Specific Heat Study of the Non-centrosymmetric Superconductor LaPt$_3$Si in Magnetic Fields

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Abstract. We have measured the specific heat of poly- and single-crystalline LaPt$_3$Si samples in various magnetic fields. In zero magnetic field, we observed distinct superconducting transitions at $T_c \sim 0.64$ K and 0.61 K for the poly- and single crystals, respectively. Temperature $T$ dependences of the specific heat $C$ of both samples around $T_c$ resembled each other and could be well described by an exponential equation for a conventional superconductor at low temperatures. In a magnetic field, a characteristic peak of $C/T$ appeared while the $T_c$ was considerably suppressed. These trends were pronounced for the single crystal. The transition of the polycrystal became broad above 40 Oe and a characteristic tail appeared at temperatures above the peak of $C/T$. We suggest that the tail is induced by domains that have crystallographic disorders of the non-centrosymmetry and a higher critical magnetic field than that of the bulk. These domains became superconducting at temperatures above the peak of $C/T$, and then formed the tail. Both the poly- and single-crystalline LaPt$_3$Si samples have been found to show characteristics that are in-between those of type-I and type-II superconductors.

1. Introduction
Non-centrosymmetric superconductors are attracting a great deal of interest[1]. In such systems, the conventional classification of the odd or even pair wave function for the orbital part and the singlet or triplet for the spin part is not valid anymore[2]. Thus, unconventional superconductivity with a nontrivial pair wave function is expected to appear. Two kinds of Fermi surfaces that are due to the antisymmetric spin-orbit interaction have been clearly observed for LaPt$_3$Si by the de Haas van Alphen measurements[3]. LaPt$_3$Si exhibits superconducting transition at $T_c \sim 0.6$ K without f-electron magnetism[4]. In order to investigate the characteristics of the non-centrosymmetric superconductor LaPt$_3$Si, we measured specific heat in various magnetic fields[5].

2. Experimental methods
A polycrystalline LaPt$_3$Si sample was synthesized by arc melting and a single-crystalline LaPt$_3$Si sample was grown by the Bridgman method and/or mineralization. These sample preparations were described in refs. 5 and 6. Specific heat was measured using the adiabatic heat pulse method over the temperature range of 0.1 to 0.8 K and the magnetic field range of 0 to 80 Oe. It was conducted in the heating up process after applying a magnetic field at $\sim 1$ K and cooling...
3. Results

Figures 1(a) and (b) show the \( T \) dependences of \( C/T \) of the single-crystalline sample in several selected \( H \) applied along the a- and c-axes, where \( T \), \( C \), and, \( H \) are temperature, specific heat, and, external magnetic field, respectively. In \( H = 0 \) Oe , \( C/T \) exhibited a sharp jump at \( T_c = 0.61 \) K. \( T_c \) is defined as the temperature at which the entropy is dominantly released from the sample. The value of \( \Delta C/\gamma_n T_c \) was 1.1 (BCS value = 1.43). \( C(T) \) below 0.55 K is well described by a single-exponential equation as
\[
C = A \exp(-\Delta/T) + \gamma_s T,
\]
where \( \gamma_s = 0.5 \) mJ/(K\(^2\)·mol), \( A = 67 \) mJ/(K·mol), and \( \Delta = 1.0 \) K. \( 2\Delta/k_B T_c \) is 3.2 (BCS value = 3.53). \( C \) at \( H = 0 \) Oe looks like that of the s-wave superconductor with an isotropic superconducting gap. \( T_c \) decreased with increasing \( H \) and superconductivity was almost completely suppressed above approximately 60 Oe. We should note here that a characteristic sharp peak appeared at just below the superconducting transition temperature in the magnetic fields. At first glance, the transition looked like that of a type-I superconductor in the magnetic field. This result may lead us to recall that the superconducting transition of the single-crystalline sample in the magnetic field is of the first order. However, residual \( \gamma_s \) increases with increasing \( H \). Therefore, the sample has normal areas where superconductivity is broken by external magnetic fields in spite of in a superconducting phase. In order to further our understanding, we calculated the entropy \( S \) of the single-crystalline sample with the heat capacity data. \( T \) dependences of \( S \) are shown in Figs. 1(c) and (d). \( S \) decreased rapidly below \( T_c \), especially \( H = 20 \sim 50 \) Oe. The rapid decreases correspond to the sharp peaks of \( C/T \). However, it should be noted that \( S \) decreased smoothly below \( T_c \). No discontinuity of \( S \) was observed around \( T_c \). These results may imply that the transition is not of the first order, but of the second order indicating that the crystal is a type-II superconductor.

