



## Thermal expansion of the non-Fermi liquid system Th<sub>1-x</sub>U<sub>x</sub>Ru<sub>2</sub>Si<sub>2</sub> ( $x \leq 0.07$ )

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### Abstract

We have studied non-Fermi liquid properties of the tetragonal diluted uranium alloys Th<sub>1-x</sub>U<sub>x</sub>Ru<sub>2</sub>Si<sub>2</sub> ( $x \leq 0.07$ ), by means of thermal expansion measurements in the temperature range 0.5–100 K. As the temperature is lowered below about 10 K, the volume effect  $\alpha_v(T)/T$  ( $= (\alpha_c + 2\alpha_a)/T$ ) is found to exhibit the tendency to diverge logarithmically, similar to other quantities of the system:  $C/T \sim -\ln T$ ,  $\chi \sim -\ln T$ ,  $\rho \sim \ln T$  (or  $T^{1/2}$ ). Interestingly, the sign of the anomaly in  $\alpha_v(T)$  is negative, opposite to the usual Kondo shrink behavior. As a possible interpretation, we discuss the two-channel Kondo model, proposing c-f hybridization effects dominated by the virtual  $f^2$ - $f^3$  charge fluctuations.

**Keywords:** Non-Fermi liquid system; Th<sub>1-x</sub>U<sub>x</sub>Ru<sub>2</sub>Si<sub>2</sub>; Thermal expansion; Kondo effect

Our previous reports of the diluted uranium alloys Th<sub>1-x</sub>U<sub>x</sub>Ru<sub>2</sub>Si<sub>2</sub> ( $x \leq 0.07$ ) have revealed non-Fermi liquid (NFL) behavior in the transport, thermal and magnetic properties at low temperatures [1]: The *Sf* electronic specific heat  $C_{sf}(T)/T$  and the magnetic susceptibility  $\chi(T)$  follow a  $-\ln T$  function over two decades of temperature below 10 K down to  $\sim 0.1$  K. The unusual feature of the local *f* states is further pronounced in the electrical resistivity  $\rho(T)$  that exhibits a steep decrease as the temperature drops, following  $\sim \ln T$  ( $1 \text{ K} < T < 10 \text{ K}$ ) and  $\sim T^{1/2}$  ( $0.1 \text{ K} < T < 1 \text{ K}$ ).

Although there is much similarity found between the NFL phenomena in various materials [2], difference in environments of *f* ions admits of at least two possible interpretations to the problem, i.e. the two-channel Kondo effects (TCKE) [3] and a second-order phase transition at  $T = 0$  [4].

In Th<sub>1-x</sub>U<sub>x</sub>Ru<sub>2</sub>Si<sub>2</sub>, however, the latter case may be ruled out, because the systematical dilution of

uranium ions ensures that the NFL behavior is ascribed to the nature of a single uranium ion doped in ThRu<sub>2</sub>Si<sub>2</sub>. At present, the most promising explanation may be given in terms of the single-impurity model of TCKE that reproduces the temperature variations of *C* and  $\chi$  fairly well by adopting  $T_K \sim 11 \text{ K}$  [1]. In addition, recent development of theoretical work has predicted the possibility of the decrease in  $\rho(T)$  as  $T \rightarrow 0$  [5, 6]. In this paper, we have performed the thermal expansion measurements to further elucidate the NFL properties of this dilute system.

Single-crystalline Th<sub>1-x</sub>U<sub>x</sub>Ru<sub>2</sub>Si<sub>2</sub> samples ( $x = 0, 0.03, 0.07$ ) were prepared in a tri-arc furnace under an argon atmosphere, using the Czochralski technique [1]. The crystals were carefully shaped by means of a spark erosion machine into the parallelepiped ( $1 \times 1 \times 2 \text{ mm}^3$ ), with each pair of the end surfaces parallel to within 5  $\mu\text{m}$ . The coefficients of linear thermal expansion,  $\alpha = L^{-1}dL/dT$ , were measured by means of a high-precision three-terminal capacitance technique [7] in the

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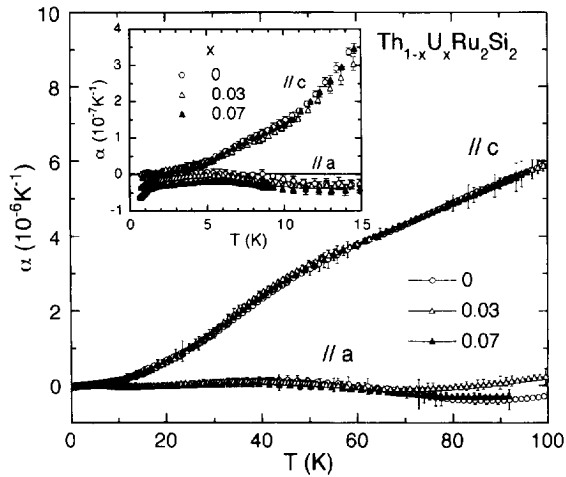


Fig. 1. The thermal-expansion coefficient along the two crystallographic axes of the tetragonal diluted alloys  $\text{Th}_{1-x}\text{U}_x\text{Ru}_2\text{Si}_2$  ( $x = 0, 0.03, 0.07$ ).

