Highly Anisotropic Pressure Dependence of the Fermi Surface of CeSb

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We report a de Haas–van Alphen effect study of CeSb under pressure in the ferromagnetic phase. The frequencies of the electron Fermi surfaces and one particular hole surface increase considerably with increasing pressure, while those of the other hole surfaces decrease slightly. On the other hand, the effective masses increase similarly for all the hole surfaces with pressure, while those of the electron surfaces change little. The frequency changes seem to be explained qualitatively by considering the anisotropic p-f mixing model. However, it is difficult to understand the changes of the effective masses in terms of the f content expected for the p-f mixing model. [S0031-9007(97)02710-5]

Strongly correlated f-electron systems (SCES) have been studied for more than a decade because of their interesting physical properties. However, our understanding of their properties is far from complete. The ground states of U-based systems were classified by Hill according to their U-U spacing, which is known as the Hill plot [1]. A similar classification was also performed for Ce-based systems [2]. Although these classifications, based on the interatomic distance, may roughly explain the properties of SCES, many systems cannot be classified by these simple schemes.

To clarify the relationship between the properties and the interatomic distance, studies under pressure have been performed. These studies have revealed that the properties of SCES are very sensitive to pressure compared to those of normal metals [3], which indicates that the electronic structures of SCES are very sensitive to the interatomic distance. Therefore, it is expected that a de Haas–van Alphen (dHvA) effect study under pressure is very useful for revealing the nature of SCES because the dHvA effect is a powerful tool to clarify the electronic structure of the material.

CeSb is one of the intriguing SCES which has been studied intensively. It crystallizes in the NaCl structure. In the paramagnetic state, the temperature dependence of the electrical resistivity clearly shows typical dense Kondo behavior [4] which has also been observed in Ce-based metallic systems such as CeB_6 [5] or $CeCu_6$ [6]. However, CeSb is different from those compounds in that it is a semimetal whose carrier concentration is a few percent per Ce atom.

In the ordered states, CeSb shows a complex magnetic phase diagram [7]. At low temperatures, the spin arrangements are described by the stacking of ferromagnetic (001) planes with magnetic moments perpendicular to the planes. When a magnetic field of about 4 T is applied parallel to the [001] direction, the system enters a ferromagnetic phase where all the spins are aligned parallel to the magnetic field. The magnetic moment is $2.10\mu_B$, simi-

lar to the saturated moment for the $|J, J_Z\rangle = |5/2, 5/2\rangle$ state. The direction of the magnetic moment is strongly pinned along one of the $\langle 100 \rangle$ directions close to the applied field direction.

The Fermi surface (FS) properties of CeSb have been studied experimentally [8–10]. The topology of the FS's in the ferromagnetic phase can be explained by band structure calculations based on the anisotropic p-f mixing model [11–13]. In the p-f mixing model of CeSb the anisotropic mixing effect between the $4f$-Γ_8 states and the $5p$-Γ_8 bands is considered to be important. Because of p-f mixing, the $4f$ states are pulled down, and the $5p$ bands are pushed up by the bonding-antibonding effect to gain energy. According to experiments and the band structure calculation, the FS's consist of a singlefold ellipsoidal electron FS centered on the X_z point (α), twofold ellipsoidal electron FS's which exhibit d-f exchange splitting centered on the X_x and X_y points (γ), and four singlefold closed-hole FS's centered on the Γ point (β_1, β_2, β_3, and β_4). Here the Greek letters in parentheses denote the corresponding frequencies observed in the experiments. The electron FS's arise mostly from the $5d$ bands of Ce, while the hole FS's arise mostly from the $5p$ bands of Sb. The β_4 frequency can be explained as arising from the particular valence band which mixes strongly with the $4f$ $|J, J_Z\rangle = |5/2, 5/2\rangle$ state and is pushed up due to p-f mixing. The other hole FS's are ascribed to the valence bands which have a little p-f mixing.

Although the topology of the FS's observed in experiments can be explained theoretically, the enhancement of the effective mass (m^*) has been controversial. The low temperature electronic specific heat coefficient obtained by specific heat measurement at zero magnetic field was 3 mJ/mole K^2 [14], which is three times larger than the value 1 mJ/mole K^2 estimated with the m^*'s determined by quantum oscillation experiments [10]. To understand this point, it is necessary to clarify how the f states affect the m^*'s, as well as the topology of the FS's.

In this paper we report a dHvA study of the pressure dependence of the frequencies and the m^*'s for all the FS's of CeSb in the ferromagnetic phase. From these results the highly anisotropic change of the FS properties has been revealed for the first time in SCES. Results will be discussed in light of the p-f mixing model.

