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Observation of Doppler-shifted cyclotron resonance in LaSb

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Cyclotron resonance (CR) measurements on a single crystal of LaSb have been performed at temperatures between 1.6 and 40 K in the frequency range from 50 to 190 GHz. In addition to ‘‘normal’’ CR, we have observed two anomalous absorption lines that show nonlinear behavior on the frequency-field diagram. The nonlinear behavior is explained by the Doppler-shifted cyclotron resonance coupled with a magnetoplasma wave (Alfven wave) propagating in the quite high-quality single crystal of LaSb.

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LaSb is a simple semimetal with an NaCl-type crystal structure and is considered to be a typical reference compound for CeSb, which shows anomalous physical properties such as dense Kondo behavior and complicated magnetic states.[1,2] Many detailed experimental and theoretical studies on LaSb have been performed as well as on CeSb in order to understand the origin of the anomalous physical properties of CeSb.[3–6] In particular, determination of the Fermi surfaces of LaSb and CeSb have been done in detail by de Haas–van Alphen (dHvA) effect measurements, and the topology of the Fermi surface of CeSb has been understood by considering the p-f mixing effect on the basis of the Fermi surface of LaSb.[7–11]

Measurements of quantum oscillations such as dHvA and Shubnikov–de Haas effects have proven to be powerful methods for investigating Fermi surfaces of metals. On the other hand, although cyclotron resonance (CR) is considered to be the best way to determine cyclotron effective mass of a substance and is supposed to give us other information about the carriers at the Fermi surface,[12–17] there have been only a few reports on rare-earth or uranium compounds due to the difficulty of conducting CR experiments in comparison with dHvA effect experiments.

In a previous paper,[18] we reported the results of CR measurements on LaSb, but it was not satisfactory because the observed absorption was one broad line and the three branches (α, β, and γ branches) of the Fermi surface were not discriminated. In order to obtain more precise and detailed information on this material, we synthesized high-quality single crystals of LaSb. The typical residual resistivity ρ_0 and residual resistivity ratio of the crystals are $\sim$0.13 $\mu\Omega$ cm and $\sim$490, respectively, which showed that the crystals were about 10-times higher in quality than crystals previously used.

Using these single crystals, we successfully observed the CR absorption lines discriminating the α, β, and γ branches in the lower-field region. These lines show a linear relation between frequency and magnetic field according to the usual resonance condition

$$\omega_c = \frac{eB}{m_{\mathrm{CR}}{}^*}, \tag{1}$$

where ω_c is the cyclotron frequency, e the charge of the carrier, B the magnetic field, and $m_{\mathrm{CR}}{}^*$ the cyclotron effective mass. In addition to these lines, we have also observed ‘‘anomalous’’ cyclotron resonances lines that show nonlinear behavior with respect to magnetic field in the higher-field region. The anomalous behavior can be explained by Doppler-shifted cyclotron resonance (DSCR) with Alfven waves in the semimetal LaSb. The purpose of this paper is to present the detailed results of DSCR in LaSb first observed in rare-earth compounds. Detailed results of the ‘‘normal’’ CR have been reported in Ref. 19.

The single crystal was cut into small plates with a size of $1\times1\times0.2\ \mathrm{mm}^3$ along the (001) cleavage plane. Cyclotron resonance measurements for the single crystal have been performed using a vector network analyzer (AB Millimeter Company, Limited) in the frequency region from 50 to 190 GHz in magnetic fields up to 14 T using a superconducting magnet at the High Field Laboratory for Superconducting Materials, Institute for Materials Research, Tohoku University. To observe the resonance lines clearly, sensitivity of the microwave system has to be high and for this purpose, cylindrical cavities were used. We made the cavity using copper in such a way that the resonant frequencies for TE_{011} and TE_{012} modes are 58 and 72 GHz, respectively. In the cavity, the sample was placed at the position where the rf electric field of the microwave is maximum for the TE_{011} or TE_{012} mode. The applied magnetic field B was perpendicular to the rf electric fields of the TE modes. In this work, we performed CR measurements using other TE or TM modes at the same sample position, because the sample experiences the components of the rf electric fields and the signal can be detected.

Figure 1 shows the cavity transmissions for various frequencies at 1.6 K in magnetic fields B parallel to the [001] direction. Here, the B direction is perpendicular to the sample surface, which is not the Azbel-Kaner configuration. The transmissions decrease with increasing magnetic field, which seems to be due to increase of the transversal magnetoresistance. The inset of Fig. 1 shows the spectra at the low-field region. In Fig. 1 and its inset, we can see four absorption lines at each frequency, and these absorption lines are labeled as A–D, respectively.

