Development of non-metallic diamond anvil cell and quantum oscillation measurement of CePt$_2$In$_7$ in a pulsed-magnet

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Abstract. In order to combine the extreme conditions of high pressure and magnetic field, we have developed a non-metallic diamond anvil cell to avoid Joule-heating in a pulsed-magnetic field. Although the cell is deformed with loading, a maximum pressure of $\sim$ 8.5 GPa can be applied. Quantum oscillation measurements using tunnel diode oscillator have been performed for heavy fermion antiferromagnet CePt$_2$In$_7$ in a pulsed field and at ambient pressure. The detail of the pressure cell and field-induced Fermi surface changes of CePt$_2$In$_7$ are discussed.

1. Introduction

Magnetic field and pressure are important and powerful to tune the electronic states in strongly correlated electron systems. As expanding the region of extreme conditions, novel states have appeared with big surprises. In the static magnetic field, a pressure cell made from hard and non-magnetic materials, such as CuBe-alloy having good electrical and thermal conductivity, are usually employed. Rather small field sweep rate $dB/dt$ can avoid the Joule heating due to Eddy current even below 100 mK. Situation completely changes in a pulsed-field. The heating power is proportional to the square of the $dB/dt$. The field durations of pulsed-field are usually of the order of several tens of milliseconds, inducing the huge Eddy current for conducting material. In order to avoid heating in a pulsed-field, it is very important to make a pressure cell with poor electrical conductivity and/or to be longer pulse duration. In addition, the sample space in a pulse-magnet is usually limited less than 20 mm in diameter, and thus the pressure cell should be thinner to fit.

Some groups have already developed such multiple extreme conditions, namely combination of pressure and pulsed magnetic fields [1, 2, 3, 4]. There are a number of pressure apparatus with advantage and disadvantage. Depending on material and construction, pressure range is limited. In order to change significantly the electronic state, the higher pressure is desired; e.g. magnetic quantum critical point of CePt$_2$In$_7$ studied here is 3.4 GPa. Here, we have selected an opposed-anvil-type high-pressure cell. One of the most compact ones is a diamond anvil cell.
Pressure can be simply generated in between flat surfaces of two opposed diamond anvils. Although the sample space is quite limited as usually ~0.3 mm in diameter and 0.1 mm in height for the 0.6-0.8 mm culet anvil, pressures higher than 20 GPa can be applied using a CuBe DAC. Compared to other pressure cells, the much smaller area of diamond anvil culet reduces the loading force very much. It is possible to change the material of DAC from metallic to non-metallic one with much smaller tensile strength, i.e. very compact turnbuckle-type plastic DAC [2]. Here, we have developed a diamond anvil cell made of a plastic material, polybenzimidazole (PBI). Although our DAC is much larger than that introduced in Ref. [2], maximum pressure up to ~8.5 GPa can be generated. The detail of pressure cell and its performance are discussed.

Using the non-metallic pressure cell and a pulsed-field, pressure and magnetic field dependence of electronic properties, especially for Fermi surface evolution, can be investigated. CeRhIn₅, for example, shows drastic change in FS across antiferromagnetic quantum critical point (QCP), where superconducting phase emerges [6]. In addition, magnetic field induced Fermi surface reconstruction at ambient pressure is observed below the antiferromagnetic quantum critical field [7]. It is very important to reveal how the field-induced Fermi surface reconstruction connects to pressure-induced QCP, and thus the emergence of superconductivity. We report brief results of quantum oscillation measurement on CePt₂In₇ up to 56 T and at ambient pressure through the contactless resistance measurements using a tunnel diode oscillator in a pulsed-magnet. CePt₂In₇ orders below \( T_N = 5.2 \) K antiferromagnetically at ambient pressure, and shows pressure-induced superconducting transition with the highest \( T_c = 2.1 \) K at \( P_c \approx 3.4 \) GPa [8]. Result of the quantum oscillation with the same method has been already reported [9]. They report the metamagnetic-like anomaly in the shift in resonance frequency \( \Delta f \) at \( H_m \approx 45 \) T along the c-axis, which seems to be distinct from the field-induced antiferromagnetic to the polarized paramagnetic transition. Moreover, significant Fermi surface changes occur below \( H_m \). Here, we shed light on more detail of the Fermi surface evolution as a function of magnetic field. To combine the non-metallic DAC and the pulsed-magnet, we also discuss some problems and the solutions.

\[ \text{(DAC)} [5]. \]

![Figure 1](image-url)  
**Figure 1.** (a)Photographs of DAC made of PBI (left) and CuBe-alloy (right). The scale deviation is 1 mm. They have almost same construction except for the upper diamond holder. (b) The overview of cross-sectional view of BPI-DAC. A : upper diamond holder, B : backing plate for the upper diamond anvil, C : diamond anvils, D : gasket, E : backing plate for the lower anvil, F : screw for XY-adjustment, G : piston, H : main body, I : ball bearing, and J : loading nut. Pressure can be controlled by tightening or loosening the loading nut. 

