Optical spectra measured on cleaved surfaces of double-exchange ferromagnet La$_{1-x}$Sr$_x$MnO$_3$

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Abstract

Optical reflectivity spectra were measured on cleaved surfaces of La$_{1-x}$Sr$_x$MnO$_3$ single crystals ($x=0.175$, $T_C=283$ K) over a temperature range 10–295 K. The optical conductivity $\sigma(\omega)$ shows, keeping single-component nature, incoherent-to-coherent crossover with increase of electrical conductivity. The $\sigma(\omega)$ spectrum of low-temperature ferromagnetic-metallic phase (10 K) exhibits a pronounced Drude-like component with large spectral weight, contrary to the previous result. The present result indicates that the optical spectrum of the manganites is sensitive to condition of sample surfaces.

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Optical reflectivity studies are important from both fundamental and practical viewpoints, since it enables us not only to deduce the dielectric function, but also to examine separately two key-elements of the charge transport, \textit{i.e.}, the carrier density (or Drude weight) and the scattering time. The charge transport is one of the central concerns for both camps. However, for the case of the double-exchange ferromagnetic-metal manganites, which have recently attracted renewed interest because of its intriguing phenomenon, colossal magnetoresistance (CMR) \cite{1}, the previous results of the reflectivity studies are rather confused and controversial \cite{2,3,4}. We report the optical reflectivity spectra \( R(\omega) \) of \( La_{1-x}Sr_xMnO_3 \) \((x=0.175, T_C=283 \text{ K})\) measured on cleaved surfaces of single crystals over a wide temperature range \((10–295 \text{ K})\). The optical conductivity \( \sigma(\omega) \) exhibits a single-component with large spectral weight, contrary to the previous results. The present result indicates that the optical spectrum of the manganites is very sensitive to condition of surfaces, which can partly explain the above confusion \cite{5}.

Single crystals of \( La_{1-x}Sr_xMnO_3 \) were grown by a floating zone method \cite{6}. The size of the cleaved surface was at largest \( 1.0 \times 1.0 \text{ mm}^2 \). Temperature-dependent optical reflectivity was measured using a Fourier-type interferometer \((0.02–1.6 \text{ eV})\) and a grating spectrometer \((0.8–6.6 \text{ eV})\). The experimental error of reflectivity, \( \Delta R \), determined by the reproducibility, is less than 1\% for the far-IR to visible region and less than 2\% for the ultraviolet region.

Figure 1 shows the temperature-dependent \((10–295 \text{ K})\) reflectivity spectra measured on the cleaved surfaces of \( La_{0.825}Sr_{0.175}MnO_3 \) single crystals on a logarithmic energy scale in the range \( 0.01–6.0 \text{ eV} \). The Curie temperature \( T_C \) measured by dc resistivity was 283 K. With decreasing temperature, the reflectivity spectrum \( R(\omega) \) changes gradually from insulating to metallic behavior: the reflectivity edge at about \( 1.6 \text{ eV} \) becomes sharpened, though its position does not shift appreciably, and the optical phonons are screened corresponding to increase of electrical conductivity. Below 100 K, the optical phonons almost fade away and the spectrum is characterized by a sharp edge and a large spectral weight below it. The present spectrum measured on cleaved surfaces is much higher at a mid-IR-to-visible region compared with the
previous results measured on polished surfaces [2, 3].

In order to make more detailed discussions, we deduce optical conductivity \( \sigma(\omega) \) (Fig. 2) from \( R(\omega) \) shown in Fig. 1 via a Kramers-Kronig transformation. We measured reflectivity spectra at each temperature below 6.6 eV and above 6.6 eV we assumed the data measured at room temperature (295 K) using a Seye-Namioka type spectrometer for vacuum-ultraviolet (VUV) synchrotron radiation up to 40 eV at Institute for Molecular Science, Okazaki National Research Institutes. Such a procedure is possible and reasonable because the variation of \( R(\omega) \) at 6.0–6.6 eV is negligible small and the data below 6.6 eV could be connected smoothly with the VUV-data. For the extrapolation at the lower-energy part, we assumed a constant \( R(\omega) \) for the insulating phase \((T=295 \text{ K})\). For the metallic phase \((T \leq 278 \text{ K})\), we make a smooth extrapolation using a Hagen-Rubens formula. Extrapolating parameter \( \sigma(0) \) is roughly in accord with the dc value [4]. Variation of the extrapolation procedures had negligible effect on \( \sigma(\omega) \) in the energy region of interest \((0.03–6.0 \text{ eV})\).

