Normal-state charge dynamics of ternary platinum germanide superconductor La₂Pt₃Ge₅

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Abstract

We report on the infrared spectroscopic studies of the normal-state electronic response of rare-earth ternary platinum germanide superconductor $La_2Pt_3Ge_5$. We analyzed the temperature-dependent optical conductivity spectra using the Drude-Lorentz oscillator model. We found that the two Drude responses with distinct scattering rates are required to explain the charge dynamics at 10 K while a single Drude mode could reproduce the far-infrared conductivity at higher temperatures. Our results indicated the two-band character of the electronic structure and highlighted the disparate temperature evolution of the electrodynamics of the two electronic states.

Keywords: La₂Pt₃Ge₅, multiband, infrared spectroscopy, optical conductivity

1. INTRODUCTION

Rare-earth ternary intermetallic compounds $R_2M_3X_5$ (R =rare-earth element, M = transition metal, X = s-p metal) have been attracting attention due to their intriguing magnetic and superconducting properties. Among the intermetallic compounds, the ternary iron silicide R_2 Fe₃Si₅ has been under intensive studies due to the fact that the highest superconducting transition temperature among any known iron compounds was observed in Lu₂Fe₃Si₅ with T_c =6.1 K [1]. Low-temperature heat capacity data of Lu₂Fe₃Si₅ revealed anomalous properties of the superconducting state [2]. Lu₂Fe₃Si₅ showed a large linear term in the heat capacity in the superconducting state and a reduced normalized jump in the heat capacity at T_c smaller than the BCS value of 1.43 [2]. These unusual phenomena were explained in terms of the two-band model in which one band remains normal in the superconducting states [2]. A recent heat-capacity study of high-quality Lu₂Fe₃Si₅ single crystals found that the specific heat divided by temperature showed a sudden drop at about $T_c/5$ and became zero with further decreasing temperature [3]. These results indicated the presence of the two superconducting gaps. Hall measurements and band structure calculations further supported the multiband nature of the electronic structure of Lu₂Fe₃Si₅ [3]. Penetration depth and the thermal conductivity data also indicated the two-gap superconductivity of Lu₂Fe₃Si₅[4, 5].

Among the rare-earth ternary intermetallic superconductors, a platinum germanide $La_2Pt_3Ge_5$ has the highest T_c of 8.1 K [6]. The specific heat data of $La_2Pt_3Ge_5$ showed unusual behavior below T_c , which is similar to the case of $Lu_2Fe_3Si_5$ [6]. The

specific heat jump at T_c was found to be about 0.36, which is much smaller than the BCS value. The electronic specific heat divided by temperature showed almost linear temperature dependence below T_c . These results suggest a possible multigap superconductivity of La₂Pt₃Ge₅.

In order to identify the multiband nature of the electronic states, spectroscopic studies are required. Infrared spectroscopy is well suited to investigate the electronic structure of multiband systems. Indeed extensive infrared spectroscopic studies have revealed the multiband character of exotic superconductors including MgB₂ [7, 8] and the Fe-based compounds [9-14].

In this paper, we report on the normal-state charge dynamics of $La_2Pt_3Ge_5$ single crystals investigated by using infrared spectroscopy. We analyzed the temperature evolution of the optical conductivity $\sigma_1(\omega)$ spectra. The far-infrared conductivity is dominated by strong Drude response, indicating the metallic character of $La_2Pt_3Ge_5$. The far-infrared conductivity at 295 K, 200 K, and 100 K can be described by a single Drude mode. In contrast, we found that the far-infrared conductivity at 10 K should be described by two Drude modes with distinct values of scattering rates. Our results demonstrate the presence of the two electronic bands governing the low-energy charge dynamics and the disparate evolution of the two bands with temperature in $La_2Pt_3Ge_5$.

2. EXPERIMENTAL DETAILS

The single crystals of La₂Pt₃Ge₅ were provided by Bank for Quantum Electronic Materials. The single crystals were grown by the high-temperature flux method using a Pt-Ge

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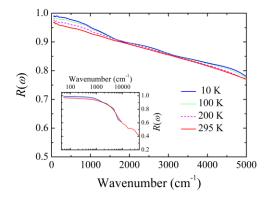


Fig. 1. Temperature-dependent $\it bc$ -plane reflectance spectra of $\rm La_2Pt_3Ge_5$. Inset: the reflectance over broad frequency up to $45000~\rm cm^{-1}$.

mixture as a self-flux. The as-grown crystals have large and flat bc plane. Details of the growth procedure are described elsewhere [6]. We measured the bc-plane reflectance $R(\omega)$ spectra of La₂Pt₃Ge₅ in the energy region between 50 and 8000 cm⁻¹ by using Fourier-transform spectrometer (Bruker Vertex 70v). The absolute value of the reflectance spectra were determined by using in-situ gold overcoating technique [15]. Reflectance spectra were recorded at selected temperatures from 10 K to 295 K. We used the spectroscopic ellipsometer (V-VASE, J. A. Woollam Co.) to obtain the dielectric constants of La₂Pt₃Ge₅ in the energy region between 5600 and 44000 cm⁻¹, from which we calculated the reflectance spectra in the corresponding frequency region. The real part of the bc-plane optical conductivity $\sigma_1(\omega)$ spectra in the frequency region from 50 to 44000 cm⁻¹ were determined by Kramers-Kronig analysis of the reflectance spectra [16].

