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Thermodynamic aspects of heavy fermion compounds under high pressure

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Abstract

High pressure studies on the thermal expansion and electrical resistance of the heavy fermion U-compound have been carried out to examine the electronic state under volume contraction. Clear anomalies around 17 K were found in the temperature dependence of the thermal expansion coefficient and electrical resistivity due to antiferromagnetic ordering. A T^2 dependence in electrical resistivity and T-linear term in the thermal expansion coefficient were obtained at low temperature. These results are analyzed on the basis of thermodynamic relations and discussed by using volume-dependent Grüneisen parameter in comparison with the results for other concentrated Kondo compounds. © 1998 Published by Elsevier Science S.A.

Keywords: Heavy fermion; Electrical resistance; Thermal expansion; High pressure; Analysis

1. Introduction

It is well known that the physical properties of concentrated Kondo (CK) compounds are extremely sensitive to a change in pressure and hence in volume. This is due to unstable f-electrons which hybridize with a conduction band. The main effect of pressure is to vary the strength of the hybridization, and it appears clearly in physical properties through changes of the characteristic energy scale, so-called T_K. We have shown that the electronic state of CK compounds is strikingly changed by pressure, up to now, through results of high pressure measurements of the electrical resistivity and the thermal expansion [1]. According to a quantitative analysis for the volume dependence of T_K, it has been revealed that instability of the f-electron is reflected in a large value of the Grüneisen parameter of T_K, which typically reaches on the order of 100 in heavy fermion (HF) compounds and around 10 in intermediate valence (IV) compounds [2–4]. Ce- or U-based HF compounds show a pressure-induced crossover from a low-T_K HF state to a high-T_K IV state accompanied by an enhancement of T_K and decrease in parameter [1,5].

In this paper we review results of electrical resistivity and thermal expansion measurements on HF compounds at high pressure, in which the U-based HF compound URu_2Si_2 is described as an example. Thermodynamic analysis associated with the volume dependence of T_K is carried out based on the experimental results for URu_2Si_2 and is compared with other CK compounds.

2. Brief survey of the experimental results for URu_2Si_2

URu_2Si_2 is classified as a HF compound [6–8] that has a moderately large electronic specific heat coefficient γ of 65 mJ/mol·K^2. The specific characteristic of this substance is the coexistence of antiferromagnetic order and superconductivity. Antiferromagnetic order is reported to occur below the Néel temperature $T_N = 17.5$ K and to be of the spin-density-wave type. Superconductivity is found below T_c of around 1 K.

Fig. 1 shows the temperature-dependent part of the electrical resistivity, $\Delta\rho = \rho - \rho_0$, of URu_2Si_2 at various pressures. The residual resistivity ρ_0 ($\simeq 15$ μΩcm) is almost independent of pressure.

ρ at room temperature, which is little influenced by pressure, amounts to about 500 μΩcm. $\rho(T)$ increases initially with decreasing temperature but decreases rather rapidly upon cooling below T_{max} where $\rho(T)$ shows a broad peak. A hump-like anomaly is observed at low temperature where the sample orders antiferromagnetically. The overall shape of $\rho(T)$ is shifted towards higher temperature and the peak tends to be collapsed by pressure. The temperature at which ρ has a maximum value

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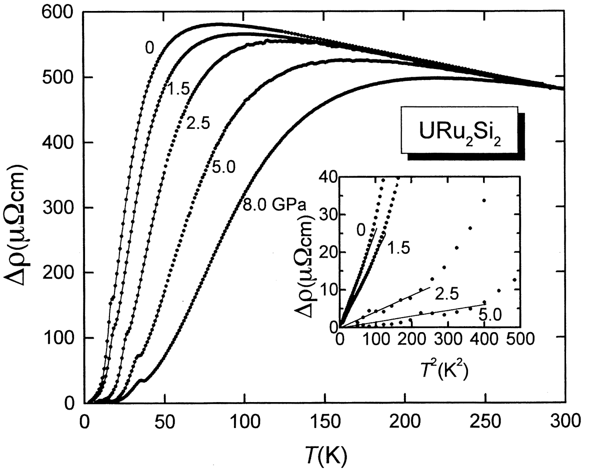

Fig. 1. Temperature dependence of the electrical resistivity of URu_2Si_2 at high pressures. T^2 dependence at low temperatures is shown in the inset.

increases with pressure. It means the characteristic energy of the system, such as the Kondo temperature T_K, is enhanced by volume reduction, which enhances the strength of hybridization between f- and conduction-band electrons in these CK compounds.

