital fluctuation scenario. We also suggested that some observed features of $\sigma(\omega)$ could be explained by a dimensional crossover from a 3D to a 2D metal near the Néel temperature. [Note that the direct transport data which demonstrates Pr$_{1/2}$Sr$_{1/2}$MnO$_3$ has a 2D metallic state are still lacking due to the unavailability of a single domain crystal.]

In this paper, we will report $H$-dependent $\sigma(\omega)$ of Pr$_{1/2}$Sr$_{1/2}$MnO$_3$. At 0.0 T in 4.2 K, $\sigma(\omega)$ show a Drude-like peak in the far-infrared region and a broad absorption peak in the mid-infrared region. With increasing $H$, the Drude-like peak increases and the large spectral weight changes are observed below 4.0 eV. These spectral weight changes can be explained within the polaron picture. Moreover, the $H$-dependences of the spectral weights and dielectric constant spectra $\varepsilon(\omega)$ of Pr$_{1/2}$Sr$_{1/2}$MnO$_3$ are different from those of Nd$_{1/2}$Sr$_{1/2}$MnO$_3$, which has the CE-type antiferromagnetic spin ordering. These experimental observations provide further supports for the occurrence of the dimensional crossover in Pr$_{1/2}$Sr$_{1/2}$MnO$_3$.

A Pr$_{1/2}$Sr$_{1/2}$MnO$_3$ single crystal was grown by the floating zone method. Details of sample growth and characterization were reported elsewhere. Its $H$-dependent...
$dc$ resistivity was measured by the conventional four-probe method using a 20 T superconducting magnet. For optical measurements, the crystal was polished up to 0.3 $\mu m$ using diamond paste. To remove surface damages due to the polishing process, we carefully annealed the sample again in an $O_2$ atmosphere at 1000 °C just before optical measurements. The $H$-dependent reflectivity in the energy region between 5.0 meV and 4.0 eV were measured using the spectrophotometers at National High Magnetic Field Laboratory. [All of the optical spectra reported in this paper were measured at 4.2 K.] Using the Kramers-Kronig transformation, $\sigma(\omega)$ and $\varepsilon(\omega)$ were obtained. For the high frequency extrapolations, the room temperature data between 4.0 and 30 eV were used.

The solid lines in Fig. 1 show the $H$-dependent $dc$ resistivity curves of Pr$_{1/2}$Sr$_{1/2}$MnO$_3$, which were taken at $T = 4.2$ K. With increasing $H$, the $dc$ resistivity value ($\sim 10^{-2}$ $\Omega$ cm at 0.0 T) is nearly constant up to 7.0 T, and then it starts to decrease above 7.0 T. The $dc$ resistivity value does not change at all above 11.5 T within our experimental error. With decreasing $H$, the $dc$ resistivity becomes nearly constant down to 9.5 T and then starts to increase. Note that the $dc$ resistivity shows a strong hysteresis.

For comparison, the $H$-dependent $dc$ resistivity curves of Nd$_{1/2}$Sr$_{1/2}$MnO$_3$ are also shown as dotted lines in Fig. 1. Compared to the Pr$_{1/2}$Sr$_{1/2}$MnO$_3$ case, the $H$-dependent $dc$ resistivity change of Nd$_{1/2}$Sr$_{1/2}$MnO$_3$ is quite large. With increasing $H$, the high resistivity value ($\sim 10^2$ $\Omega$ cm at 0.0 T) is sharply decreased near 13.0 T and also shows a very strong hysteresis. Note that the $dc$ resistivity value at 0.0 T in Nd$_{1/2}$Sr$_{1/2}$MnO$_3$ in the $H$-decreasing run is quite smaller than that in the $H$-increasing run.

In Fig. 2(a), we show the $H$-dependent $\sigma(\omega)$ of Pr$_{1/2}$Sr$_{1/2}$MnO$_3$ during the $H$-increasing run. At 0.0 T, there are broad peaks around 1.0 and 4.0 eV. As $H$ increases, the spectral weights near 1.0 eV and 3.0 eV are transferred to a lower energy region. The gap-like feature in $\sigma(\omega)$ at 0.0 T changes into a Drude-like peak above 10.0 T. The low frequency details of transferred spectral weights below 0.1 eV can be seen in Fig. 2(b). At 0.0 T, there are sharp peaks due to the optic phonon modes and a hint of small rise in $\sigma(\omega)$ in the low frequency limit, suggesting the existence of the small Drude-like peak. As $H$ increases, the phonon peaks become screened and the Drude-like peak becomes more clear below 0.04 eV. The solid circle represents the $dc$ conductivity value at 14.0 T. Therefore, $\sigma(\omega)$ below 0.1 eV at the high $H$ should be viewed as two parts, i.e., the Drude and the large incoherent absorption peaks.

