Magnetic phase diagram and Néel critical field in CeCu$_2$Ge$_2$

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Abstract. We present magnetic phase diagrams to determine Néel critical field on the basis of electrical resistivity, specific heat and magnetic susceptibility measurements at magnetic fields. The magnetic phase diagrams indicate that Néel temperatures were suppressed completely at 35T for [100] and 31T for [001], respectively. These results indicate Quantum Critical Point exists in a range from 30 to 40T. This phase diagrams should be powerful guides for investigating electronic states near QCP.

1. Introduction
The heavy-fermion antiferromagnet CeCu$_2$Ge$_2$ is a good reference compound to the prototype heavy fermion superconductor CeCu$_2$Si$_2$ because both compounds crystallize into the same ThCr$_2$Si$_2$-type tetragonal structure. CeCu$_2$Ge$_2$ also shows superconductivity above 7.5GPa when antiferromagnetic order is suppressed by high pressure. CeCu$_2$Ge$_2$ orders at 4K to incommensurate magnetic structure with a propagation vector of $\mathbf{Q}=(0.28, 0.28, 0.54)$. The magnetic moment is 0.74$\mu_B$ at $T=1.5$ K, which is considerably smaller than the moment 1.54$\mu_B$ estimated from the $\Gamma_7$-ground state split by crystalline electrical field (CEF) [1].

A quantum critical point (QCP) occurs when an ordered state of matter becomes unstable at the absolute zero temperature. When antiferromagnetic order parameter vanishes, quantum fluctuations emerge to mediate exotic superconductivity sometimes. In CeCu$_2$X$_2$(X=Si, Ge), many high pressure ($p$) versus temperature ($T$) and alloying (Si-Ge substitution) versus $T$ studies have been performed to draw magnetic phase diagram [2, 3, 4]. Contrasting to various high pressure studies among heavy fermion systems including CeCu$_2$Ge$_2$, $H$-$T$ phase diagram is not well investigated, although magnetic field is an alternative powerful external parameter for tuning electronic states of condensed matter. In this paper, we present the magnetic phase diagrams and critical fields for a- and c-axes in CeCu$_2$Ge$_2$. 

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2. Experiment

Single crystals were grown by the self-flux method [5]. Typical dimension of single crystals are 3×3×1 mm³. The crystals are confirmed to be in single phase of CeCu₂Ge₂ by X-ray powder diffraction. Those crystals are characterized by back Laue photography. Electrical resistivity was measured by using standard four-probe method in Physical Property Measurement System (PPMS). Specific heat was measured by quasi-adiabatic method in PPMS. Magnetic susceptibility is measured by SQUID magnetometer of Magnetic Property Measurement System (MPMS).

3. Results and discussions

3.1. Electrical resistivity, Specific Heats and Magnetic Susceptibility measurements

3.1.1. Electrical resistivity

Figure 1 shows temperature dependences of electrical resistivity ratio (ρ/ρ₃₀₀K) at magnetic fields where (a) H ||[100] (a-axis) and (b) H ||[001] (c-axis). Electric current is along [100] axis. We see double peaks around 100K and 6K in CeCu₂Ge₂. This behavior is typical to heavy fermion system. Black solid arrows point shoulders seen near 4K, which relate to the Néel temperature in both panels. These kinks shift to lower temperatures when magnetic field increases in a- and c-axes. We summarized field dependence of Néel temperature from electrical resistivity to Fig. 4

3.1.2. Specific Heat

Figure 2 shows temperature dependence specific heat (C) below 8K at magnetic fields where (a) H ||a-axis and (b) H ||c-axis. In these figures, we observed two anomalies in a-axis and one anomaly in c-axis. We indicate anomalies near 4K in both axes by black solid arrows. Black arrowed anomalies in Fig. 2 (a) and (b) are due to antiferromagnetic ordering. The Néel temperature is suppressed with increasing magnetic field. Although no anomaly is seen without antiferromagnetic ordering in c-axis, one small anomaly is observed around 2.5K at magnetic fields ranging from 11T to 14T for a-axis. This anomaly, indicated by blue arrow in a-axis, corresponds to small jump of magnetization for a-axis around 11T reported by Sugiyama et.al. [7]. The small jump in magnetization may be arisen by spin-flip in the a-a plane. With respect to Q-vector with (0.28, 0.28, 0.54), the magnitude of magnetic moment is 0.28×2⁻¹/²~0.2µB for a-axis before spin-flip. If magnetic field increases along a-axis, minor spin-flip may occur. The magnitude of aligned moment is 0.28µB in a-axis. The difference of moment before and after spin-flip, if there is, is calculated to be about 0.08µB. In magnetization measurement for H ||a-axis, magnitude of small jump in a range from 11T to 14T is read approximately 0.06µB from Ref. [7]. Estimated and read values are comparable to each other. Moreover Sugiyama et.al. reported that the small jump merges to saturation field when temperature increases. Thus, the additional small anomalies in specific heat are from the same magnetic origin as the small jump of magnetization. We summarize field dependence of Néel temperature and temperature at small anomaly in Fig. 4.