The Néel temperature T_N is defined by the minimum of the temperature derivative of ρ and its pressure dependence has been reported elsewhere in detail [8,9]. T_N increases with pressure and saturates at higher pressures to a value near 36 K, $T_N(P)$ may have a broad peak in the vicinity of 8 GPa. Such a peak in the pressure dependence of T_N is expected in a system in which Kondo and RKKY interactions compete, as is seen in the phase diagram proposed by Doniach [10].

It was suggested that the resistivity of URu_2Si_2 for $T<T_N$ is the sum of an ordinary Fermi-liquid type T^2-term and an exponential contribution, which is seen in an antiferromagnet with an energy gap Δ ($\simeq$70 K at 0 GPa) [11]. Since T^2 behaviour is dominant for $T \ll \Delta$, we plot ρ as a function of T^2 at low temperatures, which is shown in the inset of Fig. 1. The solid lines show a quadratic dependence on temperature. At higher pressures the slope is less and the region of T^2 dependence extends to higher temperatures. This behaviour implies also an enhancement of T_K which has been observed in other CK compounds [1,3,5].

Fig. 2 shows the pressure dependence of T_{max} and the coefficient A of the T^2-term. T_{max} at 8 GPa is almost three times larger than at 0 GPa. Assuming that T_{max} is proportional to T_K [12], an increase of T_{max} indicates a corresponding enhancement of T_K. For γ=65 mJ/mol·K^2, the value of A seems to be larger than expected from the Kadowaki–Woods relation [13], such a deviation may be due to antiferromagnetic correlations [14]. A pressure of 8 GPa reduces the value of A by almost two orders-of-magnitude. Since $A \propto 1/T_K^2$, the large decrease in A indicates a large enhancement of T_K and is roughly consistent with conclusions from $T_{max}(P)$.

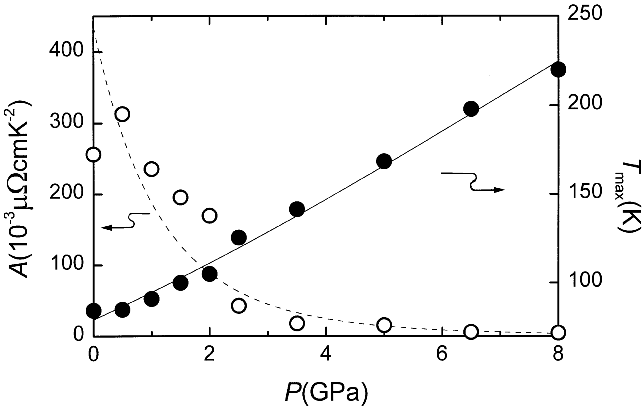

Fig. 2. Pressure dpendence of the ρ-maximum temperature T_{max} and the coefficient A of the T^2-term at low temperature.

In previous work [9] on an X-ray diffraction study of URu_2Si_2 at high pressure, we have reported that the bulk modulus B_0 and its pressure derivative B_0', in the equation $P=B_0/B_0'[(V_0/V)^{B_0'}-1]$, are estimated to be 215 GPa and 3.3, respectively. This means that a pressure of 8 GPa corresponds to a 3.4% decrease in volume. The quite large change in the electronic state that is reflected in the multiplicative enhancement in T_K is caused by only a slight change in volume.

Next we mention the effect of pressure on the thermal expansion of URu_2Si_2. Fig. 3 shows the linear thermal-expansion coefficient α of URu_2Si_2 polycrystalline at 0 and 1.0 GPa for temperatures below 100 K. These were obtained by numerical differentiation of the relative change in length with temperature. We can see a lambda-shaped jump in $\alpha(T)$, which corresponds to antiferromagnetic ordering. $\alpha(T)$ exhibits a temperature dependence quite similar to that of the specific heat $C(T)$ [6–8]. One

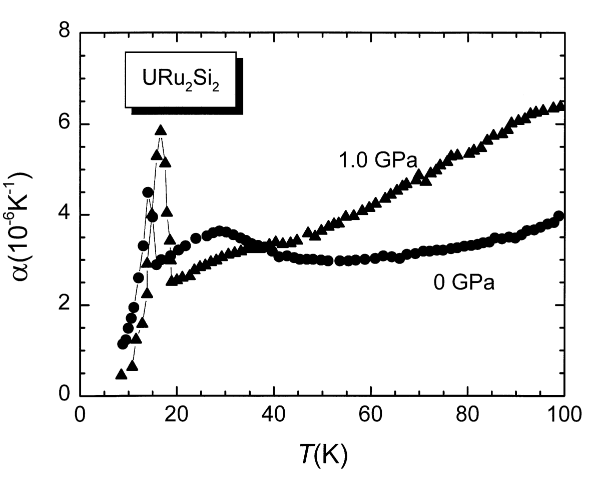

Fig. 3. Linear thermal expansion coefficient of polycrystalline URu_2Si_2 at 0 and 1.0 GPa.