The details of the sample preparation have been published elsewhere [15]. The dHvA measurements were performed in a top-loading dilution refrigerator. Magnetic fields were applied parallel to the [001] direction of the sample by a superconducting magnet. The maximum field used was 14 T. Since the dHvA oscillation is periodic in $1/B$ rather than $1/H$, we have corrected the effect of the magnetization to obtain the true dHvA frequency. Here $B = H + 4\pi(1 - D)M$, M is the magnetization, and D is the effective demagnetization coefficient of the sample. The sample shape is a rectangular parallelepiped, and D is calculated to be 0.14. Values of the dHvA frequency thus obtained are larger than the values reported earlier by a few percent [8–10]. This correction is independent of pressure, because the pressure dependence of both the magnetization [16] and D is negligible.

A standard Cu-Be pressure clamp was used to apply pressure to the sample. The maximum pressure used was 6.5 kbar. We used a 1:1 mixture of Daphne 7373 oil and kerosene as the pressure transmitting medium. The pressure at low temperatures was calibrated by measuring the superconducting transition temperature of Sn [17]. Deviation from hydrostatic pressure and its effect on the present measurement are estimated to be negligible by observing that the superconducting transition width of Sn does not change and amplitudes of the dHvA oscillations in CeSb do not show an appreciable decrease under the pressure used in this study.

The dHvA oscillations were measured with a standard field modulation technique. The modulation field was applied from the outside of the pressure clamp. Relatively low frequencies, between 2 and 7 Hz, were employed to reduce the amount of the Joule heating of the pressure clamp due to eddy currents generated by the modulation field.

Since the pressure clamp is heated because of the modulation field, there is a difference in temperatures between the thermometers and the sample in the pressure clamp. Therefore, the measured m^*'s can be different from the true ones. We will show that the true ones were determined within the experimental error as follows: The modulation amplitude and frequency used mostly in the m^* measurements were 70 G and 2 Hz, respectively. This frequency and amplitude were chosen so that the heating power is small enough, and at the same time the dHvA signal is large enough. The temperature inside the pressure clamp was measured by a RuO$_2$ resistance thermometer as functions of modulation frequency, amplitude, and temperature before the dHvA experiments. The temperatures during the experiments were measured by RuO$_2$, Ge, and carbon re-

sistance thermometers placed in the mixing chamber. The m^* measurements were performed at temperatures above 200 mK, where the difference between the temperatures of the thermometers and the sample is not significant with this modulation condition. The m^*'s are measured at ambient pressure with this modulation condition in two ways, i.e., by the standard setting without the pressure clamp and with the sample inside the pressure clamp. Both values agree well within the experimental error.

Particular care was taken when measuring the m^* of β_4 because it is the largest one, and, consequently, the measurement is most affected by the temperature error. The m^* was measured as a function of the heating power, i.e., modulation amplitude h and/or frequency ω and was plotted against the heating power $h^2\omega^2$. Then the data were extrapolated to the zero-heating condition. The difference between the obtained value and the one measured using the modulation condition mentioned above is $0.1m_0$. Namely, they are in agreement with each other within the experimental error.

Figure 1 shows a recorder trace of the dHvA oscillations observed at 6.5 kbar. The corresponding Fourier spectrum is shown in the upper panels of Fig. 2, while the lower panels show the spectrum at 1.5 kbar. It can be clearly seen that the frequencies α, γ, and β_4 at 6.5 kbar are considerably higher than those at 1.5 kbar. On the other hand, those of β_1, β_2, and β_3 at 6.5 kbar are lower than those at 1.5 kbar, and the magnitudes of the changes are much smaller than those of α, γ, and β_4.

The Fourier peak of the γ frequency splits into two or more, depending on the pressure or the field range used for the Fourier transform. Similar splitting phenomena are observed at ambient pressure. Although no clear explanation has been given for the splitting behavior so far, we think that the artifact of the Fourier analysis, combined with the field dependence of the exchange splitting, may be responsible.
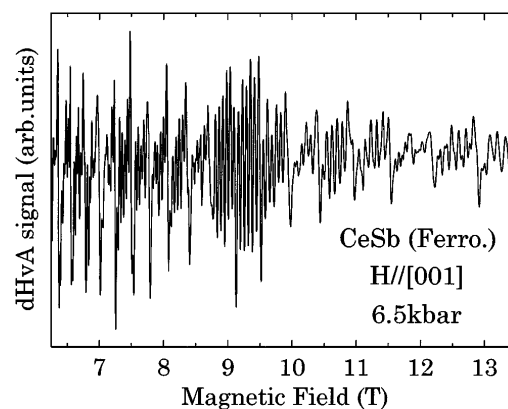

