Development of Field Angle Resolved Specific Heat Measurement System for Unconventional Superconductors

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Abstract. We developed a measurement system for field angle resolved specific heat under multiple extreme conditions at low temperature down to 50 mK, in magnetic field up to 7 T, and under high pressure up to 10 GPa. We demonstrated the performance of our developed system by measuring field angle dependence of specific heat of pressure induced unconventional superconductor CeIrSi3.

1. Introduction

Unconventional superconductors have attracted much attention in the field of strongly correlated electron systems since the discovery in the first heavy fermion superconductor CeCu2Si2 [1]. Most of them have nodes (zeros) in the gap function along certain directions. Since the location of the nodes is closely related to the pairing interaction, its clarification is crucial for understanding the pairing mechanism.

The field angle resolved specific heat and thermal conductivity measurements have been successful in determining the nodal structure because they directly probe the location of nodes in the momentum space [2][3]. The most significant effect on these techniques comes from the Doppler shift of the quasiparticle energy spectrum, $E(p) \rightarrow E(p) - v_s \cdot p$, in the circulating supercurrent $v_s$ in the mixed states of superconductors. Here, $p$ is the momentum of quasiparticle. This effect is important at the positions where the gap becomes smaller than the Doppler term ($\Delta < v_s \cdot p$). In superconductors with nodes, therefore, the Doppler shift gives rise to finite density of states (DOS) at the Fermi level and consequently the DOS contributes to the specific heat and thermal conductivity. Since the magnitude of the Doppler shift strongly depends on the relative orientation of the node and the field $H$, the effect gives rise to the oscillation of the density of states as a function of the field orientation.

To date, many unconventional superconductors have been discovered but most of them exhibit superconductivity under pressure [4]. Although it is important to reveal the gap symmetry of such pressure-induced superconductors for understanding their pairing mechanism, most of
their gap symmetry remain unsolved due to the difficulty in detecting the nodal structure under pressures. Thus, a method to determine the superconducting gap symmetry under pressures is highly desired. In this study, we developed a system for the field angle resolved specific heat measurements under pressures.

2. Experiments

Figure 1 shows the system to measure the specific heat under high pressures. The ac-calorimetry method was employed for the measurement in this study. By combining a diamond anvil cell (DAC) and vector magnets, and a $^3\text{He}-^4\text{He}$ dilution refrigerator, we achieved to measure the angular dependence of the specific heat at low temperature down to 50 mK, in magnetic field up to 7 T, and under high pressure up to $P=10$ GPa. In the following, we present details of the system.

![Figure 1.](image.png)

**Figure 1.** Schematic of the specific heat measurement system. The sample and tiny ruby are placed center of the gasket in a DAC. The DAC and transformer are installed in a $^3\text{He}-^4\text{He}$ dilution refrigerator.

2.1. **Pressure Cell**

We employed a DAC which allows us to generate the pressure up to 10 GPa. The sample is placed at the center of the gasket, between the diamond anvils as shown in Fig. 2(a) and 2(b). We used liquid Argon as a pressure-transmitting medium because of its good hydrostaticity. In fact, liquid Argon can realize better hydrostaticity than the Daphne oil which is often used in high pressure experiments [5]. By using the optical fiber in the dilution refrigerator, we measured the fluorescence spectrum [6] of tiny ruby placed next to the sample in order to estimate the pressure at low temperatures. As shown in Fig. 2(c), we observed a sharp peak in the ruby fluorescence spectrum at 4.2 K, indicating good hydrostaticity.

2.2. **Vector magnet system**

We used a vector magnet which consists of two superconducting magnets generating horizontal and vertical fields up to 7 T and 3 T, respectively. The vector magnet is installed in a dewar seating on a mechanical rotating stage to apply the magnetic fields with the arbitrary angle to the crystal axis.
In high-pressure experiments, it is difficult to realize adiabatic condition because the sample is surrounded by a pressure medium. Furthermore, the sample space is limited in a high-pressure cell, such as the DAC. Considering these limitations, ac-calorimetry is a suitable method to measure the specific heat under pressure [7]. In addition, it is sensitive to relative change of the specific heat, thus it is a powerful way to detect the specific heat oscillations associated with the nodes in the superconducting gap function.