3. RESULTS AND DISCUSSION

Fig. 1 shows the bc-plane reflectance $R(\omega)$ spectra of La₂Pt₃Ge₅ at several temperatures. The reflectance data showed the characteristic behavior of a good metal. The absolute value of the reflectance data in the far-infrared frequency region was larger than 0.95. The reflectance increased towards lower frequency. As the temperature decreased, the reflectance below 5000 cm⁻¹ increased, indicating the enhancement of coherent electronic response at low temperatures. Inset of Fig. 1 shows that the changes in the optical response with the variation in temperature were confined in the energy region below about 9000 cm⁻¹.

Insight into the electronic response can be gained from the analysis of the optical conductivity spectra. The real part of the *bc*-plane optical conductivity of La₂Pt₃Ge₅ is displayed in Fig. 2. Metallic behavior was evident in the *bc*-plane optical conductivity. A strong Drude-like response centered at zero frequency dominated the far-infrared conductivity, followed by a featureless mid-infrared continuum between 1500 and 6000 cm⁻¹. However, the far-infrared conductivity could not be described by the Drude peaks. Conspicuous absorption

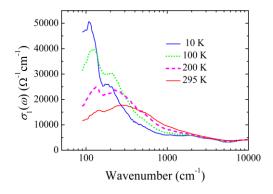


Fig. 2. Real part of the bc-plane optical conductivity spectra of La₂Pt₃Ge₅ at several temperatures.

peaks at about 140 and 300 cm⁻¹ were observed in the optical conductivity data at 295 K.

As the temperature was lowered, the Drude-like response became narrower, indicating the suppression of the scattering of the itinerant carriers. Accordingly, the spectral weigh (SW) was shifted to lower frequencies. The absorption structures registered at about 140 and 300 cm⁻¹ in the conductivity data at 295 K persisted in the optical conductivity at lower temperatures. They also became narrower and shifted to lower frequencies with decreasing the temperature.

Sum rule analysis indicated that the temperature-dependent changes in the optical response involved the redistribution of the SW in the frequency region between 0 and 5000 cm⁻¹. Fig. 3 shows the frequency- and temperature-dependent SW obtained by integrating the real part of optical conductivity:

$$SW(\omega,T) = \int_0^{\omega} \sigma_1(\omega',T) d\omega' \cdot \tag{1}$$

The above integral is global oscillator strength sum rule when the high-frequency cutoff extends to infinity. Depending on the cutoff frequency, the sum rule for different electronic states should be satisfied. We note that the sum rule was satisfied at the cutoff frequency of about 5000 cm⁻¹ for La₂Pt₃Ge₅. This indicates that the variation in temperature affected the electronic states located within the frequency window of 5000 cm⁻¹ near the Fermi level.

In order to get quantitative information on the evolution of the electronic response with temperature, we analyzed the optical conductivity spectra by using the Drude-Lorentz oscillator model:

$$\sigma_{1}(\omega) = \frac{1}{4\pi} \sum_{i} \frac{\omega_{p,i}^{2} / \tau_{i}}{\omega^{2} + (1/\tau_{i})^{2}} + \frac{1}{4\pi} \sum_{i} \frac{S_{j}^{2} \omega^{2} \gamma_{j}^{2}}{(\omega_{i}^{2} - \omega^{2})^{2} + \omega^{2} \gamma_{i}^{2}} . (2)$$

The first term in (2) represents the contribution from the Drude response. Here $\omega_{\rm p,i}$ and $1/\tau_{\rm i}$ are the plasma frequency and the scattering rate of the *i*th Drude peak. The second term, Lorentz oscillator, represents the contribution from optical absorption at finite energies. $S_{\rm j}$, $\omega_{\rm j}$, and $\gamma_{\rm j}$ are the strength, the resonance frequency, and the width of the *j*th bound excitation, respectively.

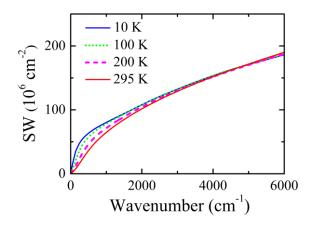


Fig. 3. Frequency- and temperature-dependent spectral weight obtained by integrating the real part of optical conductivity spectra of La₂Pt₃Ge₅.