FIG. 1. Recorder trace of the dHvA oscillations in the ferromagnetic phase observed at 6.5 kbar for the magnetic field parallel to the [001] direction. The temperature of the mixture during the measurement is 200 mK.
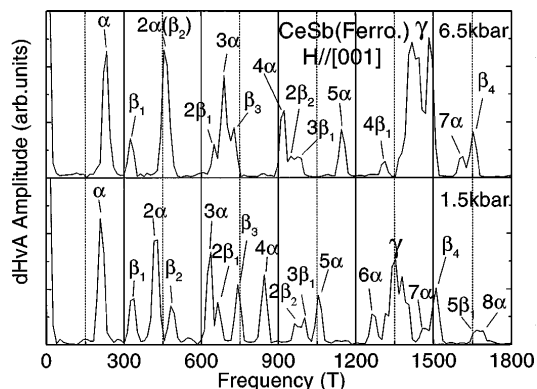
FIG. 2. Upper panel: Fourier spectrum of the oscillations shown in Fig. 1. Lower panel: Fourier spectrum of the oscillations observed at 1.5 kbar.

Figure 3 shows the fractional changes of the frequencies, i.e., $\Delta F/F(0) = \{F(P) - F(0)\}/F(0)$ as a function of pressure. Table I is a summary of the frequencies at ambient pressure and their pressure coefficients $d \ln F/dP$ determined by fitting a straight line to each set of data in $\ln F$ vs P plots. The α and γ frequencies which arise from the electron FS's increase considerably with pressure. On the other hand, the pressure effect of the hole FS's are qualitatively different among them. The frequency of β_4 increases at a larger rate than those of α and γ, while those of other hole FS's (β_1, β_2, and β_3) decrease and the magnitude of the rate is less than one-third that of β_4.

The pressure dependence of the frequencies are quite different from those observed in the non-f reference
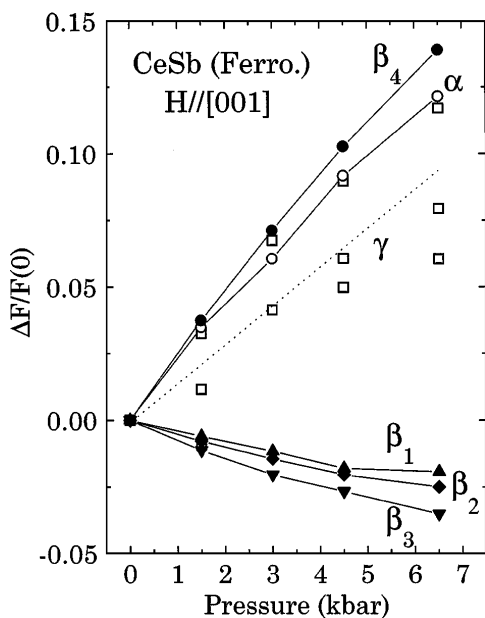

FIG. 3. Fractional changes of the frequencies $\Delta F/F(0) = \{F(P) - F(0)\}/F(0)$ as a function of pressure. The solid lines and a broken line are guides for the eye.

TABLE I. Frequencies at ambient pressure and their pressure coefficient $d \ln F/dP$. The pressure coefficient of γ has been estimated by fitting a line to all the data points of γ in Fig. 3.

	F(T)	$d \ln F/dP$ ($\times 10^{-3}$ kbar^{-1})
Electron FS's		
α	205	+18
γ	1330	+14
Hole FS's		
β_1	330	−3
β_2	490	−4
β_3	750	−5
β_4	1450	+20

material LaSb [18] which has the same crystal structure and whose carrier concentration is two-thirds that of CeSb [10]. In LaSb all the frequencies increase with pressure. The pressure coefficients are between about $+3 \times 10^{-3}$ kbar^{-1} and $+7 \times 10^{-3}$ kbar^{-1}, which are much less than those of α, γ, and β_4.

Table II is a summary of the m^* data measured both at ambient pressure and at 6.5 kbar for all the frequencies. The data at ambient pressure shown here were measured by the standard setting, and they are in agreement with those reported previously [8,19], except for that of β_4. The m^* of β_4 at ambient pressure has been determined to be $(1.9 \pm 0.15)m_0$. All the m^*'s of the electron FS's do not change within the experimental error. On the other hand, the m^*'s of the hole FS's increase similarly with pressure. The increase for the m^*'s of β_2 and β_4-FS's are obvious beyond the experimental error. However, considering the large experimental error, those of β_1 and β_3-FS's might be small [20].