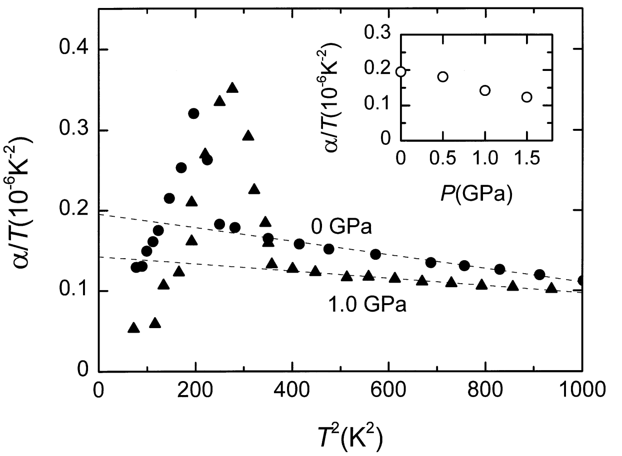

Fig. 4. α/T as a function of T^2. Pressure dependence of the value of α/T extrapolated to $T=0$ K is illustrated in the inset.

exception is the broad peak seen just above T_N where C shows almost a linear temperature dependence without any large anomaly. The peak tends to disappear at higher pressure and we can see only a remnant of it in $\alpha(T)$ at 1.0 GPa. The origin of the peak is not clear at present but may be caused by a crystalline-electric field. If we determine T_N from the temperature at the midpoint of the jump, there is good agreement with T_N obtained from $\rho(T)$ measurements mentioned above.

α/T is plotted as a function of T^2 in Fig. 4 below 1000 K^2 ($T \lesssim 35$ K). Just above T_N, α/T amounts to about 2×10^{-7} K^{-2}, which is one or two orders of magnitude larger than in a noble metal and reflects the large effective electron mass of this substance [15]. We estimate the Grüneisen parameter, according to the expression $\Gamma = 3\alpha V/\kappa C$, to be 23 using values just above T_N both for α/T and C/T, 0.17×10^{-6} K^{-2} and 0.23 J/mol·K^2 [8], respectively. This value is sufficiently large to show that URu_2Si_2 is in a strongly correlated state even in this temperature region.

The value of α/T extrapolated to 0 K, a, is plotted as a function of pressure in the inset of Fig. 3. We find that a decreases with increasing pressure to two thirds of its ambient-pressure value at 1.5 GPa. The specific heat and the thermal expansion coefficient may be expressed at low temperature as $C=\gamma T$ and $\alpha = aT$, respectively. Thus, the value of a could be treated as $a=\kappa\gamma\Gamma/3V$. The γ-value decreases with pressure as T_K increases because of the relation, $\gamma \propto 1/T_K$, which is consistent with the decrease of a.

3. Analysis and discussion

Now we try to analyze the data taking the volume dependence of T_K into account. The values T_{max} and A in the electrical resistivity and a in the thermal expansion coefficient reflect a certain energy scale in the electronic state of URu_2Si_2. In the simplest way, we can consider the following, $T_{max} \propto T_K$, $A \propto 1/T_K^2$ and $a \propto \kappa\Gamma/3VT_K$. To get at the essence of the change in the electronic state, we have to consider these quantities as a function of not the pressure but the volume.

Fig. 5 is a plot of the relative change of T_{max} against that of volume on a logarithmic scale. This plot has a positive slope, because T_{max} increases with pressure (and hence with decreasing volume), with negative curvature. Considering the relation between T_{max} and T_K, the Grüneisen parameter, $\Gamma \equiv -\partial \ln T_K/\partial \ln V$, corresponds to the slope in this plot and is estimated to be 40 at ambient pressure by calculating a tangent at the origin. The slope becomes smaller with volume contraction and is estimated to be 30 at 8 GPa. The decrease of Γ with increasing T_K is consistent with the fact that the values of Γ for CK materials having a large T_K are smaller than those of low-T_K materials.

