Electrical Resistivity and Thermal Expansion Measurements of URu₂Si₂ under Pressure

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We carried out simultaneous measurements of electrical resistivity and thermal expansion of the heavy-fermion compound URu₂Si₂ under pressure using a single crystal. We observed a phase transition anomaly between hidden (HO) and antiferromagnetic (AFM) ordered states at T_M in the temperature dependence of both measurements. For the electrical resistivity, the anomaly at T_M was very small compared with the distinct hump anomaly at the phase transition temperature T_0 between the paramagnetic state (PM) and HO, and exhibited only a slight increase and decrease for the I // a-axis and c-axis, respectively. We estimated each excitation gap of HO, Δ_HO, and AFM, Δ_AFM, from the temperature dependence of electrical resistivity: Δ_HO and Δ_AFM have different pressure dependences from each other. On the other hand, the temperature dependence of thermal expansion exhibited a small anomaly at T_0 and a large anomaly at T_M. The pressure dependence of the phase boundaries of T_0 and T_M indicates that there is no critical end point and the two phase boundaries meet at the critical point.

KEYWORDS: URu₂Si₂, hidden order, antiferromagnetism, heavy-fermion superconductor, thermal expansion, electrical resistivity

URu₂Si₂ is a heavy-fermion superconductor with a superconducting transition temperature T_c ∼ 1.5 K.¹ Furthermore, the compound undergoes a successive phase transition at T_0 ∼ 17.5 K. At this temperature, specific heat exhibits a sharp and large jump of ∼ 0.3 J/(K²·mol). Additionally, there appears a clear kink and a clear hump in the curves of magnetic susceptibility and electrical resistivity plotted as a function of temperature T, respectively.² These features show a weak sample dependence. On the other hand, in many neutron diffraction experiments, only a tiny staggered moment of about ∼ 0.03 μ_B/U was observed, and there were strong sample dependences on its magnitude and onset.³⁻⁶ These results have led to many speculations that the true order parameter is not the weak magnetic dipole moment, but another unknown symmetry such as quadrupoles.

Amitsuka et al. examined the T dependence of dc magnetization under pressure, and observed the anomaly at the phase transition from HO to AFM.¹³,¹⁴ The authors argued the presence of the bicritical point on the basis of the P dependence of the parasitic ferromagnetic anomaly T_{FM}(P) of ∼ 35 K for P = 0. Moreover, it was revealed that the superconductivity of this system coexists only in HO but not in AFM. However, Bourdarot et al. argued the presence of the critical end point of the T(M)(P) on the basis of their neutron diffraction measurements.¹⁵ Recently, Hasinger et al. observed the anomaly at the phase transition from HO to AFM in the electrical resistivity and specific heat, and showed the P dependences of these phase boundaries that met at the critical point.¹⁶ Whether or not T_M(P) and T_0(P) meet is important information concerning the symmetry of the order parameter of the HO state.¹⁷ However past experiments are insufficient to conclude whether or not T_M(P) and T_0(P) meet. The thermal expansion measurement was sensitive to T_M but it yielded no details of T_0, particularly at around P_c. A smaller anomaly was smeared out within a predominant anomaly when T_M approached T_0. On the other hand, electrical resistivity and specific heat were sensitive to only T_0, that is, these measurements yielded no details of T_M at around P_c. In this work, we carried out electrical resistivity and thermal expansion measurements in parallel. In the entire P range, even at around P_c, T_M and T_0 could be accurately determined from the data of thermal expansion and electrical resistivity, respectively, which were measured at the same time under pressure to avoid the ambiguity between the two measurements.

We first synthesized a polycrystalline material by melting a stoichiometric amount of the constituent elements natural U, Ru, and Si, which had purities of 99.9 %, 99.99 %, and 99.9999 %. Then we grew a single crystal...
by the Czochralski pulling method from the polycrystalline material in high-purity argon atmosphere using a laboratory-made tri-arc furnace. The sample for measurements was cut from the as-grown single crystal. The size of the sample was about $\sim 2\times2\times2$ mm$^3$. We chose a sample that showed a distinct anomaly at $T_M$ in order to determine $T_0(P)$ and $T_M(P)$, because the anomaly of $T_M$ exhibits a strong sample dependence, whereas the anomaly of $T_0$ has only a weak dependence. We measured electrical resistivity and thermal expansion by the conventional dc 4-terminal method and the strain gauge technique with a copper block as a dummy sample, respectively. The measurements were performed using a $^4$He cryostat down to 4 K. Pressure was generated using a copper-beryllium clamp-type cylinder with a piston made of tungsten carbide. The pressure-transmitting medium was Dafune7373. We determined pressure by measuring the superconducting transition temperature of indium. The electrical resistivity and thermal expansion measurements were carried out concurrently to eliminate measurement ambiguities in pressure and temperature.