\[ \text{(DAC)} [5]. \]
2. Experimental

Our design of DAC is based on the piston-screw type one described in Ref. [10]. The overall photographs of DACs made from CuBe alloy and polybenzimidazole plastic (PBI) [11] are shown in Fig. 1(a). The dimensions of a cylinder part is approximately 20 mm in diameter and 30 mm long, which size is determined to fit into the Physical Property Measurement System of Quantum Design and pulsed-magnet installed at the Institute for Solid State Physics, the University of Tokyo [12]. In order to grip DAC while changing pressure by the loading nut, the cylinder has flats of 18 mm width. The PBI has one of the highest tensile strength among the plastic materials, ~160 MPa and is commercially available [13]. Cross-sectional drawing of PBI-DAC is illustrated in Fig. 1(b). The diamond anvils are glued to the backing plates made from CuBe and ceramic alumina for CuBe- and PBI-DAC, respectively. Using the screw, the lower diamond anvil can be adjusted to be coaxial with the upper one. The CuBe-DAC has a hemispherical support of the upper diamond anvil to parallel the surface of the anvils, in similar to Ref. [10]. The difference of PBI-DAC from CuBe one is a lack of such structure, because the support made of PBI fails with increasing pressure. Even without the tilt alignment for PBI-DAC, there is no elliptic form of the sample chamber, which occurs due to misalignment of the anvils, up to the highest pressure of 8.5 GPa in this work. This fact indicates that the parallelism between anvils is achieved through precise machining [14]. Advantages of this type of DAC are that pressure can be precisely controlled by the loading nut without hydraulic press, and the inside of sample chamber and thus pressure can be monitored through the diamond anvil window during loading.

In order to test the functionality of PBI-DAC, the usual CuBe-DAC having almost same design of PBI-DAC is compared. Diamond anvils having bevel of 0.8 mm inner and 1.0 mm outer culet with a taper angle of 12° are used in both DACs. Non-beveled diamond anvils with 0.6 mm diameter are also used for PBI-DAC. The stainless steel gasket is pre-indented to 0.1 mm from initial thickness 0.3 mm, and is drilled a hole with a half of culet diameter. The sample chamber is filled with glycerin as pressure-transmitting medium. Pressure is determined with conventional Ruby fluorescence method.

Quantum oscillation of CePt$_2$In$_7$ at ambient pressure without using pressure cell and in a pulsed-field is measured at 1.3 K through the contactless resistance measurement using tunnel diode oscillator (TDO), as described in Ref. [9, 15]. Magnetic fields up to 56 T with 36 ms duration were generated by a pulsed magnet installed in our facility [12]. Single-crystalline CePt$_2$In$_7$ of dimensions ~0.5×0.5×0.1 mm$^3$, which the shortest length is along the c-axis, is located in a 7 turns coil with 0.8 mm in diameter. The coil is a part of a TDO circuit. The TDO circuit except for the tank coil is located at room temperature, meaning that the circuit includes the ~1 m coaxial line. The magnetic field is applied along the c-axis, and thus the field inhomogeneity is negligibly small even in a pulsed magnetic field. The resonance frequency expressed as $f = \frac{1}{2\pi\sqrt{LC}}$, where $L$ is the inductance of the coil and $C$ is capacitance of a capacitor in tank circuit. Change in the penetration depth and magnetic susceptibility reflects $L$, and thus $f$. The dissipation of the LC-circuit is compensated by the negative resistance of the properly biased tunnel diode.

3. Result and Discussion

3.1. Non-metallic diamond anvil cell

Pressure efficiency using PBI- and CuBe-DAC are compared in Fig. 2. The horizontal axis is presented loading force as piston stroke length. The pressure efficiency in PBI- and CuBe-DACs are clearly different. Comparing the case of 0.8-1.0 mm anvils, the efficiency of PBI cell is quite poor. It is simply explained by the difference of the tensile strength of the materials used; the tensile strength of PBI is one order of magnitude smaller than that of CuBe. For PBI-DAC, the loading force not only squeezes the sample chamber but expands the cell body, e.g. nearly 0.2 mm expansion of the cylinder along the loading axis at a piston stroke of 400 μm.
Figure 2. Force dependence of pressure for CuBe- and PBI-DAC with different shape of diamond anvils. The horizontal axis represents the loading motion of the piston stroke. Open square and circle are the results of CuBe and PBI ones with the same beveled anvils, 0.8 mm inner culet and 1.0 mm outer culet with a taper angle of 12°. Closed circle is for PBI-DAC with non-beveled 0.6 mm culet anvil. Load is expressed as piston stroke estimated by angle of the loading nut having 1 mm pitch.

In order to suppress the expansion of the cell, we change the culet size from 0.8 mm to 0.6 mm in diameter, reducing the loading anvil area to the nearly half. The efficiency is improved drastically, as shown in Fig. 2. With the non-beveled 0.6 mm anvil, we have succeeded to apply pressures up to 8.5 GPa. The deformation of the cell along the pressure loading axis is significantly reduced. At the present, the attainable pressure is limited by the deformation of the bottom part of DAC. This part is screwed by loading nut (see Fig. 1), and the deformation of the body exceeds the the inner diameter of the jig to grip during tightening of the nut. The tensile strength of PBI is 160 MPa. The smallest area of our DAC cylinder part, i.e. window part, is ~94 mm². One can simply estimate the maximum pressure, exceeding the ultimate tensile strength, as ~ 53 GPa for the φ0.6 mm anvil. Although this estimation is too simple and optimistic, the pressure maximum could increase with modifying the design. Optimization of PBI-DAC by making the cylinder wall thicker with keeping or reducing the outer diameter is in progress.

