Observation of Temperature Induced Plasma Frequency Shift in an Extremely Large Magnetoresistance Compound LaSb *

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(Received 19 April 2017)

We report an optical spectroscopy study on LaSb, a compound recently identified to exhibit extremely large magnetoresistance. Our optical measurement indicates that the material has a low carrier density. More interestingly, the study reveals that the plasma frequency increases with decreasing temperature. This phenomenon suggests either an increase of the conducting carrier density or/and a decrease of the effective mass of carriers with decreasing temperature. We attribute it primarily to the latter effect. Two possible scenarios on its physical origin are examined and discussed. The study offers new insight into the electronic structure of this compound.

PACS: 78.20.−e DOI: 10.1088/0256-307X/34/7/077804

Recently, the extremely large magnetoresistance (XMR) phenomenon discovered in a few semimetals such as WTe_2,[1] NbP,[2] Cd_3As_2,[3] TaAs,[4] $TmPn_2$ (Tm=Ta/Nb, Pn=As/Sb)[5–9] and LnX (Ln=La/Y, X=Sb/Bi)[10–12] has attracted a great deal of attention in the condensed matter community not only due to the high interest of elucidating the underlying physical origin but also because of its potential applications in magnetic sensors, switches and memories. Several mechanisms have been proposed to explain the XMR behavior, including electron-hole compensation from a semiclassical two-band model,[1,6,13] suppression of backscattering channels at zero field,[14] a quantum viewpoint by incorporating Landau levels,[15,16] field-induced Fermi surface (FS) changes[5] and nontrivial band topology.[10] Nevertheless, a consensus has not been achieved.

Among those materials, the binary LaSb is one of the simplest compounds. It has a rock salt structure with relatively simple band structure. There are two hole pockets, one spherical and one intersecting-ellipsoidal shape, around the Brillouin zone (BZ) center Γ point and one ellipsoidal electron pocket at the BZ boundary X.[17] From the material perspective, the compound has the advantage of facilitating the investigation on the origin of XMR. Many works have been carried out on this material.[10,16–19] This compound is found to have low carrier density of the order of $10^{20}\,\mathrm{cm}^{-3}$, but ultrahigh mobility. The carrier mobilities are 10^4–$10^5\,\mathrm{cm}^2\mathrm{V}^{-1}\mathrm{s}^{-1}$ derived from low-temperature transport experiments.[10,17] From the temperature-dependent Hall coefficient data, the charge carriers change from electron-type at 300 K to hole-type at a slightly lower temperature and eventually to electron-type at $T < 40\,\mathrm{K}$.[10] The electron and hole carrier density calculated from the enclosed FS volumes is perfectly compensated for, being consistent with the results of quantum oscillation (QO) and angular resolved photoemission spectroscopy (ARPES) measurements.[17–19] More interestingly, the ARPES measurement revealed that LaSb has a linearly dispersive bulk band and shares many similarities with the Weyl semimetal TaAs family which consists of hole FSs from the normal parabolic bands and electron FSs from the linear Weyl bands in the bulk electronic structure.[19] A recent ARPES measurement even revealed a nontrivial surface state.[18] Furthermore, the carrier compensation is unaffected by temperature as the FSs show no changes with temperature based on ARPES results.

Infrared optical spectroscopy can provide much useful information about charge dynamics, carrier density, and electronic structure of a material. In this work, we study the infrared response of the newly discovered extremely large magnetoresistance material LaSb. The study reveals a low energy scale of the screened plasma edge near $3000\,\mathrm{cm}^{-1}$. It yields optical evidence for a low-carrier density semimetal, which is consistent with the results obtained from density function theory calculations and ARPES measurements. More interestingly, we observe a temperature induced shift of plasma frequency: the plasma frequency increases with decreasing temperature. This phenomenon suggests either an increase of the conducting carrier density or/and a decrease of the effective mass of carriers with decreasing temperature.

*Supported by the National Natural Science Foundation of China under Grant Nos 11327806 and GZ1123, and the National Key Research and Development Program of China under Grant No 2016YFA0300902.

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We elaborate that the peculiar phenomenon is mainly caused by the latter effect.