Figure 2 shows the frequency-field diagram for the $B\|[001]$ direction. The resonance points A and B are on the

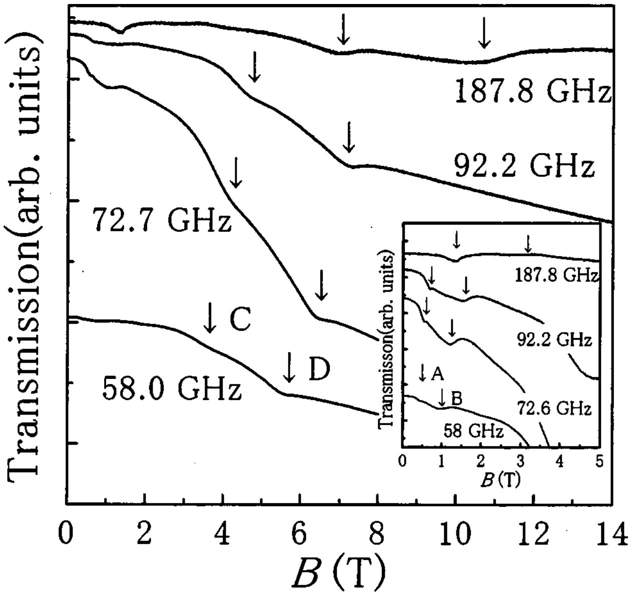

FIG. 1. Cavity transmissions for various frequencies at 1.6 K in magnetic fields parallel to the [001] direction. The inset shows the spectra at the low-field region. The arrows indicate the resonance features.

straight lines satisfying Eq. (1). From the temperature and angular dependence the absorption lines A and B are identified as the cyclotron resonance of the carriers in the α and γ branches, respectively.[19] The determined $m_{\mathrm{CR}}{}^*$ are $0.20m_0$ and $0.45m_0$ for the α and γ branches, respectively. On the other hand, the resonance frequencies of C and D show nonlinear behavior with respect to the magnetic field. In addition, their resonance fields are fairly high. That is, if these C and D resonances are assumed to be CR lines, the effective masses are estimated to be over $1.0m_0$, of which values are much larger than the expected masses in LaSb.[5,6] Then it is impossible to explain such features by the normal CR relation. However, in increasing the temperature, the absorption lines C and D are simultaneously undetectable with the A and B lines as shown in Fig. 3. This fact indicates that the C and D lines are originated from the cyclotron motion of the carriers as are the A and B lines.

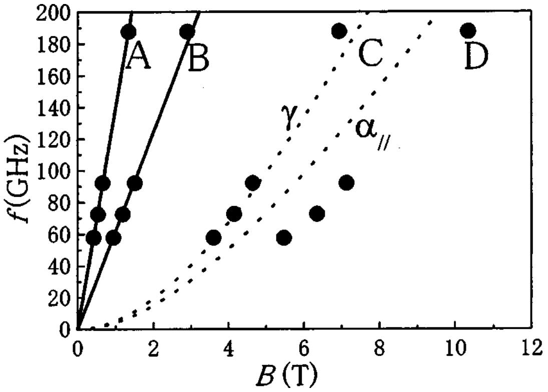

FIG. 2. Frequency-field diagram of LaSb at 1.6 K for $B \parallel [001]$. The dotted lines are calculated from Eq. (3) and correspond to the edges of the γ and $\alpha_{\parallel}$ branches, respectively.

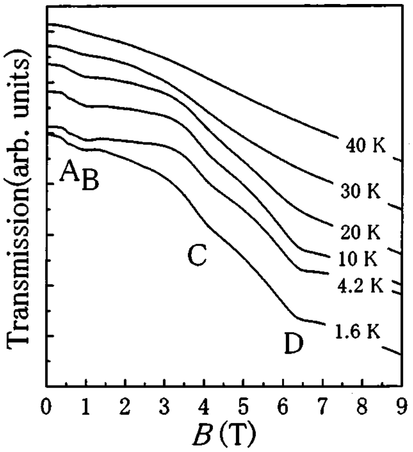

FIG. 3. Cavity transmissions at various temperatures at 72.8 GHz in magnetic fields parallel to the [001] direction.

In general, the microwave is very rapidly attenuated due to the skin effect as it penetrates into the metals or semimetals. In these cases, only a few carriers in the skin depth directly couple with the microwave, resulting in the normal CR. Most of the carriers are inside the skin depth, and they cannot couple with the microwave on the surface of the material. It is known, however, that the electromagnetic wave of the frequency $\omega \ll \omega_c$ can propagate inside the bulk of the metal or semimetal as a magnetoplasma wave in high magnetic field if the sample quality is very high.[20,21] For a compensated semimetal such as LaSb, this wave is called the Alfven wave. In this case, there is a possibility that the carriers couple with a microwave even in the bulk.

In our experimental condition, in which the B direction is perpendicular to both the sample surface and the rf electric field, the Alfven wave propagates along the B direction (z direction). A carrier that has a velocity component v_z along the z direction couples with the rf field of the Alfven wave with the Doppler-shifted frequency $\omega \pm k v_z$, where ω and k are the frequency and wave number of the Alfven wave, respectively. In this case, DSCR occurs when the cyclotron frequency ω_c coincides with $\omega \pm k v_z$.