3.2. Quantum oscillation measurement of CePt₂In₇ at ambient pressure and up to 56 T

As an example of TDO measurements, we present the results of the heavy-fermion antiferromagnet CePt₂In₇ at ambient pressure. Magnetic field dependence of the resonance frequency f at the lowest studied temperature of 1.3 K is shown in Fig. 3(a). Similarly to the previous work, the inflection point at $H_m \sim 45$ T indicated by an arrow is clearly observed [9]. $H_m$ is considered to be the loading point from the antiferromagnetic transition, because the anomaly is observed for both above and below $T_N$. The inset of Fig. 3(a) is the quantum oscillation signal subtracted polynomial background as a function of the inverse of field, corresponding to the field range between 30 and $H_m = 45$ T. Above $\sim 35$ T, i.e. $1/\mu_0 H < 0.028$ T⁻¹, the higher frequencies than that at lower field appear. It is clearly confirmed by the Fourier spectra with small field windows of 10-13 T, as shown in Fig 3(b). The prominent frequencies are indicated by the dashed lines and Greek characters used in Ref. [9]. Above 15 T, γ-branch at $\sim 1570$ T becomes
Figure 3. (a) Magnetic field dependence of the resonance frequency $f$ for the fields applied along the $c$-axis at 1.3 K. The arrow indicates $H_m \sim 45$ T. The inset represents the oscillating signal subtracted by the polynomial background. (b) Fourier spectra of the quantum oscillation of CePt$_2$In$_7$ along the $c$-axis at 1.3 K, for different field windows up to 56 T. The Greek characters indicate the prominent frequency, which are the same notations used in Ref. [9]. It is noted that the branches denoted as $\kappa$ and $\lambda$ appear well below $H_m$. The dashed lines are guide for eyes.

The peak is more pronounced with increasing field and becomes attenuated at $H_m$. The other conduction bands were not observed at low field. These results are good agreement with the previous report studied at lower temperature than this experiment [9]. Interestingly, the larger bands reported for $H \geq H_m$, which are explained by the band calculations of the localized picture [9], appear well below $H_m$. For example, the $\kappa$-branch is clearly detected for the FFT window well below $H_m$. At $H_m$, the larger bands do not seem to show any characteristics. At least, the FFT amplitudes monotonically develop as a function of magnetic fields even crossing $H_m$, which are expected for usual quantum oscillation. Important finding in this study is that the larger Fermi surface corresponding to the localized 4f-electron appears well below $H_m$.

It is important for CePt$_2$In$_7$ to reveal whether the Fermi surface reconstruction field is in AFM phase. At the present, origin of the $H_m$ may not correspond to the AFM to PM [9] and is not revealed through this experiment. In order to directly determine the phase boundary, thermodynamic experiments, such as magnetization and specific heat, are required and in progress.

Similar Fermi surface changes below $H_c$ is observed in an analogous compound CeRhIn$_5$ [7]. The authors suggested the existence of another QCP separating the AFM phases having small and large Fermi surface below $H_c$. Another possibility of the appearance of new branches at
high field is due to the magnetic breakdown through the AFM gap. When the magnetic field becomes comparable to the AFM energy gap, conduction electrons can tunnel from orbits on AFM- to PM-Fermi surface. Pressure experiments, which may control the gap, validate the magnetic breakdown scenario.

TDO experiment is very powerful to investigate the Fermi surface. This is great advantage for pressure experiments. One can escape from the difficulty of attaching electrodes to very small sample and the extrinsic stress from the electrodes. Quantum oscillation measurements using TDO under pressure and in a pulsed magnetic field are in progress. The PBI-DAC in the superfluid $^4$He is not significantly heated up in a pulsed-field with ~30 ms duration. At present, we have not succeeded to detect the quantum oscillation signals using the developed DAC installed a small coil of 0.2 mm inner diameter with several turns and in a pulsed-field. Since the $L$ of tank circuit in the pressure chamber is quite small compared to the other contribution, e.g. coaxial line between TDO circuit locating at the top of probe and sample coil, the reduced line distance between the circuit and sample induces the contribution from sample.

4. Summary
We have developed a non-metallic diamond anvil cell made of polybenzimidazole, which can introduce into a pulsed-magnet. Even with deformation of the pressure cell along the loading axis, we have succeeded to generate pressure up to 8.5 GPa. By means of quantum oscillation measurement using tunnel diode oscillator, we have detected field-evolution of Fermi surface in CePt$_2$In$_7$ at ambient pressure. In order to discuss the Fermi surface changes of CePt$_2$In$_7$ across $P_c$, quantum oscillation measurement using pressure cell developed here and a pulsed magnetic field are in progress.

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