3.1.3. Magnetic Susceptibility

Figure 3 shows temperature dependence of magnetic susceptibilities at 10mT (red circles and lines), 5T (green circles and lines), 3.5T (blue circles and lines) and 7T (black circles and lines) for H ||a- and c-axes. In a-axis, sharp peaks near 4K were observed. As is seen in hard axis of standard antiferromagnet, magnetic susceptibility saturates in c-axis below 4K. The shoulders are caused from antiferromagnetic ordering. We summarize field dependence of Néel temperature in Fig. 4.

3.2. Magnetic Phase Diagrams

Figure 4 shows Tₙ versus H for (a) a- and (b) c-axes. Green, black and blue circles show Tₙs determined by electrical resistivity, specific heat and magnetic susceptibility measurements.
Gray open circles show an additional anomaly observed in specific heat in a range from 11T to 14T in a-axis. When we estimate Néel critical field for $H||a-$axis, we used black and blue data for fitting. Because it is rare that $H$-$T$ phase diagram is drawn in heavy fermion system, the $H$-$T$ power low scaling is not well established. In high field study in heavy fermion antiferromagnet CeIn$_3$, the power law scaling of $T_N^*$ is determined to be expressed pretty well by $T_N^* \sim [1-(H/H_c)^2]$ where $T_N^*$ is estimated Néel temperature and $H_c$ is Néel critical field [6]. We adopt this power law to estimate the critical field of CeCu$_2$Ge$_2$.

In Fig. 4, black lines are fitting curve as a function of $T_N^* = T_{N,0} [1-(H/H_c)^2]$. The $T_{N,0}$, which indicates $T_N$ at 0T, is estimated to be 4.2K for both axes. The Néel critical fields are estimated to be 35T for a-axis and 31T for c-axis. Critical field for a-axis is about 30% overestimated than saturation field (27T) reported in Ref. [7]. In c-axis, the estimated field is about twice as large as saturation field of magnetization in the paper. While the a-a plane in CeCu$_2$Ge$_2$ shows the same symmetry as that in CeIn$_3$, a-c plane in CeCu$_2$Ge$_2$ is different. Quadratic fitting is more appropriate to be used for a-a plane of CeCu$_2$Ge$_2$. Moreover, precise measurements at static high magnetic fields and low temperature could help to estimate critical fields more precisely. However, it is obvious that electronic states are in paramagnetic region above estimated critical fields in this work. We could present phase lines for a- and c- axes in CeCu$_2$Ge$_2$.

Summary
We revealed $H$-$T$ phase diagrams in single crystal of CeCu$_2$Ge$_2$. The antiferromagnetic order was gradually suppressed as magnetic field increased. By using quadratic fitting, Néel critical fields were estimated to be 35T for a-axis and 31T for c-axis. At those points, it is suggested that field induced quantum critical points emerge. These phase diagrams are powerful guide to investigate electronic states near quantum critical points.

Acknowledgments
We express many thanks to Dr. Watanabe and Professor Miyake for their fruitful discussions. Works by T. E. were performed under the auspices of ”100 Tesla spin science” and of basic research (C) by MEXT. T. E. is also acknowledges to Asahi Glass Foundation, CASIO Science Promotion Foundation and Suzuki Foundation.

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Figure 1. Electrical resistivity at various magnetic fields for (a) $H \parallel a$- and (b) $c$-axes where $J \parallel a$-axis. Antiferromagnetic ordering causes shoulders around 4K indicated by arrows. The shoulder shifts to lower temperature when magnetic field increases.
Figure 2. Low temperature specific heat for (a) $H \parallel a$- and (b) $c$-axes. Black solid arrows indicate anomalies due to antiferromagnetic transitions. Blue arrow points an additional anomaly that emerges when magnetic field applied above 11T.
Figure 3. Magnetic susceptibility for (a) $H \parallel a$- and (b) $c$-axes. The Néel orders are indicated by arrows.
Figure 4. Magnetic phase diagrams for (a) $H \parallel a$- and (b) $c$-axes. Green, black and blue circles represent Néel temperatures determined from electrical resistivity, specific heat and magnetic susceptibility, respectively. Gray circles indicate anomalies seen in specific heat.