It is noted that the m^* of β_4 measured in this experiment is about half of that measured using the acoustic dHvA effect [10]. The m^* with fields in the [111] direction is also found to be small, i.e., $1.8m_0$, compared with that

TABLE II. Effective masses measured at ambient pressure and at 6.5 kbar. For the effective mass of γ, we have determined the effective mass using the Fourier spectrum of the field range between 7.6 and 4.6 T, where the Fourier peak of γ splits into about two peaks. The γ (higher) and the γ (lower) refers to the data of the higher frequency and those of the lower frequency, respectively.

	$m^*(m_0)$ (ambient pressure)	$m^*(m_0)$ (6.5 kbar)
Electron FS's		
α	0.23 ± 0.02	0.22 ± 0.03
γ (higher)	0.8 ± 0.1	0.9 ± 0.1
γ (lower)	0.9 ± 0.1	0.9 ± 0.1
Hole FS's		
β_1	0.5 ± 0.05	0.6 ± 0.05
β_2	0.9 ± 0.1	1.4 ± 0.2
β_3	1.2 ± 0.1	1.5 ± 0.2
β_4	1.9 ± 0.15	2.4 ± 0.25

obtained by the acoustic dHvA effect. The relatively small m^* can be qualitatively confirmed by observing that the amplitude of the β_4 oscillation is the largest among other frequencies and that the signal can be observed in fields down to 6 T and at temperatures up to 1.5 K in the [111] direction. Moreover, since the m^*'s, except for that of β_4, are in good agreement with those measured by two different groups with different samples and experimental settings [8,19], it is unlikely that the difference comes from sample differences or experimental mistakes.

The highly anisotropic pressure dependence of all the frequencies in this study seems to be explained qualitatively by taking the anisotropic p-f mixing model into account [11–13]. We discuss two effects which change all the FS's almost uniformly.

(1) With increasing pressure, the volume of the reciprocal lattice or the FS increases because the volume of the real-space lattice decreases. The pressure coefficient $d \ln F / dP$ due to this effect is given by $2\kappa/3$, where κ is the volume compressibility. We estimate the value of $2\kappa/3$ to be $+1 \times 10^{-3}$ kbar^{-1} [21], which is much smaller than those observed in this study.

(2) Other effects which can increase the frequencies considerably have to be considered. As the pressure is increased, the overlap of the wave functions between the p bands of Sb and the d bands of Ce increases. Therefore, the system becomes more metallic, or the valence bands go up and the conduction bands come down with respect to the Fermi energy E_F, i.e., both the hole and electron FS's increase. However, this effect cannot explain the very anisotropic change among the hole FS's.

In order to understand the pressure dependence of all the frequencies, it is necessary to consider an anisotropic effect. According to the p-f mixing model, the p-f mixing is strong in the particular valence band from which β_4 arises [11–13]. With increasing pressure, the p-f mixing becomes stronger because the overlap of the wave functions between the p bands of Sb and the f states of Ce increases. Therefore, it is expected that this particular valence band is preferentially pushed up further with pressure. This effect causes the volume of the β_4-FS to increase. Then, the E_F increases to compensate the numbers of holes and electrons. Consequently, the volumes of the electron FS's increase, while the volumes of the hole FS's, other than the β_4-FS, decrease.

If we take the last effect into account, in addition to the effects (1) and (2), the anisotropic frequency changes may be explained. Namely, all three effects cause an increase in the volumes of the electron FS's and the β_4 FS, while the last effect is compensated by (1) and (2) for the other hole FS's to give only small changes.

It is difficult to explain the pressure dependence of the m^*'s in a straightforward manner by the p-f mixing model. It is expected from dHvA effect studies and the band structure calculations of many other SCES that the mass enhancement is large for the band whose f content is large. According to the interpretation of the pressure dependence of the frequencies based on the p-f mixing model, it may be assumed that the f content of the β_4-FS increases considerably with pressure, while those of other hole FS's do not change very much. Therefore, it is difficult to explain the result that the increase of the m^* for β_4-FS shows a similar trend to that of the other hole FS's. The present observation might imply that the f content is not significant for the mass enhancement in CeSb, or the band masses change to compensate the mass enhancement due to the many-body effect. A band structure calculation study under pressure will be helpful to clarify this point.

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[20] When the m^*'s at both ambient pressure and 6.5 kbar are determined by using the same experimental condition, i.e., same field range, same temperature range, etc., the experimental errors are smaller than those given in Table II and the m^*'s always increase with pressure for all the hole FS's beyond experimental error. However, the value of the m^* itself depends slightly on the experimental condition. The values of the m^*'s and their experimental error in Table II have been determined by considering several sources of error and also the differences between different experimental conditions.

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