The LaSb single crystals were grown by a self-flux method similar to the procedure described in Ref. [18]. The optical reflectance measurements were performed on as-grown shinny surfaces of the single crystals with a combination of Bruker 113v and Vertex 80v in the frequency range of 40–20000 cm^{-1}. An *in-situ* gold and aluminum over-coating technique was used to obtain the reflectance.[20] Then the measured reflectance was corrected by multiplying the available curves of gold and aluminum reflectivity at different temperatures. The dielectric function was derived from the standard Kramers–Kronig transformation.

Figures 1(a) and 1(b) show the reflectance spectra $R(\omega)$ of LaSb in different frequency ranges at selected temperatures. As can be seen, $R(\omega)$ shows metallic frequency and temperature dependence, and $R(\omega)$ at low energy increases rapidly with decreasing frequency and approaches unity at the zero frequency limit at all temperatures. In addition, $R(\omega)$ at low energy increases with decreasing temperature. Both of them are consistent with the metallic behavior in the resistivity measurement.[10,18,19]

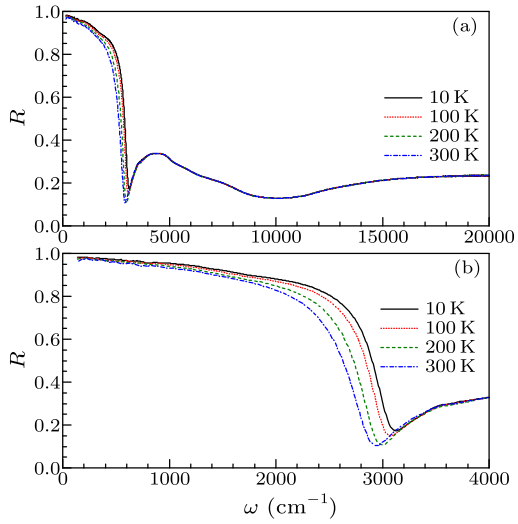

Fig. 1. (a) The temperature-dependent $R(\omega)$ in the frequency range from 0 to 20000 cm^{-1}. (b) The expanded plot of $R(\omega)$ from 0 to 4000 cm^{-1}.

With increasing frequency, $R(\omega)$ drops quickly to a minimum value near 3000 cm^{-1}, being usually referred to as the plasma edge.[21–25] The energy of the plasma edge corresponds to the 'screened' plasma frequency ω_{P}^{*}, which is linked to the plasma frequency by the relation $\omega_{\mathrm{P}}^{*} = \omega_{\mathrm{P}}/\sqrt{\varepsilon_{\infty}}$. The plasma frequency satisfies the equation $\omega_{\mathrm{P}}^{2} = 4\pi ne^{2}/m^{*}$, where n is the carrier density, m^{*} is the effective mass, and ε_{∞} is the dielectric constant at high frequency. For ordinary metals, the plasma edges appear at a rather high energy scale, usually over 10000 cm^{-1}. As a comparison, the energy scale of the plasma edge of LaSb is considerably lower. The low energy scale of the plasma edge reveals a relatively small value of carrier density divided by the effective mass, namely n/m^{*}. Since the effective mass of charge carriers is not expected to be heavy, the carrier density of the compound must be small.

With decreasing temperature, the plasma edge sharpens slightly, indicating a reduced scattering for charge carriers. Most interestingly, the plasma edge displays a clear shift towards higher energy (i.e., a blueshift). Above the edge frequency, the reflectance becomes roughly temperature independent. Several peak-like features in the $R(\omega)$ spectra can be ascribed to interband transitions.

The formation of the plasma edge and its evolution with temperature can be clearly seen in the real part of the dielectric function $\epsilon_1(\omega)$, as shown in Fig. 2. In $\epsilon_1(\omega)$, the zero-crossing frequency represents the 'screened' plasma frequency ω_{P}^{*}. With decreasing temperature, the screened plasma frequency increases as listed in Table 1. The data are in good agreement with the shift of plasma edge as seen in reflectance spectra.

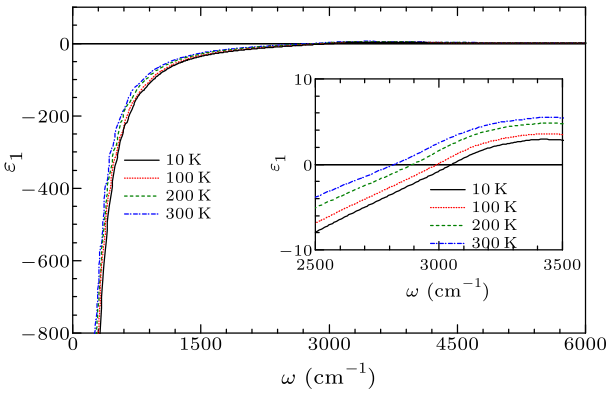

Fig. 2. The real part of the dielectric functions $\epsilon_1(\omega)$ of LaSb in the frequency range of 0–6000 cm^{-1}. Inset: the expanded plot of $\epsilon_1(\omega)$ from 2500 to 3500 cm^{-1}.