Thermal response of a sample with respect to ac-heat applied by laser was obtained from voltage response of the thermocouple AuFe/Au attached to the sample as shown in Fig. 2(a) and 2(b). The voltage signal is amplified by a low-temperature transformer located in a dilution refrigerator and a preamplifier. Optimizing the frequency (\(f \sim 0.5 - 5\) kHz), we can obtain the specific heat \(C\) as

\[ C = \frac{Q_0}{2\pi \omega_{ac}} \sin \theta = \frac{Q_0 S}{2\pi \omega_{ac}} \sin \theta, \]

where \(Q_0\) and \(\omega\) are amplitude and angular frequency of the ac-heat, respectively, and \(\theta\) is the phase difference of the thermal response. The amplitude of thermal response \(T_{ac}\) is obtained from that of voltage response \(V_{ac}\) of the thermocouple with the Seebeck coefficients \(S\) of AuFe.
1 % above 200 mK under the magnetic field of 3 T. We must take these results into consideration in measuring field angle dependence of specific heat.

Figure 3. (a) Experimental configuration of AuFe wire to measure the angular dependence of Seebeck coefficient. (b) Field dependence of Seebeck coefficient $S/T$ at selected temperatures. (c) Polar angle $\theta$ dependence of $S(\theta)/T$ at 100 mK and 200 mK under $|\mu_0 H| = 3$ T. (d) Temperature variation of the amplitude of the normalized twofold term $S_{2\theta}$ at $|\mu_0 H| = 3$ T.

3.2. Field angle resolved specific heat of CeIrSi$_3$

To demonstrate the performance of our developed system, we measured the field angle dependence of the specific heat of the non-centrosymmetric superconductor CeIrSi$_3$, in which the superconductivity is realized only under pressure \cite{8}. In this compound, the temperature dependence of the nuclear-spin lattice relaxation rate $1/T_1$ exhibits a $T^3$ behavior without any coherence peak just below $T_c$ \cite{9}. This result suggests the presence of line nodes in the superconducting gap. However the location of node remains unresolved.

We attached the thermocouple AuFe/Au on the sample by a spot welding method, so that the AuFe wire (Au wire) is aligned along the $c$-axis as shown in Fig. 5(a). In this measurement the field is rotated along the $ac$ plane. First, we checked stability of voltage signal and phase difference as function of time at 100 mK under zero field. As shown in Fig. 4(a) and 4(b), the signals are stable within 1% showing high stability of the system. Figure 5(b) shows the angular variation of the normalized specific heat of CeIrSi$_3$ measured with rotating field along $ac$ plane at 100 mK. We can clearly observed the specific heat showing a maximum in the field parallel to the $c$-axis. This angular variation of $C$ consists of constant $C_0$ and twofold term $C_2$ as following
expression \( C(\theta) = C_0 + C_2(2\theta) = C_0(1 + C_2\cos 2\theta) \) where \( C_2 \) is the normalized amplitude of the twofold term.

The anisotropy of thermocouple can not be the origin of the twofold term with the maximum along the \( c \)-axis because it will cause a minimum along the \( c \)-axis as mentioned above. Thus we can immediately rule out the artificial origin of twofold oscillation due to the anisotropy of \( S \).

As shown in Fig. 5(b), the amplitude of the twofold term is about 2% of the total specific heat of the sample. According to the previous study of field angle resolved specific heat measurement, the amplitude of specific heat oscillations associated with the gap structure is several percent of the total specific heat [2]. This fact indicates that the resolution of our developed system is high enough to detect gap structure of unconventional superconductors at low temperature even under pressures.

Figure 4. Stability of (a) the voltage signal \( V_{ac} \) and (b) phase difference \( \theta \) at \( T = 100 \) mK under \( P = 2.6 \) GPa and zero field.

Figure 5. (a) Experimental configuration for specific heat measurements. \( \theta \) is an angle between the direction of magnetic field \( H \) and the \( c \)-axis along the \( ac \) plane. (b) \( \theta \) dependence of the normalized specific heat \( C/C_0 \) at \( T = 100 \) mK under \( -\mu_0 H = 1 \) T and \( P = 2.6 \) GPa.
4. Summary

We developed a system for field angle resolved specific heat measurements under the pressure to reveal the gap symmetry of pressure induced unconventional superconductors. To apply pressures, we used a DAC that has ability to apply the high-hydrostatic pressure up to the order of 10 GPa, and has optical windows allowed us to perform optical measurement. We employed ac-calorimetry method which enables us to perform high-resolution measurements of the specific heat.

As the result, our system achieved to measure a small angular dependence of specific heat under multiple extreme conditions, at low temperatures, in magnetic fields, and under high pressures. We demonstrated the performance of our system by measuring field angle dependence of specific heat of CeIrSi$_3$. We succeeded to detect the small oscillations of the specific heat at 100 mK under pressure of 2.6 GPa, showing high-resolution of the system.

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