The optical conductivity \( \sigma(\omega) \) shows, keeping single-component nature, incoherent-to-coherent crossover with decreasing temperature: Above \( T_C \), the \( \sigma(\omega) \) spectrum is characterized solely by a broad peak centered at \( \sim 1.5 \text{ eV} \). At the temperature range \( 278–220 \text{ K} \), this broad peak gradually develops and its position shifts downwards as \( T \) decreases but a Drude-like component is not confirmed in the present experiment, though the material is ferromagnetic-metallic. Below 155 K, on the other hand, the spectrum exhibits a single Drude-like component centered at \( \omega=0 \) and it becomes narrow as \( T \) decreases without increase of spectral weight.

Integrated spectral weight defined as

\[
N^\ast_{\text{eff}}(\omega) = \frac{2m_0V}{\pi e^2} \int_0^\omega \sigma(\omega') d\omega'
\]

\((m_0: \text{a bare electron mass; } V: \text{the unit-cell volume})\) represents an effective density of carriers contributing to optical transitions below a certain cutoff energy \( \hbar\omega \) (inset of Fig. 2). The characteristic single-component which shows incoherent-to-coherent crossover consists of the spectral weight transferred from the two bands at \( \sim 3 \text{ eV} \) and at \( \sim 5 \text{ eV} \). The present result suggests that the exchange-split down-spin band [6] consists of two
bands. Because the curves of $N_{\text{eff}}^*(\omega)$ merge into almost a single line above 6 eV, the down-spin band does not seem to split to more than two bands. Imperfect convergence is most likely due to the increasing experimental error on $\sigma(\omega)$ with $\omega$. This partly justifies our procedure that the data above 6.6 eV at room temperature is connected with the data measured at each temperature.

Finally we show that the discrepancy between our result and the previous result may originate from sensitivity of the spectrum to the condition of the surface. In Fig. 3 are shown the room-temperature (295 K) reflectivity spectra measured on a cleaved surface (solid line) as well as that measured on a surface polished by lapping films with diamond powder (dotted line). It is found that polishing dramatically alters $R(\omega)$ for the ferromagnetic metal La$_{0.70}$Sr$_{0.30}$MnO$_3$ [Fig. 3(b)] whereas it affects only slightly the spectrum for the undoped LaMnO$_3$ [Fig. 3(a)]. The previous data [2, 3] resembles closely the spectrum measured on the polished surface. The damage of the surface probably localizes the carriers. However, light with long wavelength reaches inside the bulk and hence $R(\omega)$ recovers the intrinsic spectrum, which is consistent with the observation that the discrepancy almost disappears below 0.03 eV [4]. “Small Drude weight” may originate from the above restoration process [inset of Fig. 3(b)].

In summary, we have reported the temperature-dependent optical spectra of the prototypical double-exchange system La$_{1-x}$Sr$_x$MnO$_3$ measured on the cleaved surfaces. The optical conductivity spectra are characterized by a single component which shows incoherent-to-coherent crossover with increase of the electrical conductivity and a pronounced Drude-like component is observed for the ferromagnetic-metallic phase at low temperatures. The present result indicates that the optical spectrum of the doped manganites is sensitive to condition of surfaces. The charge dynamics of the doped manganites might be extremely sensitive to static imperfections.

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Figure Captions

Fig. 1 Temperature-dependent optical reflectivity spectra measured on cleaved surfaces of \( \text{La}_{0.825}\text{Sr}_{0.175}\text{MnO}_3 \) \( (T_C = 283 \text{ K}) \).

Fig. 2 Temperature-dependent optical conductivity spectra of \( \text{La}_{0.825}\text{Sr}_{0.175}\text{MnO}_3 \) deduced from the reflectivity spectra measured on the cleaved surfaces (shown in Fig. 1) via a Kramers-Kronig transformation. Inset: Effective carrier number per Mn-atom \( N^*_{\text{eff}}(\omega) \) defined as the integration of \( \sigma(\omega) \).

Fig. 3 Room-temperature (295 K) optical reflectivity spectra measured on the cleaved (solid line) and polished (dotted line) surfaces: (a) \( \text{LaMnO}_3 \) and (b) \( \text{La}_{0.70}\text{Sr}_{0.30}\text{MnO}_3 \). Inset: Optical conductivity spectra deduced from the Kramers-Kronig transformation of the reflectivity spectra shown in Fig. 3(b).