In our earlier studies on the temperature dependent $\sigma(\omega)$ of Pr$_{1/2}$Sr$_{1/2}$MnO$_3$, we observed similar spectral weight changes. In the high frequency region above 2.0 eV, there are two contributions. Around 4.0 eV, there is a peak due to the charge transfer transition from the O 2p level to the Mn 3d level. And, there is a broad peak due to the optical transition between the Hund’s rule split bands near 3.0 eV, however it is much weaker than the charge transfer transition. And, it was also shown that the polaron scenario was better to describe the optical spectra in the low frequency region than the
orbital fluctuation scenario.

After subtracting the charge transfer transition contribution from the measured $\sigma(\omega)$, we analyzed $H$-dependent $\sigma(\omega)$ below 2.0 eV within the polaron picture:

$$\sigma(\omega < 2.0 \text{ eV}) = \sigma_D(\omega) + \sigma_I(\omega) + \sigma_{II}(\omega),$$

where $\sigma_D(\omega)$, $\sigma_I(\omega)$, and $\sigma_{II}(\omega)$ represent the contributions due to free carriers, incoherent polaron absorption, and the interorbital transition between the Jahn-Teller split levels, respectively. The simple Drude model was used for $\sigma_D(\omega)$, and two Gaussian functions were used for $\sigma_I(\omega)$ and $\sigma_{II}(\omega)$. Then, we estimated the $H$-dependent optical strengths $S_D$, $S_I$, and $S_{II}$ by integrating $\sigma_D(\omega)$, $\sigma_I(\omega)$, and $\sigma_{II}(\omega)$, respectively. Details of this procedure were published elsewhere.

Although there are no direct experimental evidences such as anisotropic transport measurements on a single domain crystal, $\text{Pr}_{1/2}\text{Sr}_{1/2}\text{MnO}_3$ is believed to have a 2D metallic behavior in the $A$-type spin ordered states. On the other hand, the charge/orbital ordered state of $\text{Nd}_{1/2}\text{Sr}_{1/2}\text{MnO}_3$ with the CE-type ordering should be insulating. The weaker $H$-dependences of $S_I$ and $S_D$ for $\text{Pr}_{1/2}\text{Sr}_{1/2}\text{MnO}_3$ can be easily understood. Moreover, $\sigma(\omega)$ of $\text{Pr}_{1/2}\text{Sr}_{1/2}\text{MnO}_3$, shown in Fig. 2(b), show a weak Drude-like peak even at $H = 0.0 \text{ T}$. To get more informations on the ground state, we estimated the value of the total polaron absorption $S_{tot} (= S_I + S_D)$. It is found that the value of $S_{tot}$ at 0.0 T is by about 65 \% ($\sim 2/3$) smaller than that at 14.0 T. The existence of the Drude-like peak and the decrease of $S_{tot}$ by an amount of $\sim 1/3$ at zero field support that the ground state of $\text{Pr}_{1/2}\text{Sr}_{1/2}\text{MnO}_3$ should be a 2D metal.

**FIG. 3.** Optical strengths (a) $S_I$, (b) $S_{II}$, and (c) $S_D$ for $\text{Pr}_{1/2}\text{Sr}_{1/2}\text{MnO}_3$ (solid circles) and $\text{Nd}_{1/2}\text{Sr}_{1/2}\text{MnO}_3$ (solid squares). [All the units are $\Omega^{-1}\text{cm}^{-1}\text{eV}$.]

Figures 3(a), (b), and (c) show the $H$-dependence of $S_I$, $S_{II}$, and $S_D$ for $\text{Pr}_{1/2}\text{Sr}_{1/2}\text{MnO}_3$ (solid circles) during the $H$-increasing run, respectively. [For comparison, $\text{Nd}_{1/2}\text{Sr}_{1/2}\text{MnO}_3$ data were also shown as solid squares.] With increasing $H$, $S_I$ and $S_D$ increase smoothly, but $S_{II}$ shows the opposite behavior. The $H$-dependences of $S_I$, $S_{II}$, and $S_D$ for $\text{Nd}_{1/2}\text{Sr}_{1/2}\text{MnO}_3$ are similar to those for $\text{Pr}_{1/2}\text{Sr}_{1/2}\text{MnO}_3$. And, at high $H$, the values of $S_I$ and $S_D$ are nearly the same for both samples. However, there are a few important differences between $\text{Pr}_{1/2}\text{Sr}_{1/2}\text{MnO}_3$ and $\text{Nd}_{1/2}\text{Sr}_{1/2}\text{MnO}_3$ data. First, at $H = 0.0 \text{ T}$, $S$ of $\text{Pr}_{1/2}\text{Sr}_{1/2}\text{MnO}_3$ remains as a finite value, while that of $\text{Nd}_{1/2}\text{Sr}_{1/2}\text{MnO}_3$ becomes nearly zero. Second, the $H$-dependences of $S_I$ and $S_D$ are much weaker for $\text{Pr}_{1/2}\text{Sr}_{1/2}\text{MnO}_3$. These $H$-dependences agree with Fig. 1, where the resistivity of $\text{Pr}_{1/2}\text{Sr}_{1/2}\text{MnO}_3$ is smaller than that of $\text{Nd}_{1/2}\text{Sr}_{1/2}\text{MnO}_3$ by four orders of magnitude at zero field but becomes nearly the same at high $H$.