To estimate the volume dependence of Γ, the Grüneisen parameter for the strength of hybridization is defined as $q=-|JN(0)|$. The value of q usually has a value between 6 and 8 [16]. Considering the expression of the T_K associated with the strength of hybridization, the volume dependence of Γ is written as $-q=-\partial\ln\Gamma/\partial\ln V$ or $\Gamma = \Gamma_0(V/V_0)^q$ [2]. From the least-squares fit of the data in Fig. 5, the initial values of Γ, Γ_0, and q are calculated to be 40 and 8, respectively. The same analysis is also valid for the pressure dependence of A in Fig. 2 [17].

The volume dependence of a is more complex because all values in the expression $a \propto \kappa\Gamma/3VT_K$ depend on volume. The volume dependence of T_K, Γ and κ/V are defined to be Γ, $-q$ and $-B_0'$, respectively. Thus the volume dependence of a is written as $-\Gamma - q - B_0'$ and calculated to be -51. This means that the pressure of 1.5 GPa reduces the a-value to 70%. The result of the thermal expansion measurement is well explained by the thermodynamical analysis.

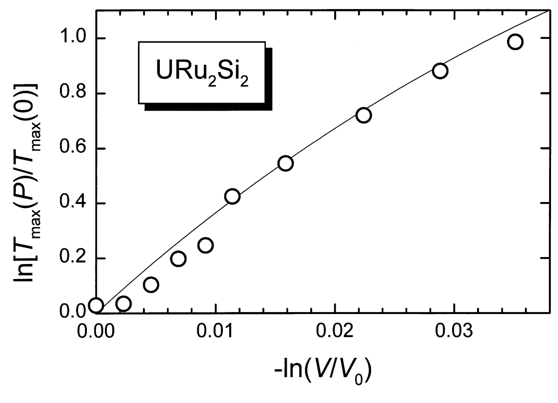

Fig. 5. Logarithmic plot of the relative change of T_{max} and V for URu_2Si_2. The solid line shows the result of fitting to a Grüneisen analysis.

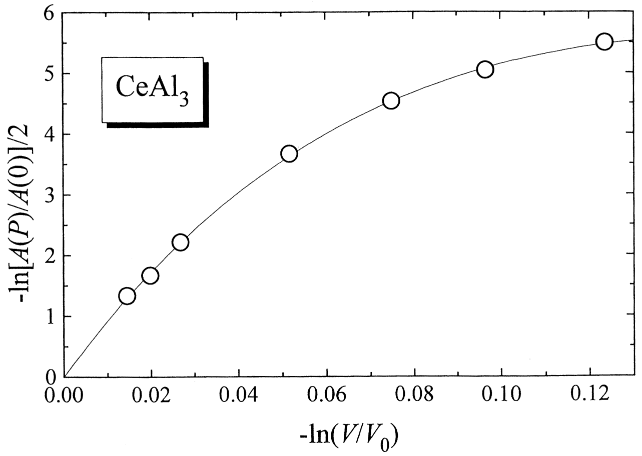

Fig. 6. Logarithmic plot of the relative change of the coefficient of the T^2-term in the electrical resistivity A and volume for CeAl$_3$.

Finally, we report briefly another example showing that these analyses are valid for other CK compounds. Fig. 6 gives the logarithm of the volume dependence of the T^2-coefficient of resistivity A for a typical HF compound CeAl$_3$. We plot -ln[$A(P)/A(0)$]/2 on the vertical axis, considering that the A-value is inversely proportional to T_K^2 [1]. A decrease in the volume by about 10% induces a large enhancement of T_K and hence a crossover from a HF to an IV state. Values of Γ_0 and q are calculated from the slope and curvature of the curve in Fig. 6 to be 97 and 6, respectively. The large value of Γ_0 reflects the pressure-sensitive electronic structure of a low-T_K HF compound. We observe that Γ becomes smaller as the electronic state changes into a high-T_K IV state.

This analysis has been applied also to several other CK materials that have a range of T_K values and it shows that the initial value of Γ is commensurate with T_K at ambient pressure and the q-value is roughly independent of T_K. It should be emphasized that this analysis of the electrical resistance and the thermal expansion coefficients has broad application for describing the electronic state of CK compounds.

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