The dispersion relation of isotropic Alfven waves under the conditions $\omega_c \tau \gg 1$ and $\omega \ll \omega_c$ is given by

$$k^2 = \frac{\mu \Sigma_j n_j m_j}{B^2}\omega^2 = \frac{\lambda}{B^2}\omega^2, \tag{2}$$

where μ is the magnetic susceptibility, n_j the density of the jth carrier, and m_j is its mass.[20,21] Substituting Eq. (2) in the resonance condition, we have the magnetic-field dependence of the resonance frequency ω_c' for DSCR:

$$\omega_c' = \frac{\omega_c}{1 + (\nu_z \sqrt{\lambda})/B}. \tag{3}$$

Here, we adopt the condition $\omega_c = \omega + k v_z$. The other condition, $\omega_c = \omega - k v_z$, means that the resonance field shifts to the lower-field side and the Alfven wave with this frequency cannot propagate because the condition $\omega \leqslant \omega_c$ is not satisfied. Consequently no resonance absorption is observed at this frequency.

The relation of Eq. (2) is the isotropic case. But the Fermi surface of LaSb is anisotropic and consists of three branches α, β, and γ. So we have to determine λ in Eq. (2) more carefully according to the empirical data of this crystal. The α branch is the electron Fermi surface with an ellipsoidal shape at the X point. The β branch is the hole Fermi surface, almost spherical and centered at the Γ point. The γ branch is also the hole surface at the Γ point and is slightly elongated along the $\langle 100 \rangle$ direction.[5,6] For simplification, we assume the γ branch to be a sphere. Then the dispersion relation of the Alfven waves in LaSb is written as

$$k^2 = \frac{\mu[(2/3)n_\alpha m_t + (1/3)n_\alpha m_1 + n_\beta m_\beta + n_\gamma m_\gamma]}{B^2}\,\omega^2 = \frac{\lambda}{B^2}\,\omega^2, \tag{4}$$

where n_α, n_β, and n_γ are the densities of the carriers in the α, β, and γ branches; m_t and m_l are the transverse and longitudinal masses of the α branch; and m_β and m_γ are the effective mass of the β and γ branches, respectively. In DSCR measurements, a detected anomaly is mainly due to the carriers around the edge of the Fermi surface in which the carrier has the maximum Fermi velocity along the B direction.[20,21] LaSb has four edges of the Fermi surface for the [001] direction, that is, the edges of the β branch, the γ branch, and the two types of the α branch ($\alpha_\perp$: the longitudinal axis of the spheroid is parallel to the [001] direction; $\alpha_\parallel$: the axis is perpendicular to the [001] direction). The dotted lines in Fig. 2 are calculated from Eq. (3) using the parameters obtained experimentally for the edges of the γ and $\alpha_\parallel$ branches, respectively. The agreement between the experimental data of C and D and the calculated lines is reasonably good. Therefore, we conclude that the C and D lines are due to DSCR of the carriers at the edges of the γ and $\alpha_\parallel$ branches, respectively. Here, the carrier densities n_α $(=2.0\times10^{26})$, n_β $(=0.5\times10^{26})$, and n_γ $(=1.5\times10^{26}\ \mathrm{m}^{-3})$ are taken from Ref. 22 and the effective-mass parameters m_t $(=0.20)$, m_β $(=0.17)$, and m_γ $(=0.45m_0)$ are deduced from the CR measurement in Ref. 19. Unfortunately, m_1 is not determined experimentally, so we use $m_1 = 2.6m_0$, assuming that the mass ratio m_t/m_1 $(=0.077)$ is equal to that of the band calculation ($m_t = 0.13$ and $\sqrt{m_t m_1} = 0.47m_0$).[5] Since the Fermi velocities v_F at the edges also have not been reported, v_F $(=0.46\times10^6$ for the $\alpha_\parallel$ branch and 0.6×10^6 m/s for the γ branch) are roughly estimated from the results of the dHvA measurements using the relation $\nu_F = \hbar k_F/m^*$.[6]

In this work, the edges of the β and $\alpha_\perp$ branches have not been observed. The carriers in a branch have various velocity components along the B direction in the range of $-v_F \leqslant v_z \leqslant v_F$ and damp the Alfven waves in the region $\omega - kv_F \leqslant \omega_c \leqslant \omega + kv_F$. Therefore, the edge that shifts to the highest field would be observed most clearly and the rest of the carriers in this branch prevent DSCR of the other branches. In the case of LaSb, according to our estimation the resonance field of the $\alpha_\parallel$ branch is largest and the resonance fields of the β and $\alpha_\perp$ branches are smaller than those of the γ and $\alpha_\parallel$ branches at a fixed frequency. Therefore, the edge of the $\alpha_\parallel$ branch can be observed most clearly, and it would be more difficult to observe the edges of the β and $\alpha_\perp$ branches.

In summary, we have performed the cyclotron resonance measurements on a single crystal of LaSb at temperatures between 1.6 and 40 K in the frequency range from 50 to 190 GHz. In addition to "normal" CR, we have observed two anomalous absorption lines that show nonlinear behavior on the frequency-field diagram. The nonlinear behavior is explained by Doppler-shifted cyclotron resonance with Alfven waves.

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