To quantitatively characterize the variation of the carrier density and its damping, we tried to reproduce the reflectance spectra $R(\omega)$ from a simple Drude–Lorentz model.[26] The overall dielectric function can be expressed as

$$\epsilon(\omega) = \epsilon_{\infty} - \sum_{i} \frac{\omega_{\mathrm{p}i}^{2}}{\omega^{2} + i\omega\Gamma_{\mathrm{D}i}} + \sum_{j} \frac{S_{j}^{2}}{\omega_{j}^{2} - \omega^{2} - i\omega\Gamma_{j}}, \tag{1}$$

where ε_{∞} is the dielectric constant at high energy, $\omega_{\mathrm{p}i}$ and $\Gamma_{\mathrm{D}i}$ are the plasma frequency and the relaxation rate of each conduction band, while ω_j, Γ_j and S_j are the resonance frequency, the damping and the mode strength of each Lorentz oscillator, respectively. We apply a Drude term and three Lorentz terms to reproduce the reflectance spectra below 20000 cm^{-1} at all temperatures. However, as $R(\omega)$ drops so sharply, we cannot give a perfect fit with a single simple Drude

component. The calculated curve deviates from the experimental spectrum, nevertheless it reflects the evolution of the plasma frequencies $\omega_{\rm p}$ and scattering rate Γ with a change of temperature. Figure 3 illustrates the reflectance spectra at 10 and 300 K together with the Drude–Lorentz fitting results for LaSb. The plasma frequencies $\omega_{\rm p}$ and scattering rate Γ are also listed in Table 1. We can see that the plasma frequency increases from 8000 to 8650 $\rm cm^{-1}$ with decreasing temperature, in the meantime, the scattering rate decreases from 220 to 170 $\rm cm^{-1}$. As already mentioned above, the blue shift of the plasma frequency indicates either an increase of carrier density or/and a decrease of the effective mass of the conducting charge carriers with decreasing temperature.

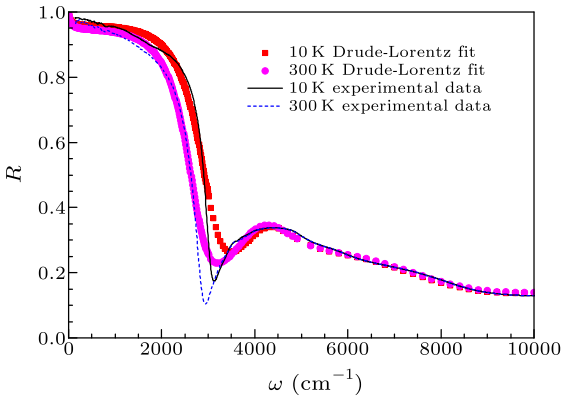

Fig. 3. The experimental reflectance spectra $R(\omega)$ along with the decomposed Drude and Lorentz components of LaSb at 10 K and 300 K.

Table 1. The screened plasma frequency $\omega_{\rm P}^{*}$ of LaSb obtained from the zero-crossing frequency of the dielectric function $\epsilon_1(\omega)$, the plasma frequency and the scattering rate obtained from the Drude–Lorentz fit. The unit of these quantities is $\rm cm^{-1}$.

T (K)	300 K	200 K	100 K	10 K
$\omega_{\rm P}^{*}$	2820	2890	2990	3050
$\omega_{\rm P}$	8000	8200	8500	8650
Γ	220	210	190	170

It is well known that, for an ordinary metal or semimetal, the carrier density is not a temperature-dependent quantity, instead it is determined by its crystal structure. The charge carrier number could be changed by thermal activation only for a semiconductor with a small band gap or a metal with very small Fermi energy.[27] However, the number of such thermally activated carriers can only decrease with decreasing temperature, which is opposite to the trend observed here. According to the ARPES measurement on LaSb, no visible temperature induced changes of the FSs were indicated,[19] therefore LaSb should belong to a system with constant carrier density.