Figure 1(a) shows the $T$ dependence of the thermal expansion coefficient for the $a$-axis, $\alpha_a$, at different pressures. A mean-field-like discontinuous anomaly corresponding to the phase transition at $T_0$ between the paramagnetic state (PM) and HO was observed at 17 K at 0 GPa, and it could be observed only below 0.83 GPa within the accuracy of the $\alpha_a$ measurement. An anomaly that was identified as the phase transition between the HO and AFM appeared at 0.57 GPa and $T_M \sim 13.5$ K. This anomaly was greater than the anomaly at $T_0$. It is clear that $T_M$ shifts to higher temperatures accompanied by a change in the shape of the anomaly, and finally becomes a large mean-field-like anomaly. These behaviors are the same as those described in our previous paper.

Next, Fig. 1(b) shows the $T$ dependence of the derivative of the resistivity for the $a$-axis, $d\rho_a/dT$, at different pressures. There is a sharp and deep dip at $T_0$ in $d\rho_a/dT(T)$. It shifts to higher temperatures, remaining sharp and deep, throughout the entire range of pressure. On the other hand, a small convex-upward anomaly is also seen at $T_M$ in $d\rho_a/dT(T)$ above 0.57 GPa. This anomaly was too small, in comparison with the deep dip, to observe the $P$ dependence of $T_M$. When $T_M$ was close to $T_0$, it was smeared out in the dip. The measurements of $\alpha_a$ and $d\rho_a/dT$ were carried out at the same time. Therefore, the $P$ and $T$ of $\alpha_a$ are identical to those of $d\rho_a/dT$, although there may be a slight error in absolute value. $T_0$ and $T_M$ were defined as the maximum temperature in the data of both $\alpha_a$ and $d\rho_a/dT$, and are indicated by marks in Figs. 1(a) and 1(b). The error ranges of $T_0$ and $T_M$ were determined from the full width at half-maximum of the peak or the full width at half-minimum of the dip. The $P$ dependences of $T_0$ and $T_M$ are plotted in Fig. 4(a). These results are described below.

Figures 2(a)-2(c) represent the $T$ dependences of $\alpha_a$ and $d\rho_a/dT$ at 0, 0.57, 0.73, 0.83, and 1.09 GPa. At ambient pressure, we should observe only the phase transition between PM and HO. Our $\alpha_a$ and $d\rho_a/dT$ data certainly showed the anomaly at the same temperature $T_0$. Next, in Fig. 2(b), we could observe the $P$ dependence of the anomalies of $T_0$ and $T_M$ from 0.57 to 0.83 GPa: $\alpha_a$ is sensitive to the transition at $T_M$, whereas $d\rho_a/dT$ is sensitive to the transition at $T_0$. Moreover, $d\rho_a/dT(T)$ evidently shows a convex-upward anomaly at $T_M$. When $d\rho_a/dT(T)$ has a convex-upward anomaly, $\rho_a(T)$ must have a steplike anomaly at $T_M$. We show $\rho_a(T)$ in Fig. 3; these results are described below. At 1.09 GPa, in Fig. 2(c), we observed a large peak of $\alpha_a$ and a deep dip of $d\rho_a/dT$ at the same temperature. The large peak of $\alpha_a$ corresponds to $T_M$ and the deep dip of $d\rho_a/dT$ corresponds to $T_0$. Therefore, $T_0$ and $T_M$ have the same value at 1.09 GPa. The phase boundaries of $T_0$ and $T_M$ met and constructed the phase transition between PM and AFM at the temperature $T_N$. Moreover, the anomaly of the large peak of $\alpha_a$ was retained upto 1.69 GPa in our previous study, and the anomaly of $\rho(T)$ was retained upto over $\sim 2$ GPa in previous studies. We consider that the anomalies observed at $T_0$ and $T_M$ occur at the same temperature as the anomaly of $T_N$ at pressures higher than $P_c$. Figure 2(d) shows the $T$ de-
the expansion coefficient for the c-axis, $\alpha_c$, and the derivative of the resistivity for the c-axis, $d\rho_c/dT$, at 0.37, 0.55, and 0.58 GPa. There is a large anisotropy between $\rho_a$ and $\rho_c$; $\rho_c$ is one-tenth of $\rho_a$. Therefore, it was difficult to obtain the absolute value of $\rho_c$; consequently, we show $d\rho_c/dT$ and $\alpha_c$ in arbitrary units. We also observed anomalies at $T_M$ in $\alpha_c$ and $d\rho_c/dT$, although these anomalies were small. These small anomalies were consistent with the previous results. Here, note that the anomaly in $d\rho_c/dT$ at $T_M$ is convex downward. The anomaly in $\rho$ at $T_M$ clearly exhibits anisotropy.