Figure 2(a) shows the \( T \) dependences of \( C/T \) of the polycrystalline sample. The obtained parameters for the polycrystal are as follows: \( T_c = 0.64 \) K, \( \Delta C/\gamma_n T_c = 1.3 \), \( \gamma_s = 0.6 \) mJ/(K\(^2\)·mol), \( A = 79 \) mJ/(K·mol), \( \Delta = 1.1 \) K, and \( 2\Delta/k_B T_c = 3.3 \). \( T \) dependences of \( S \) of the polycrystalline sample are presented in Fig. 2(b). The behavior of \( S \) is similar to that of the single crystal. However, \( S \) decreased more smoothly below \( T_c \) than that of the single crystal, even \( H = 20 \sim 50 \) Oe. \( T \) dependence of \( C/T \) in zero magnetic field is quite similar to that of the single crystal, as shown in Fig. 3(a). The similarity of the \( T \) dependence of \( C/T \) between the
Figure 2. (a): $T$ dependences of $C/T$ of polycrystalline LaPt$_3$Si in several selected $H$. (b): $T$ dependences of $S$ derived from the data shown in fig. 2(a).

Figure 3. Comparisons of the $T$ dependences of $C/T$ between the poly- and single-crystalline samples at (a) $H = 0$ Oe, (b) $H = 40$ Oe.

single crystal and the polycrystal was preserved in the low magnetic fields. However, for $H = 40$ Oe, the large peak immediately below $T_c$ was suppressed for the polycrystal and an additional broad tail appeared above the temperature where the jump appeared, as shown in Fig. 3(b). The onset of the superconducting transition ($T_{c,\text{onset}}$) is 0.1 K higher than $T_c$ of 0.42 K. This broadening of transition for the polycrystal cannot be explained by the small anisotropy of $T_c$ observed for the single crystal. The broadening became remarkably large with increasing $H$.

We note that the above-mentioned broad tail for the polycrystal above $T_c$ appeared only in a magnetic field that was larger than approximately 40 Oe. No such tail was observed in zero or a smaller magnetic field. It is natural to consider that the polycrystal includes some local domains where the suppression of the $T_c$ by $H$ is relatively small. This interpretation may be represented in terms of the critical magnetic field $H_c$, so that the local domains have larger $H_c$ than that in the bulk of the sample. We have pointed out in our previous work that the polycrystal includes a considerable amount of crystal disorders, particularly disorders of the 'non-centrosymmetry' [6, 5]. Such disorders should considerably decrease the coherence length of the Cooper pair and increase $H_c$ in the local domains. We consider that the above-mentioned broad tail is due to this reason; that is, the broad tail appeared because there was a considerable amount of local domains having crystallographic disorders. As noted above, $S$ of the polycrystal decreased below $T_c$ more slowly than that of the single crystal. We consider that this behavior is caused by the local domains. We should also note that the $T_c$ of the polycrystal is almost the same in zero magnetic field as that of the single crystal. Therefore, the crystal disorders change directly not the $T_c$ but the $H_c$ in the local domains.

We show the $T$ dependences of the superconducting critical magnetic field $H_c(T)$ ($H$-$T$ phase diagram) of poly- and single-crystalline (a- and c-axes) LaPt$_3$Si in Fig. 4. The transition temperature $T_c$ for each $H$ is defined as the midpoint temperature between the onset and the peak of $C/T$. For the polycrystal, $T_{c,\text{onset}}$ is also plotted in the fig. 4(b). For the single crystal, there is a small anisotropy in the initial slope of the $H$-$T$ phase diagram between $H//a$ and $H//c$. Applying the conventional formula $H_c(T) = H_c(0)[1 - (T/T_c(0))^2]$, the $H_c(0)$ values were obtained as 66 and 61 Oe for $H//a$ and $H//c$ of the single crystal, respectively. $H_c(0)$
of the polycrystalline sample is slightly larger than that of the single crystal, \( H_c(0) = 71 \) Oe. The \( H-T \) phase diagram of the polycrystal illustrated by a solid line is very similar to those of the single crystal. However, in terms of \( H_c \) obtained from \( T_{c, \text{onset}} \) of the polycrystal, the superconducting phase boundary illustrated by a dashed line exhibits a steep increase above 20 Oe with decreasing \( T \). Similar \( H-T \) phase diagram was obtained from the electrical resistivity measurements, too. These properties, particularly the behavior of \( T_{c, \text{onset}} \), are consistent with the idea that the local domains have higher critical magnetic fields than that of the bulk.

4. Conclusions
We have measured the specific heat of poly- and single-crystalline LaPt\(_3\)Si samples in several magnetic fields in order to investigate the characteristics of non-centrosymmetric superconductors. For \( H = 0 \) Oe, both the polycrystal and single crystal showed almost the same \( T \) dependence of \( C/T \), which could be well described in terms of a conventional superconductor. In the presence of a magnetic field, however, \( T_c \) was considerably suppressed and a sharp peak of \( C/T \) appeared for the poly- and single-crystal. The superconducting transition of the polycrystal had a broad tail above \( H = 40 \) Oe. It is concluded that the origin of this broad tail is the domains that have crystallographic disorders of the non-centrosymmetry, in which suppression of \( T_c \) is relatively small. \( H_c(T) \) for the single crystal for \( H//a \) and \( H//c \) and the polycrystal monotonically increased with decreasing \( T \), and was well fitted by the conventional formula. \( H_c(0) \) for the single crystal exhibited a small anisotropy. Both the poly- and single-crystals show characteristics that are in-between those of type-I and type-II superconductors, although we consider that the superconducting transitions lead to the second-order phase transitions due to the disorders of the non-centrosymmetry.

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