Development of $^3$He insert for Magnetization Measurements down to $T = 0.4$ K with SQUID magnetometer

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Abstract. We have developed a 9-mm-diameter $^3$He insert for precise magnetization measurements down to $T = 0.4$ K that is attachable to a commercial superconducting quantum interference device magnetometer. The insert is made from a thin-walled stainless steel pipe with an inner diameter of 6.2 mm, which determines the maximum sample size. $^3$He gas is condensed in the pipe, which is liquefied by $^4$He gas at $T = 1.8$ K generated by the magnetometer via the heat exchanger of a Cu vacuum jacket with an outer diameter of 8.6 mm soldered to the stainless steel pipe. The temperature of the insert is decreased to $T = 0.5$ K by evacuating liquid $^3$He using a rotary pump and then to $T = 0.36$ K with a sorption pump. From the diamagnetization signal of a superconducting Al chip with a mass below 0.1 mg, the magnetization resolution with the insert is confirmed to be less than $10^{-7}$ emu. We measure the temperature dependence of magnetization down to $T = 0.5$ K in Pr$_x$La$_{1-x}$Pb$_3$, which is a good candidate for the reality of the quadrupolar Kondo effect, using the $^3$He insert. Non-Fermi liquid behavior of the nonlinear susceptibility in $\chi_3$ with a $-\ln T$ dependence is detected in the [100] and [110] directions below $T = 2.5$ K, suggesting the screening of quadrupolar moments. In contrast, $\chi_3$ in the [111] direction becomes constant below $T = 3$ K. The observed features indicate that a low-lying $\Gamma_3$ doublet plays a crucial role in the anomalous properties of Pr$_x$La$_{1-x}$Pb$_3$.

1. Introduction
Since the multichannel Kondo effect has been shown mathematically to give rise to a breakdown of the Fermi Liquid (FL) state and lead to Non-Fermi liquid (NFL) ground state [1, 2], experimental observation of the multichannel Kondo effect is one of the central issues in strongly correlated electron physics. Over the past two decades, a great deal of studies have been done to demonstrate the existence of the two-channel Kondo effect (TCKE) in many systems, such as $f$-electron compounds, quantum dots, and two-level systems.

It is well known that the quadrupolar Kondo effect (QKE) proposed by Cox is a realistic model of the TCKE [3]. He showed that QKE can be realized in a $\Gamma_3$ doublet with a quadrupolar moment in the crystal-electric-field (CEF) ground state [3, 4]. In real systems, the QKE is expected to appear in U$^{4+}$ ions with the $5f^2$ configuration or in Pr$^{3+}$ ions with the $4f^2$ configuration under cubic symmetry. Recently, much attention has been devoted to the study of the characteristic features of Pr-based compounds, which are discussed in connection with the QKE [4, 5, 6, 7, 8, 9, 10, 11], because Pr compounds are ideal systems for studying correlated electron behavior within the $f^2$ configuration due to a more localized character of the $4f$ orbital.
On the other hand, it is difficult to determine the uranium 5f configuration experimentally due to a mixed valence character the 5f orbital. This prevents us from examining the f-electron properties as a single-ion effect.

We have studied the low-temperature properties of Pr$_x$La$_{1-x}$Pb$_3$ with a $\Gamma_3$ doublet in the CEF ground state for various Pr concentrations [12, 13, 14, 15]. The observed features indicate that Pr$_x$La$_{1-x}$Pb$_3$ is a good candidate for clarifying the reality of the QKE. To obtain the direct evidence of QKE in Pr$_x$La$_{1-x}$Pb$_3$, we focus on the nonlinear susceptibility $\chi_3$, which reflects the fluctuation of the quadrupolar moment [16, 17]. $\chi_3$ is defined as the third-order term in the magnetization and expressed by

$$M = \chi_1 H + (1/3!)\chi_3 H^3 + \cdots,$$

where $M$ is the magnetization and $H$ is the applied field. It is significant that $\chi_3$ corresponds to the quadrupolar susceptibility in the Pr $\Gamma_3$ doublet ground-state system.

The precise magnetization measurements below $T = 2$ K are crucial for the investigation of $\chi_3$, because Pr$_x$La$_{1-x}$Pb$_3$ is the dilute system with nonmagnetic ground state. From these reasons, we develop a 9-mm-diameter $^3$He insert attachable to a commercial MPMS superconducting quantum interference device (SQUID) magnetometer from Quantum Design. In this paper, we show the specification and performance of the insert [18].

