Hall effect in single crystal CeCu$_2$Si$_2$ under high pressure

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Abstract. Hall effect measurements were carried out under high pressures up to 4.9 GPa in a single crystal of CeCu$_2$Si$_2$. The temperature dependence of the Hall coefficient is interpreted to be composed of two peaks below room temperature. The high-temperature peak shows strong pressure dependence, where the peak shifts from 20 K at ambient pressure to 125 K at 4.9 GPa. This peak is a consequence of the anomalous Hall effect due to skew scattering. The low-temperature peak shifts from 5 K at ambient pressure to 15 K at 4.9 GPa and the magnitude of the peak shows the maximum around 4.1 GPa at which the superconducting transition temperature also reaches the maximum.

1. Introduction

The heavy-fermion compound CeCu$_2$Si$_2$ is a superconductor with $T_c \approx 0.6$ K at ambient pressure [1]. This unconventional superconductivity is considered to be mediated by antiferromagnetic (AFM) spin fluctuations around an AFM quantum critical point such as in CePd$_2$Si$_2$ and CeIn$_3$ [2]. With increasing pressure, $T_c$ is enhanced above 3 GPa and becomes maximum around $P_c \approx 4.5$ GPa, although the AFM spin fluctuations are drastically suppressed under pressure [3]. The enhanced superconductivity soon disappears with further application of pressure above $P_c$. In the chemical substitution system CeCu$_2$(Si$_{1-x}$Ge$_x$)$_2$, the superconducting phase in the $P$-$T$ phase diagram splits into two domes, which suggests a different origin for superconductivity at ambient pressure and high pressures around $P_c$ [4].

Recently it was suggested theoretically that the possible origin of the enhancement of $T_c$ around $P_c$ is critical valence fluctuations [5, 6]. The $T$-linear dependence of resistivity and the enhancement of the residual resistivity around $P_c$ were explained by the valence fluctuation scenario [5]. The NQR study at 4.2 K revealed the downward deviation of $^{63}v_Q$ of Cu-nuclei above 3.5 GPa from the linear pressure dependence which is predicted from the volume compression [3]. This result may reflect the change of valence. The pressure dependence of the Ce valence at 14 K was recently measured by XAS [7]. The valence of Ce smoothly decreases with increasing pressure and no sharp valence transition at $P_c$ was observed. In summary, although a number of low-temperature properties suggest the presence of a valence transition, no direct evidence of such a transition has been obtained so far.
The Hall coefficient reflects the transport properties of carriers and is expected to be sensitive to the valence instability. We have measured the Hall coefficient in polycrystalline CeCu$_2$Si$_2$ under pressure [8]. The temperature dependence of the Hall coefficient has a peak at around 10 K. The magnitude of the Hall coefficient at the peak temperature shows the decreasing trend with increasing pressure and is enhanced from 3.1 to 4.2 GPa at which $T_c$ is enhanced. In this study, the Hall effect was measured under high pressure in single crystal of CeCu$_2$Si$_2$.

2. Experimental

The single crystal of S-type CeCu$_2$Si$_2$ was grown by a self-flux method combined with a Bridgman cooling technique [9]. The pressure up to 4.9 GPa was applied in an indenter cell [10] with Daphne oil 7474 [11] as a pressure transmitting medium. The pressure was determined from the superconducting transition temperature of Pb [12]. The Hall voltage $V_H$ was measured down to 1.4 K with AC resistance bridge in the field-sweep measurement. External magnetic field was applied along the [100] direction, which is the hard axis of magnetization. The Hall resistivity is given by

$$R_H(H) = f(\frac{V_H(H) - V_H(-H)}{2I})$$

where $a$ and $I$ are the thickness of the sample and the electric current, respectively. The Hall coefficient $R_H$ was obtained from the slope of $H(H)$. The electrical resistivity was measured up to 4.5 GPa for the sample cut from the same grain to check the $T_c$ using a dilution refrigerator. The pressure dependence of $T_c$ in the present sample reproduces the previous result [5].

3. Results and Discussion

![Figure 1](image1.png)  
**Figure 1.** Temperature dependence of the Hall coefficient $R_H$ in CeCu$_2$Si$_2$ at various pressures.

![Figure 2](image2.png)  
**Figure 2.** Hall coefficient $R_H$ as a function of $\rho \chi$ in CeCu$_2$Si$_2$ at ambient pressure.

Figure 1 shows the temperature dependence of the Hall coefficient $R_H$ in CeCu$_2$Si$_2$ at various pressures. The Hall resistivity increases linearly with increasing field up to 5 T in the present experimental range of temperature and pressure. The $R_H$ at ambient pressure increases with decreasing temperature and reaches the maximum at $\sim$ 5 K. This behavior is almost the same as our previous result in the polycrystalline sample [8]. In heavy fermion systems, the anomalous Hall effect $R_H^a$ due to skew scattering causes this kind of strong temperature dependence. The $R_H^a$ is theoretically proportional to the magnetic susceptibility $\chi$ [13] or the product of $\chi$ and the electrical resistivity $\rho$ [14] at temperatures higher than the coherence temperature of the Kondo effect. The $R_H^a$ decreases with decreasing temperature below the coherence temperature.
The peak temperature of \( R_H \) roughly corresponds to the coherence temperature. The \( R_H \) in CeCu_2Si_2 at ambient pressure is plotted against \( \rho \chi \) in Fig. 2. The linear relation between \( R_H \) and \( \rho \chi \) holds above 100 K.