Figure 3 shows the $T$ dependence of $\rho_a$ at 0.10, 0.37, 0.57, 0.73, and 1.09 GPa. Previous $\rho(T)$ results for URu$_2$Si$_2$ could be fitted by the sum of the $T^2$ term and the $\exp(-\Delta/T)$ term: $\rho = \rho_0 + AT^2 + B\frac{T}{\Delta}(1 + 2\frac{T}{\Delta})\exp(-\frac{\Delta}{T})$. (1)

We attempted to fit this equation to our $\rho_a$ data. $\rho_a$ was expected to be fitted easier than $\rho_c$ for the excitation feature because of the strong $T$ dependence of $\rho_a$. Our $\rho_a(T)$ data below 0.37 GPa could be fitted well with eq. (1). In this pressure region, the HO phase exists below $T_0$. That is, the $\rho_a(T)$ of the HO region could be fitted with eq. (1). Moreover, the $\rho_a(T)$ data at 1.09 GPa could also be fitted well. At this pressure, the phase boundaries of $T_0(P)$ and $T_M(P)$ meet; therefore, the AFM phase exists below the anomaly at $T_M$. $\rho_a(T)$ in the AFM region could also be fitted with eq. (1) using the appropriate parameters for the AFM state. However, the $\rho_a(T)$ data from 0.57 to 0.87 GPa show a steplike anomaly at $T_M$. When there is a steplike anomaly, we must fit separately at $T_M$. The lower and higher parts of data were fitted with eq. (1) using the appropriate parameters for HO and AFM states, respectively. It is difficult to discuss the $T$ dependence of $\rho_a$ at around $T_M$ because of the inevitable phase separation of first-order transition. We must estimate these parameters without $\rho_a(T)$ data at around $T_M$. The $P$ dependencies of the excitation gaps, $\Delta_{HO}$ and $\Delta_{AFM}$, and the coefficients of the $T^2$ contribution, $A_{HO}$ and $A_{AFM}$, are plotted in Figs. 4(b) and 4(c), respectively. In our estimation, there were small differences in the accuracy between $\rho_{a,HO}$ and $\rho_{a,AFM}$ and between $A_{HO}$ and $A_{AFM}$ for $\rho_a(T)$ at 0.57 and 0.73 GPa, respectively. Therefore, we show electrical resistivity data without the residual resistivity and Fermi liquid contribution $\rho_0 - \rho_0 - AT^2$ on a logarithmic scale as a function of $1/T$ in the inset of Fig. 3. The decreasing rates of $\log(\rho_a - \rho_0 - AT^2)$ at 0.10,
0.37, and 1.09 GPa are almost constant within the plotted range, although these rates become slightly slower owing to the coefficient of the exponential term of $\rho_a$. The decreasing rate of $\log(\rho_a/\rho_0 - AT^2)$ vs 1/T roughly corresponds to $\Delta_{\text{HO}}$ or $\Delta_{\text{AFM}}$. When the AFM phase appears, namely, a broken line turns into a full line, the rate becomes more rapid, indicating that $\Delta_{\text{AFM}} \neq \Delta_{\text{HO}}$. It is natural to have different excitation gaps for different ordered states. It is a interesting that eq. (1) well fits not only the $\rho_a(T)$ of the AFM phase but also that of the HO phase. This result may provide a clue to the order parameter of the HO phase.

In Fig. 4(a), we summarize the P-T phase diagram of URu$_2$Si$_2$ using data from $\rho_a(T)$ and $\alpha_a(T)$ measurements; it includes details about $T_0$ and $T_M$ at around $P_c$. It was experimentally verified that $T_M$ meets $T_0$ at the critical point, where $P_c$ is from 1.04 to 1.09 GPa in this sample. We took measurements only below 1.09 GPa because of the limit of our pressure cell. In Figs. 4(b) and 4(c), we plot the P dependences of $\Delta_{\text{HO}}$, $\Delta_{\text{AFM}}$, $A_{\text{HO}}$, and $A_{\text{AFM}}$, respectively. $\Delta_{\text{HO}}(P)$ and $\Delta_{\text{AFM}}(P)$ have different P dependences from each other. However, $\Delta_{\text{HO}}(P)$ and $\Delta_{\text{AFM}}(P)$ also increase gradually with increasing $P$, and also seem to show linear P dependences. These extrapolated lines seem to cross at around $P_c$. $A_{\text{HO}}(P)$ and $A_{\text{AFM}}(P)$ also decrease gradually with increasing $P$. Although the difference in P dependence between $A_{\text{HO}}(P)$ and $A_{\text{AFM}}(P)$ cannot be denied, the differences between $A_{\text{HO}}$ and $A_{\text{AFM}}$ are negligible in terms of the accuracy of the measurements and estimations.

In conclusion, our results include two significant points to be emphasized. The first one is that the HO and AFM phases are completely separated by the boundary of $T_M$, which seems to be a first-order transition. The second one is that each of the HO and AFM phases also has an excitation gap: $\Delta_{\text{HO}}(P)$ was not identical to $\Delta_{\text{AFM}}(P)$. These two results clearly indicate that the HO state is not identical to the AFM state.

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