**FIG. 4.** $H$-dependent $\varepsilon(\omega)$ of (a) $\text{Pr}_{1/2}\text{Sr}_{1/2}\text{MnO}_3$ and (b) $\text{Nd}_{1/2}\text{Sr}_{1/2}\text{MnO}_3$.

Further insights on $\text{Pr}_{1/2}\text{Sr}_{1/2}\text{MnO}_3$ can be obtained from the $H$-dependent $\varepsilon(\omega)$, shown in Fig. 4(a). At 0.0 T, $\varepsilon$ is nearly $\omega$-independent, but the value of $\varepsilon$ at the low frequency is less than zero. This result implies that $\text{Pr}_{1/2}\text{Sr}_{1/2}\text{MnO}_3$ should be a metal at 0.0 T, which is consistent with the existence of the Drude-like peak in Fig. 2(b). With increasing $H$, $\varepsilon(\omega)$ decrease further and clearly show a metallic behavior at 14.0 T. As a comparison, we also plot the $H$-dependent $\varepsilon(\omega)$ for $\text{Nd}_{1/2}\text{Sr}_{1/2}\text{MnO}_3$ in Fig. 4(b). At 0.0 T, $\varepsilon$ is positive and nearly $\omega$-independent at all frequencies, consistent with a typical insulator response. With increasing $H$ up to 13.0 T, $\varepsilon(\omega)$ in the low frequency region increases. With increasing $H$ further, $\varepsilon(\omega)$ suddenly become negative and finally show a typical metallic response at 17.0 T.

In our earlier study for the $H$-dependent $\varepsilon(\omega)$ of $\text{Nd}_{1/2}\text{Sr}_{1/2}\text{MnO}_3$, we demonstrated that the diver-
gence behavior of \( \varepsilon(\omega) \) near the insulator-metal transition boundary, i.e., 13.0 T, could be explained by a dielectric anomaly near a percolation threshold. However, in \( \text{Pr}_{1/2}\text{Sr}_{1/2}\text{MnO}_3 \), such a dielectric anomaly of \( \varepsilon(\omega) \) cannot be observed near the 10.0 T region, where \( dc \) resistivity experiences a large change.

The \( dc \) resistivity change near the 10.0 T region can be understood in terms of a dimensional crossover from 2D to 3D metals. If the zero field ground state of \( \text{Pr}_{1/2}\text{Sr}_{1/2}\text{MnO}_3 \) is a 2D metal, the sample might be composed of multi-magnetic domains with their antiferromagnetic directions randomly pointing along \( x, y, \) and \( z \) directions. For such sample, the percolating metallic paths should be formed. That is the reason why we could observe the existence of the Drude-like peak and the absence of the dielectric anomaly in \( \text{Pr}_{1/2}\text{Sr}_{1/2}\text{MnO}_3 \). In optical measurements, the light will probe the average response of the multi-magnetic domains. Effectively, \( 2/3 \) of its response comes from the metallic response within the ferrimagnetically ordered layer and \( 1/3 \) comes from the insulating response along the antiferromagnetic direction. Since the metallic response dominates in the low frequency region, the value of \( S_{tot} \) could be reduced approximately by \( 1/3 \) near the dimensional crossover, in agreement with our experimental observation. The detailed process of the dimensional crossover is not known yet. Using \( ^{55}\text{Mn} \) NMR experiment, Allodi et al. suggested the percolative origin for the ferromagnetic transition in \( \text{Pr}_{1/2}\text{Sr}_{1/2}\text{MnO}_3 \). Compared with the \( \text{Nd}_{1/2}\text{Sr}_{1/2}\text{MnO}_3 \) case, such a percolative process is quite plausible. However, more studies are required to understand details of the dimensional crossover.

In summary, we investigated the magnetic field dependent optical conductivity spectra and dielectric constant spectra of an \( A \)-type spin ordered half-doped manganite \( \text{Pr}_{1/2}\text{Sr}_{1/2}\text{MnO}_3 \). In comparison with \( \text{Nd}_{1/2}\text{Sr}_{1/2}\text{MnO}_3 \), which show the \( CE \)-type spin ordering, we showed that a dimensional crossover from a 2-dimensional to a 3-dimensional metal should occur in \( \text{Pr}_{1/2}\text{Sr}_{1/2}\text{MnO}_3 \).

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