Fig. 4 shows the results of the Drude-Lorentz oscillator fit of the optical conductivity. The far-infrared conductivity at 295 K could be fitted well with a single Drude and two Lorentz oscillators. The finite-energy optical absorptions represented by the Lorentz oscillators may originate from low-energy interband transitions. An infrared spectroscopic study of Co-doped BaFe₂As₂ superconductors which are the prominent examples of multiband systems suggested the presence of such low-energy interband transitions [14]. The optical conductivity data of BaFe₂As₂ superconductors exhibited a couple of optical excitations in the infrared frequency region. These excitations were attributed to the interband transitions involving the multiple holelike bands near the Fermi level [14]. The observation of the low-energy absorption peaks centered at infrared frequency region might suggest the complex multiband character of La₂Pt₃Ge₅. Density-functional-studies of La₂Pt₃Ge₅ are required to understand the origin of the low-energy optical transitions.

As the temperature was lowered down to 100 K, the Drude oscillator as well as the two Lorentz oscillators became narrower, indicating the decrease in the scattering rate of the itinerant carriers and the increase in the lifetime of the bound optical excitations.

A notable finding of the Drude-Lorentz oscillator model analysis was the presence of the two distinct Drude modes in the electronic response at 10 K. As shown in Fig. 4(d), the far-infrared conductivity could be described well only when the two Drude oscillators with different scattering rates were used. If we used a single Drude component with larger scattering rate, we could not reproduce the rapid increase in the conductivity in the frequency region below about 200 cm⁻¹. On the other hand, if we employed a Drude component with smaller scattering rate, we could not explain the tail of the response of the itinerant carriers located in the frequency region between 300 and 1000 cm⁻¹. The magnitudes of the scattering rates of the narrow and broad Drude responses were estimated to be about 80 and 400 cm⁻¹, respectively. This result suggests that the existence of two groups of charge carries with different scattering rate at 10 K, thus highlighting the multiband nature of La₂Pt₃Ge₅.

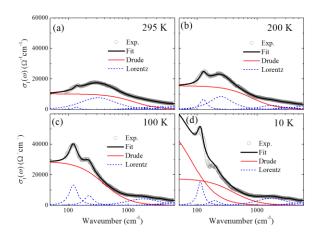


Fig. 4. Results of the fitting of the optical conductivity spectra at (a) 295 K, (b) 200 K, (c) 100 K, and (d) 10 K by using the Drude-Lorentz oscillator model.

Having established the presence of the two distinct Drude components in the optical response at 10 K, we now revisit the optical conductivity spectra at higher temperatures. We note that the far-infrared conductivity taken at higher temperatures could be reproduced well by a single Drude oscillator. Together with the fact that the resistivity and specific heat data did not show any anomaly in the temperature region between 10 and 100 K, this finding indicates that the two electronic states were present at high temperatures but were indistinguishable; if the scattering rates of the two charge carriers were nearly the same, the corresponding coherent response could be represented by a single Drude oscillator with the plasma

frequency of
$$\omega_{p,total} = \sqrt{\sum_{i=1}^{2} \omega_{p,i}^{2}}$$
, where $\omega_{p,i}$ denotes

the plasma frequency of the ith Drude response. Indeed the square of the plasma frequencies of the Drude peak at 295 K $\omega^2_{\rm p,total} \approx 7.5 \times 10^8 \, {\rm cm}^{-2}$ was found to be close to the sum of the squares of the plasma frequencies of two Drude peaks at 10 K which is about $7.0 \times 10^8 \, {\rm cm}^{-2}$. Our analysis of the infrared response of La₂Pt₃Ge₅ suggests that the far-infrared responses of the two electronic states had nearly the same scattering rate at temperatures higher than 100 K and one of the two became much narrower that the other at 10 K: the disparity in the temperature evolution of the charge dynamics of the two electronic states at the Fermi level.

Our finding of the existence of the two Drude components is in line with the results of the specific heat measurements of $La_2Pt_3Ge_5$ which suggested the possibility of the multigap superconductivity. We note that a number of infrared spectroscopic studies of the multiband Fe-based superconductors revealed the presence of the two or more electronic bands near the Fermi level [9]-[14] and their one-to-one connection to the respective superconducting gaps. Infrared spectroscopy experiments in the superconducting states can provide the relationship between the two Drude components and the multiple superconducting gaps in $La_2Pt_3Ge_5$.

4. CONCLUSIONS

We investigated the normal-state charge dynamics of $La_2Pt_3Ge_5$ by employing infrared spectroscopy. We analyzed the optical conductivity spectra by using the Drude-Lorentz oscillator model and obtained detailed information on the evolution of the electronic response with the variation in temperature. The analyses indicated that two Drude components with different scattering rates contributed to the charge dynamics at 10 K whereas a single Drude component governed the charge dynamics at higher temperatures. Our results revealed the two-band nature of the electronic states of $La_2Pt_3Ge_5$ and the disparity of the temperature evolution of the two bands.

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