The \( R_H \) below 100 K deviates from the linear dependence at higher temperatures. The \( R_H \) between 35 and 14 K is proportional to \( \rho \chi \) with different slope above 100 K. Note that 14 and 100 K correspond to the characteristic temperatures \( T_{\text{max}}^H \) determined from the peaks in the resistivity, respectively. The \( R_H \) deviates from the linear relationship below 14 K. The \( \rho \chi \) in this temperature region is expected to decrease with decreasing temperature because \( T_{\text{max}}^H \) corresponds to the coherence temperature. Therefore, the maximum of \( R_H \) at 5 K cannot be assigned to the contribution of \( R_H^0 \) due to skew scattering.

The \( R_H \) above 3.4 GPa clearly has two peaks, which are observed, for example, at \( T_{m}^L = 9 \) K and \( T_{m}^H = 105 \) K at 4.3 GPa, as shown in Fig. 1. The low-temperature and high-temperature peaks are denoted as \( T_{m}^L \) and \( T_{m}^H \), respectively. Note that the high-temperature peak at \( T_{m}^H \) was not clearly seen up to 4.8 GPa in our previous report for polycrystalline sample [8]. This difference may be caused by the anisotropic Hall coefficient. The pressure dependences of \( T_{m}^L \) and \( T_{m}^H \) are summarized in Fig. 3 (a). The \( T_{m}^L \) and \( T_{m}^H \) below 2.6 GPa, at which two peaks are not clearly separated, are determined from the peak and the inflection point of \( R_H \), respectively.

The pressure variation of \( T_{m}^H \) almost follows that of \( T_{1}^{\text{max}} \). Therefore, the high-temperature peak of \( R_H \) at \( T_{m}^H \) is likely due to the anomalous Hall effect.

Figure 3. Pressure dependence of (a) \( T_{m}^H \), \( T_{m}^L \) in \( R_H \) and \( T_{m}^{\text{max}} \) in \( \rho \) (see text), and (b) the magnitude of \( R_H \) at \( T_{m}^H \) and \( T_{m}^L \). The dotted curves are guide to eye.

Figure 4. Pressure dependence of (a) \( T_c \) and (b) residual resistivity \( \rho_0 \) (solid circle) and resistivity at 2 K (open circle). The dotted curves are guide to eye.

The magnitude of \( R_H \) at \( T_{m}^L \), shown in Fig. 3 (b), is almost unchanged up to 1.9 GPa, and decreases with increasing pressure up to 3.4 GPa. Then, it increases rapidly and exhibits the maximum at 4.1 GPa. The \( T_c \) of our sample reaches its maximum value at around 4.0 GPa, as shown in Fig. 4 (a). Therefore, we conclude that the enhancement of both the \( R_H \) at \( T_{m}^L \) and the \( T_c \) under pressure occurs simultaneously. The temperature \( T_{m}^L \) shows a minimum at 4.1 GPa and increases above 4.1 GPa, as shown in Fig. 3 (a). These results imply the existence...
of fluctuations around 4.1 GPa. The residual resistivity of this sample reaches the maximum at 4.6 GPa, as shown in Fig. 4 (b). This implies the existence of $P_c = 4.6$ GPa within the valence fluctuations scenario. The valence fluctuations may also influence the $R_H$. However, the pressure at which $R_H$ is enhanced is slightly lower than the pressure at which $\rho_0$ reaches the maximum.

The enhancement of the $R_H$ due to AF fluctuations was reported for CeRhIn$_5$ [15]. The $R_H$ of CeRhIn$_5$ is critically enhanced around 2 GPa, at which the AF metallic and superconducting states are separated. The enhancement of the $R_H$ decreases with increasing the magnetic field due to the suppression of AF fluctuations. In the case of CeCu$_2$Si$_2$, we did not observe a field dependent $R_H$. Therefore, the possibility of the AF fluctuations for the origin of the enhancement of the $R_H$ at 4.1 GPa is ruled out.

4. Conclusion

At ambient pressure, the temperature dependence of the Hall coefficient $R_H$ along [100] has a linear relation to $\rho_X$ above 100 K, which is explained by the sum of the ordinary Hall coefficient and the anomalous Hall coefficient due to skew scattering. The maximum in $R_H$ at 5 K is not assigned to the anomalous Hall effect. The $R_H$ under pressure above 3.4 GPa exhibits two peaks. The pressure dependence of the high-temperature peak roughly follows that of $T_{c1}^{\max}$ in the resistivity. The origin of high-temperature peak is assigned to the anomalous Hall effect due to skew scattering. The magnitude of $R_H$ at the low-temperature peak increases with increasing pressure above 3.4 GPa and reaches the maximum at 4.1 GPa, at which $T_c$ also reaches its maximum. This result indicates the close relationship between the enhancement of $R_H$ at low temperatures and the increase of $T_c$.

Acknowledgments

This work was partially supported by a Grant-in-Aid for Scientific Research on Innovative Areas “Heavy Electrons” (No. 20102008) from the Ministry of Education, Culture, Sports, Science and Technology. This work was also supported by Scientific Research B (20340090) from the Japan Society for the Promotion of Science.

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