2. Design of $^3$He insert

We designed a $^3$He insert with the following specifications: (1) The magnetometer resolution with the insert is retained at the same level as that without the insert. (2) The insert is easily attached to and removed from the 9-mm-diameter sample space within the magnetometer, and (3) the maximum sample diameter is $\sim 6$ mm. A schematic of the $^3$He insert is shown in Fig. 1(a). The insert is fitted to the magnetometer through the actuator shoe used for moving the sample rod up and down. The insert is mounted with an airlock plug by turning the slide seal clumps (see Fig. 2). Since the pressure in the $^3$He insert is kept to be much lower than 0.1 MPa in the measurements, the release of $^3$He gas is prevented by fitting a vacuum seal with a double O-ring arrangement on top of the $^3$He insert. This enables only the sample rod to move during measurements (Fig. 2). Note that the insert is attachable to other magnetometers or variable temperature inserts by changing the design of the attachment devices.

The insert is made from a thin-walled stainless steel pipe with an inner diameter of 6.2 mm and an outer diameter of 6.5 mm, which is used for bringing $^3$He gas. The length of the insert below the airlock plug is $\sim 1,300$ mm. We also attach a stainless capillary with an inner diameter of 0.7 mm and an outer diameter of 1.1 mm for evacuating the vacuum jacket. They are linked with two KF16 flanges set at the top of the insert to connect with a $^3$He gas handling system and a vacuum pump.

Figure 1(b) presents an enlargement of the low-temperature portion of the insert. A Cu foil with a thickness of 0.05 mm and a length of $\sim 150$ mm is wound at the lower end of the stainless steel pipe, where the liquid $^3$He is condensed, to increase the thermal conductivity along the pipe. The vacuum jacket, made from a copper pipe with an outer diameter of 8.6 mm and a length of $\sim 250$ mm, is soldered to the stainless steel pipe, which is also used as the heat exchanger between $^3$He and $^4$He gases at $T \sim 1.8$ K generated by the magnetometer. A RuO$_2$ thermometer for the $^3$He pot is attached at the bottom of the pipe. Manganin wires are threaded in the capillary tube and connected with a thermometer after winding around the pipe to prevent thermal conduction of heat from room temperature.

To lower the temperature below $T = 0.5$ K, we installed a charcoal sorption pump with 0.43 g of charcoal contained in a Cu pot set above the sample holder. Manganin wires for heating the charcoal are wound at the top of the pot; temperature is measured here using a Au-Fe thermometer attached to the Cu pot.
3. Performance

Figure 1. (a) Schematic of the $^3$He insert, which is made from a thin-walled stainless steel pipe with an outer diameter of 6.5 mm. The stainless steel capillary tube with an outer diameter of 1.1 mm is used for evacuating the vacuum jacket and threading the manganin wires used as a thermometer. Two ports in the KF 16 flanges at the top of the insert are used to supply $^3$He gas inside the pipe and pump the vacuum jacket. (b) Schematic of the inside of the vacuum jacket. The vacuum jacket, made from a copper pipe with an outer diameter of 8.6 mm and a length of $\sim$ 250 mm, is soldered to the stainless steel pipe. $^3$He gas condenses at the bottom of the stainless steel pipe, where a Cu foil with a thickness of 0.05 mm and a length of $\sim$ 150 mm is wound to increase the thermal conductivity along the pipe. A RuO$_2$ thermometer is placed at the bottom of the stainless steel pipe.

Figure 2. Schematic showing the insert attached to the MPMS SQUID magnetometer by clamping with the airlock plug. In measurements, only the sample rod locked at the actuator shoe is moved by the stepping motor of the MPMS magnetometer. The actuator shoe does not touch either the two stainless steel pipes for the $^3$He gas or vacuum lines during measurements.

The $^3$He insert is cooled to low temperatures using the same procedure as cooling the sample in the magnetometer without the insert. Before attaching the insert, the vacuum jacket is evacuated. After warming the sample space of the magnetometer to 320 K, then venting the airlock and sample spaces, the $^3$He insert with the sample mounted is inserted into the magnetometer. Next, the airlock and sample spaces of the magnetometer are purged. After this sequence is completed, the $^3$He gas line of the insert is evacuated. A small amount of $^3$He...
gas is then introduced into the gas line. The temperature of the insert is lowered by lowering the temperature of the MPMS magnetometer. It takes only 40–50 min. to cool the insert to liquid $^3$He temperature. The rest of $^3$He gas is added to the insert when the magnetometer temperature reaches its lowest temperature $T \sim 1.8$ K.

Using the same procedure with the $^3$He cryostat, the temperature is further lowered below 2 K. After liquefying about 2 liter of $^3$He gas over a period of about 1 h, $^4$He gas is evacuated using a rotary pump. In this way, it is possible to maintain low temperatures for 5–10 h. depending on the temperature. Figure 5 presents a typical evolution of the insert temperature during cooling as measured by a thermometer attached to the sample holder.