Since the carrier density is temperature independent, we are left with the possibility that the effective mass of charge carriers decreases with decreasing temperature. It is not unusual that the carrier effective mass changes with temperature for correlated electronic systems (e.g., heavy fermion materials), but in those systems the effective mass always increases with decreasing temperature due to the many-body renormalization effect.[28,29] It is rare to find systems in which the carrier effective mass decreases with decreasing temperature. The phenomenon was observed in a few systems with low carrier density, for example, $Cu_{0.07}TiSe_2$,[23] $Bi_2Te_{3-\delta}Sn_\delta$,[25] Cu-intercalated Bi_2Se_3,[25] and recently in $SrMnSb_2$.[30] LaSb offers a new example showing this peculiar property. We shall address this issue and examine possible explanations for its origin in the following.

In our earlier study on $Cu_{0.07}TiSe_2$,[23] we already pointed out that for a system with constant but low carrier density, the chemical potential is not fixed but shifts up with decreasing temperature. Assuming a degenerate electron gas for a simple metal, the chemical potential has the following relation with temperature, $\mu \simeq \mu_0[1-\pi^2/12(T/T_{\rm F})^2]$. For an ordinary metal, the Fermi energy $E_{\rm F}$ is very high, thus the T-induced change in μ in the temperature range of 0–300 K is negligible. However, if the Fermi energy is rather low, about 80–100 meV at 300 K then we expect to have an upward shift of chemical potential by about 5%–10% for T decreasing from 300 to 0 K. If the effective mass of electrons along the dispersive band is different, a change of effective mass at $E_{\rm F}$ could be caused by a chemical potential shift. From the ARPES measurement of LaSb,[19] the compound is a compensated semimetal with small $E_{\rm F}$. The electron band at the Brillouin zone (BZ) boundary X is parabolic along the long-axis but linearly along the short axis, while the outer hole band around the BZ center Γ point disperses not parabolically but gradually levels off from Γ–X, in other words, the slope of the energy band may increase with the upward shift of chemical potential. It is known that the band effective mass is defined as $1/m^{*} = \frac{1}{\hbar}\frac{d^2\varepsilon}{dk^2}$, thus the increase of the slope reflects the decrease of the band effective mass, which in turn results in the increase of $\omega_{\rm P}^{*}$ and $\omega_{\rm P}$. Therefore, the presence of such a peculiar nonparabolic band in a low carrier density compound may explain the blue shift of the plasma frequency with decreasing temperature.

Recently, Park *et al.* proposed a 'three-band' model, which includes two conduction bands and one valence band, to explain the observed blue shift of the plasma edge in $SrMnSb_2$.[30] The first conduction band is a conventional conduction band which crosses the Fermi level. The second conduction band is assumed to be close but above the Fermi level, moreover it is heavier or lighter than the first conduction band. Provided that $k_{\rm B}T$ is comparable with the band separation, a thermal redistribution of carriers between these two conduction bands should occur. As

the temperature increases, the number of thermally redistributed quasiparticles between the two conduction bands increases, leading to a change of the optical effective mass and, as a consequence, a change of the plasma frequency. According to this model, when the second conduction band is heavier (lighter), the plasma frequency would decrease (increase) with increasing temperature. Assuming that the LaSb compound indeed has another heavier electron band above but very close to the Fermi level, the model can also explain the observation of the blue shift of the plasma edge with decreasing temperature. So far, it is still unclear whether or not such a heavier band is present in its band structure. The present work may motivate further detailed studies on its band structure.

In summary, we have studied the optical response of a newly discovered extremely large magnetoresistance material LaSb. The study reveals a well formed plasma edge near 3000 cm^{-1}, indicating that the compound has rather low-carrier density. The observation is consistent with the results obtained from density function theory calculations and ARPES measurements. More significantly, we observe clearly a blue shift of plasma frequency with decreasing temperature. This phenomenon suggests either an increase of the conducting carrier density or/and a decrease of the effective mass of carriers with decreasing temperature. We attribute it primarily to the latter effect. We elaborate that the decrease of the effective mass of carriers in such low carrier density systems can be explained by the presence of either peculiar nonparabolic bands crossing E_{F} or a heavy electron band above but close to E_{F}. The study offers new insight into the electronic structure of the compound.

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