Large heat capacity jump at the superconducting transition temperature in the non-centrosymmetric superconductor CeIrSi$_3$ under high pressure.

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Abstract. We investigated the pressure-induced superconductor CeIrSi$_3$ without inversion center under high pressure. The electrical resistivity and ac heat capacity were measured in the same run for the same sample. The critical pressure of the antiferromagnetic state was determined to be $P_c = 2.25$ GPa. The heat capacity $C_{ac}$ shows both antiferromagnetic and superconducting transitions at pressures close to $P_c$. The coexistence of the antiferromagnetism and superconductivity is discussed. The superconducting region is extended up to about 3.5 GPa. The superconducting transition temperature $T_{sc}$ shows a maximum value of 1.6 K around 2.5$-2.7$ GPa. At 2.58 GPa, a large heat capacity anomaly was observed at $T_{sc} = 1.59$ K. The jump of the heat capacity in the form of $\Delta C_{ac}/C_{ac}(T_{sc})$ is $5.7 \pm 0.1$. This is the largest observed value among all superconductors studied previously, suggesting the strong-coupling superconductivity in CeIrSi$_3$.

1. Introduction
Recently, the discovery of non centrosymmetric superconductors such as CePt$_3$Si, UIr, CeIrSi$_3$, CeRhSi$_3$, and CeCoGe$_3$ has attracted considerable interest [1, 2, 3, 4, 5]. In these compounds, two spin degenerate bands are split due to the Rashba-type spin-orbit interaction, which strongly influences the superconducting properties, particularly the pairing symmetry of the Cooper pairs. Theoretical studies suggest a mixed-type pair function with spin triplet and singlet components [6]. Many theoretical and experimental studies have been extensively conducted in order to clarify this novel type of unconventional superconductivity. In this paper, we report our high pressure study on CeIrSi$_3$, which crystalizes in the tetragonal BaNiSi$_3$-type crystal structure ($I4mm$) without inversion center.

Figure 1 (a) shows the pressure phase diagram of CeIrSi$_3$. At ambient pressure, CeIrSi$_3$ is an antiferromagnet with a Néel temperature $T_N = 5.5$ K. Under high pressure, the Néel temperature decreases with increasing pressure and disappears at the critical pressure $P_c = 2.25$ GPa. Superconductivity was found in a wide pressure region from 1.3 GPa to about 3.5 GPa. A superconducting transition temperature $T_{sc}$ shows a maximum value of 1.6 K around 2.5$-2.7$ GPa.
Figure 1. (a) Pressure phase diagram in CeIrSi$_3$. $T_N$ and $T_{sc}$, which were determined by the previous resistivity measurements, are plotted by triangles [3, 7]. $T_{sc}$ and $T_N$ values obtained by the present resistivity and ac heat capacity measurements are shown by squares and circles, respectively. Pressure dependences of (b) $\Delta C_{ac}/C_{ac}(T_{sc})$, and (c) the width of the superconducting transition in the resistivity $\Delta T_{sc}/T_{sc}$.

GPa [3]. We performed the ac heat capacity and electrical resistivity measurements on CeIrSi$_3$ in order to study the superconductivity.

2. Experiment

The single crystal of CeIrSi$_3$ was grown by the Czochralski method in a tetra-arc furnace [7]. The residual resistivity ratio RRR ($= \rho_{RT}/\rho_0$) is 120, where $\rho_{RT}$ and $\rho_0$ are the resistivity at room temperature and the residual resistivity, respectively. This indicate the high quality of the sample. The heat capacity under high pressures was measured by the ac calorimetry method [8, 9, 15]. The sample was heated up using a heater, whose power is modulated at a frequency $\omega$. The amplitude of the temperature oscillation $T_{ac}$ is expressed as the function of the heat capacity $C_{ac}$ of the sample as $T_{ac} = P_0/(\kappa + i\omega C_{ac})$. Here, $P_0$ is an average of the power. $\kappa$ is the thermal conductivity between the sample and the environment. $T_{ac}$ was measured with a AuFe/Au thermocouple (Au + 0.07 at% Fe). The contribution from the thermocouple and Au wires to the heat capacity is very small ($\sim 0.1\%$). The resistivity measurement was also carried out for the same sample by the standard four-terminal method. Two additional Au wires were attached to the edges of the sample so as to pass the electrical current for the resistivity measurements.
Figure 2. Temperature dependences of the ac heat capacity $C_{ac}$ (left side) and electrical resistivity $\rho$ (right side) at 1.99 and 2.19 GPa in CeIrSi$_3$.

3. Results and Discussions

Figure 2 shows the temperature dependences of the heat capacity $C_{ac}$ and electrical resistivity $\rho$ at 1.99 and 2.19 GPa, below the critical pressure $P_c = 2.25$ GPa. At 1.99 GPa, $C_{ac}$ shows a clear anomaly and $\rho$ shows a kink at the Néel temperature $T_N = 2.95$ K. The resistivity shows a superconducting transition at $T_{sc} = 1.02$ K. However, there is no superconducting anomaly in $C_{ac}$ at $T_{sc}$. This indicates the absence of the bulk superconductivity. The evidence of the bulk superconducting state is not obtained also at 1.31 and 1.71 GPa (data not shown). At 2.19 GPa, $C_{ac}$ shows a broad anomaly that consists of two peak structures. Those correspond to the antiferromagnetic and superconducting transitions, respectively. From the entropy balance, the Néel temperature is determined as $T_N = 1.88$ K. The peak of the heat capacity at the lower temperature side is close to the superconducting transition at $T_{sc} = 1.40$ K where $\rho$ becomes zero.