We measure the temperature dependence of the magnetization for cerium manganite nitride (CMN). The temperature is controlled by adjusting the pumping speed of $^3$He gas with the valve control in the gas handling. Once the temperature is established, magnetization is measured by moving the sample up and down, raising the sample temperature. The temperature increase becomes notable below $T \sim 0.65$ K. For instance, the initial temperature of $T = 0.36$ K increases to $T \sim 0.42$ K after the scan. From the fact that the increase is independent of the magnitude of the magnetic field, it is reasonable to consider that the increase is caused by friction heating between the sample holder and the inside of the stainless steel pipe. When we take the average of the initial and final temperatures as the sample temperature of CMN, the temperature dependence of the magnetization is well reproduced by the Curie law. We speculate that such an enhanced heating can be suppressed by improving the sample holder.

![Figure 3](image_url)

**Figure 3.** Temperature dependence of magnetization of an Al chip in a nominal magnetic field of $H_{nom} = 2.5$ Oe. The diamagnetization following the superconducting transition appears at $T_C \sim 1.17$ K. The inset shows the SQUID output signals at $T = 1.10$ and 1.20 K. The solid lines indicate fittings from the MPMS magnetometer data. (b) Temperature dependence of magnetization of an Al chip under the nominal magnetic fields of $H_{nom} = 2.5$ and 4 Oe.

To assess the magnetization resolution with the $^3$He insert, we measured the diamagnetization signal of Al below the superconducting transition temperature $T_C$. An Al chip with a mass below 0.1 mg is mounted in a straw, which is attached to the sample rod. The inset in Fig. 3(a) shows the scanning signals above and below $T_C$. In the low field region below $H \sim 10$ Oe, the homogeneity of the magnetic field in the scanning region is lowered because of the remanent field in the superconducting magnet providing the maximum field of $H = 70$ kOe. Thus, we reduced the scanning range for the sample from 40 to 25 mm. Indeed, the signal is almost symmetric with respect to the center of the SQUID coil and is fitted well with the analytical curve of the
Figure 4. (a) Temperature dependences of $\chi_3$ in the three principal directions. The dashed lines are theoretical calculations in the [100] and [111] directions for the isolated-ion model, where quadrupolar moments are not screened by conduction electrons, based on the CEF level scheme given in the text. (b) Anisotropic part of the normalized nonlinear susceptibility, $\Delta \chi_3^H \equiv (\chi_3^H - \chi_3^{[111]}) / \chi_3^{[111]}$, for the [100] and [110] directions, where the theoretical values of $\chi_3^{[111]}$ are used. The solid lines are visual guides.

Figure 3(a) gives the temperature dependence of magnetization at the nominal magnetic field of $H_{\text{nom}} = 2.5$ Oe generated by the MPMS control system. The diamagnetization signal increases rapidly below $T = 1.17$ K. The scatter of the data is within $\pm 5 \times 10^{-8}$ emu, suggesting that the resolution of the magnetometer with the $^3$He insert is at least $\pm 10^{-7}$ emu. From the temperature-dependent magnetization curves for $H_{\text{nom}} = 2.5$ and 4 Oe shown in Fig. 3(b), the transition temperature is seen to be slightly reduced and the magnitude of the diamagnetization signal is observed to increase. The details of the insert are described in ref. [18].

We show the temperature dependences of $\chi_3$ along the three principal axes, [100], [110], and [111] in Fig. 4(a). Although the $\chi_3$ values are almost the same in the three directions at high temperatures, the anisotropy increases with decreasing temperature as expected from the theory [16]. The $\chi_3$ values in the [100] and [110] directions begin to increase below $T \approx 3$ K, while that in the [111] direction decreases gradually and becomes almost constant below $T \approx 3$ K. We check the validity of the present measurements by comparing $\chi_3$ in the [111] direction to that for the isolated-ion model. Since the QKE does not contribute to $\chi_3$ in the [111] direction, the temperature dependence of $\chi_3$ is expected to be the same as that for the isolated-ion model. We plot the estimate in Fig. 3(a), which is obtained from the energy level scheme of $\Gamma_3(0$ K)$-\Gamma_4(14.7$ K)$-\Gamma_5(21.8$ K)$-\Gamma_4(35.2$ K) with the CEF parameter $X = 0.1$ and $W = -0.277$ K, according to the definition by Lea et al. [19]. As shown in the plot, $\chi_3$ in the [111] direction is well reproduced by the theoretical values. Furthermore, the magnetic susceptibility above $T = 5$ K is well reproduced by the above CEF parameter [17].