Above the antiferromagnetic critical pressure $P_c = 2.25$ GPa, both $C_{ac}$ and $\rho$ show only the superconducting transition, as shown in Fig. 3. At 2.58 GPa, the values of $T_{sc}$ obtained from the resistivity and ac heat capacity measurements are 1.62 and 1.59 K, respectively. The values are plotted in Fig. 1 (a) as squares (heat capacity) and circles (resistivity). The jump of the heat capacity in the form of $\Delta C_{ac}/C_{ac}(T_{sc})$ is $3.4 \pm 0.3$ at 2.30 GPa and $5.7 \pm 0.1$ at 2.58 GPa. Here, $\Delta C_{ac}$ is the jump of the heat capacity at $T_{ac}$ and $C_{ac}(T_{sc})$ is the value of $C_{ac}$ just above $T_{sc}$ that corresponds to $\gamma T_{sc}$. Here, $\gamma$ is the electronic specific heat coefficient. The values of $\Delta C_{ac}/C_{ac}(T_{sc})$ are significantly larger than that of the BCS theory (1.43). The large jump of the heat capacity at $T_{sc}$ was also observed in CeCoIn$_5$ and UBe$_{13}$ where the value of $\Delta C/\gamma T_{sc}$ are 4.5 and 2.7, respectively [10, 11]. In CeIrSi$_3$, the value of $5.7 \pm 0.1$ at 2.58 GPa is the largest value among all superconducting materials. The entropy balance in the superconducting state...
of 2.58 GPa is considered, as shown by the dotted line. It is found that the value of $C_{ac}/T$ is enhanced with decreasing temperature. The value of $C_{ac}/T$ at 0 K is roughly twice larger than that at $T_{sc} = 1.59$ K. The value of $\Delta C_{ac}/(\gamma T_{sc})$ is about $2.8 \pm 0.3$ if the $C_{ac}/T$ value at 0 K is used as the $\gamma$ value. This value is still larger than the BCS value. The precise pressure dependence of $\Delta C_{ac}/C_{ac}(T_{sc})$ is shown in Fig. 1(b). The strong-coupling superconductivity is realized around 2.5 GPa in CeIrSi$_3$. The increment of $\Delta C_{ac}/C_{ac}(T_{sc})$ suggests that the superconducting coupling parameter increases with increasing pressure [12, 13].

The superconducting transition width $\Delta T_{sc}/T_{sc}$ of the resistivity decreases with increasing pressure and becomes close to zero above $P_c = 2.25$ GPa as shown in Fig. 1(c). The relation between the antiferromagnetism and superconductivity is the most interesting issue to be discussed. From the present results shown in Fig. 1(c), the superconductivity and antiferromagnetism in CeIrSi$_3$ seem to be competing with each other. For the co-existence of the superconductivity and antiferromagnetism, we point out two possibilities. One possibility is that the both states co-exist only in a small pressure region close to $P_c$. The other one is that both states do not coexist and the superconductivity exists inhomogeneously in the antiferromagnetic state below $P_c$. On the basis of the latter possibility, the pressure dependence of $\Delta T_{sc}/T_{sc}$ and the gradual increase of $\Delta C_{ac}/C_{ac}(T_{sc})$ around $P_c$ can be interpreted as the increment of the superconducting volume fraction. For further investigations on the co-existence of antiferromagnetism and superconductivity, microscopic experiments such as NMR are needed.

It is impossible to obtain the absolute value of the heat capacity by the present ac heat capacity measurement. However we can estimate the relative change of the heat capacity [8, 9]. The value of $C_{ac}/T$ just above $T_{sc}$ is determined as $100 \pm 20$ mJ/K$^2$·mol at 2.58 GPa by comparison with the value of $C_{ac}$ at ambient pressure. This $\gamma$ value indicates that the moderate

Figure 3. Temperature dependences of the ac heat capacity $C_{ac}$ (left side) and electrical resistivity $\rho$ (right side) at 2.30 and 2.58 GPa in CeIrSi$_3$. 

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heavy fermion superconductivity is realized in CeIrSi$_3$. Surprisingly, this value is approximately the same as $\gamma = 120$ or 105 mJ/K$^2$·mol at ambient pressure [7].

We summarize the present experimental results as follows. The critical pressure of the antiferromagnetic state is determined to be $P_c = 2.25$ GPa. Bulk superconductivity is realized basically above $P_c$. The highest $T_{sc} = 1.6$ K and $\Delta C_{ac}/C_{ac}(T_{sc}) = 5.7 \pm 0.1$ values are obtained at the pressure region higher than $P_c$, around 2.5 – 2.7 GPa. It indicates that CeIrSi$_3$ is a strong-coupling superconductor.

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