To see the anisotropic component of the nonlinear susceptibility, we plot the results in the form

$$\Delta \chi_3^H \equiv \frac{\chi_3^H - \chi_3^{[111]}}{\chi_3^{[111]}},$$

where $\chi_3^H$ is the nonlinear susceptibility in the [100] and [110] directions. The plots are depicted...
in Fig. 4(b). The $\chi_3$ in the [100] and [110] directions clearly shows NFL behavior with a $-\ln T$ dependence below $T \approx 2.5$ K, suggesting the screening of quadrupolar moments by conduction electrons. The details on $\chi_3$ in Pr$_x$Lax-Pb$_3$ are presented in ref. [17].

4. Conclusion
We have developed a 9-mm-diameter $^3$He insert for precise magnetization measurements down to $T = 0.4$ K that is attachable to a commercial superconducting quantum interference device magnetometer. The insert is made from a thin-walled stainless steel pipe with an inner diameter of 6.2 mm, which determines the maximum sample size. $^3$He gas is condensed in the pipe, which is liquefied by $^4$He gas at $T = 1.8$ K generated by the magnetometer via the heat exchanger of a Cu vacuum jacket with an outer diameter of 8.6 mm soldered to the stainless steel pipe. The temperature of the insert is decreased to $T = 0.5$ K by evacuating liquid $^3$He using a rotary pump and then to $T = 0.36$ K with a sorption pump. From the diamagnetization signal of a superconducting Al chip with a mass below 0.1 mg, the magnetization resolution with the insert is confirmed to be less than $10^{-7}$ emu. We measure the temperature dependence of magnetization down to $T = 0.5$ K in Pr$_{0.05}$La$_{0.95}$Pb$_3$, using the $^3$He insert. Non-Fermi liquid behavior of the nonlinear susceptibility in $\chi_3$ with a $-\ln T$ dependence is detected in the [100] and [110] directions below $T = 2.5$ K, suggesting the screening of quadrupolar moments by conduction electrons.

References
[1] Nozières P and Blandin A 1980 J. Phys. 41 193
[2] Sacramento P D and Schlottmann P 1991 Phys. Rev. B 43 13294
[3] Cox D L 1987 Phys. Rev. Lett. 59 1240
[4] Koga M and Shiba H 1995 J. Phys. Soc. Jpn. 64 4345
[5] Yatskar A, Beyermann W P, Movshovich R and Canfield P C 1996 Phys. Rev. Lett. 77 3637
[6] Onimaru T, Sakakibara T, Aso N, Yoshizawa H, Suzuki H S and Takeuchi T 2005 Phys. Rev. Lett. 94 197201
[7] Kawae T, Li C-S, Yoshida Y, Takeda K, Asano T and Kitai T 2005 J. Phys. Soc. Jpn. 74 2332
[8] Suzuki O, Suzuki H S, Kitazawa H, Kido G, Ueno T, Yamaguchi T, Nemoto Y and Goto T 2006 J. Phys. Soc. Jpn. 75 013704
[9] Tanida H, Suzuki H S, Takagi S, Onodera H and Tanigaki K 2006 J. Phys. Soc. Jpn. 75 073705
[10] Sakai A and Nakatsuji S 2011 J. Phys. Soc. Jpn. 80 063701
[11] Onimaru T, Matsumoto K T, Inoue Y F, Ueno K, Sakakibara T, Karaki Y, Kubota M and Takabatake T 2011 Phys. Rev. Lett. 106 177001
[12] Sato Y, Morodomi H, Ienaga K, Inagaki Y, Kawae T, Suzuki H S and Onimaru T 2010 J. Phys. Soc. Jpn. 79 093708
[13] Kawae T, Shimogai M, Mito M, Takeda K, Ishii H and Kitai T 2001 Phys. Rev. B 65 012409
[14] Kawae T, Yamamoto T, Yurue K, Tateiwa N, Takeda K and Kitai T 2003 J. Phys. Soc. Jpn. 72 2141
[15] Kawae T, Kinoshita K, Nakie Y, Tateiwa N, Takeda K, Suzuki H S and Kitai T 2006 Phys. Rev. Lett. 96 027210
[16] Ramirez A P, Chandra P, Coleman P, Fisk Z, Smith J L and Ott H R 1994 Phys. Rev. Lett. 73 3018
[17] Kawae T, Koga M, Sato Y, Makiyama S, Inagaki Y, Tateiwa N, Fujiwara T, Suzuki H S and Kitai T 2013 J. Phys. Soc. Jpn. 82 073701
[18] Sato Y, Makiyama S, Sakamoto Y, Hasuo T, Inagaki Y, Fujiwara T, Suzuki H S, Matsubayashi K, Uwatoko Y and Kawae T 2013 Jpn. J. Appl. Phys. 52 106702
[19] Lea K R, Leask M J M and Wolf W P 1962 Phys. Chem. Solids. 23 1381