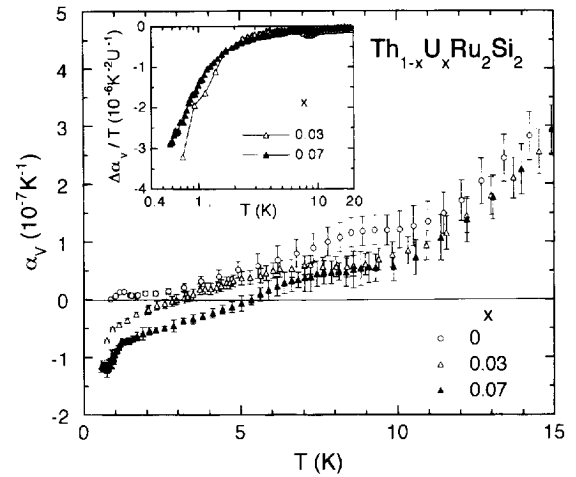


Fig. 2. The volume thermal-expansion coefficient  $\alpha_v = \alpha_c + 2\alpha_a$  of  $\text{Th}_{1-x}\text{U}_x\text{Ru}_2\text{Si}_2$  ( $x = 0, 0.03, 0.07$ ). The inset shows the temperature variations of the 5f electronic contribution to the volume effect,  $\Delta\alpha_v/T$ .

temperature range 0.5–100 K, with special care of the empty-cell correction.

Fig. 1 shows the temperature variations of the thermal expansion coefficients of  $\text{Th}_{1-x}\text{U}_x\text{Ru}_2\text{Si}_2$  ( $x = 0, 0.03, 0.07$ ),  $\alpha_{c,a}(T)$ , along the tetragonal  $c$ - and  $a$ -axis. In this overview,  $\alpha_c(T)$  increases as the temperature rises, whereas  $\alpha_a(T)$  is almost  $T$ -independent. We may attribute this uniaxial anisotropy at high temperatures to the nature of the non- $f$  matrix, since there is no systematic variation of  $\alpha$  on  $x$ . At low temperatures, on the other hand, the effect of doping uranium becomes significant with opposite anisotropy (the inset of Fig. 1): Below  $\sim 10$  K, the  $\alpha_a(T)$  curves for the doped alloys clearly deviate from the data of the host metal, while no distinctive deviation could be detected along the  $c$ -axis.

The coefficients of volume expansion,  $\alpha_v(T)$  ( $\equiv \alpha_c + 2\alpha_a$ ) are shown in Fig. 2. The anomalous decrease of  $\alpha_a$  at low temperatures is reflected by the change of the sign of  $\alpha_v$  below 5 K for  $x = 0.07$  and 3 K for  $x = 0.03$ . Note the deviation of those  $\alpha_v(T)$  curves from 0 even at the lowest temperature of 0.5 K. This implies that they should have minima below 0.5 K because of the thermodynamic constraint ( $\alpha(0) = 0$ ).

Subtracting the data of the host metal, we deduce the 5f electronic contributions to the volume

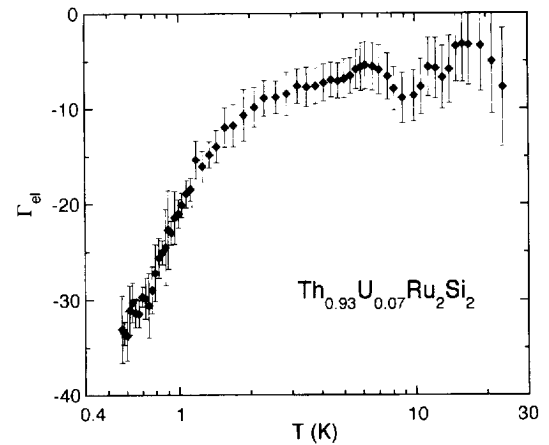


Fig. 3. The 5f electronic Grüneisen parameter of  $\text{Th}_{0.93}\text{U}_{0.07}\text{Ru}_2\text{Si}_2$  (deduced with  $\kappa = 0.92 \times 10^{-11} \text{ Pa}^{-1}$ ).

expansion,  $\Delta\alpha_v(T)$  (the inset of Fig. 2). As the temperature is lowered below 10 K,  $\Delta\alpha_v(T)/T$  gradually decreases and approaches a  $\ln T$  curve below  $\sim 2$  K. From this result and the  $C_{5f}(T)$  data [1], we obtain in Fig. 3 the 5f-electronic Grüneisen parameter  $\Gamma_{e1}(T) = \Delta\alpha_v(T)V_m/\kappa C_{5f}(T)$  for  $x = 0.07$ , where  $V_m \sim 5.20 \times 10^{-5} \text{ m}^3/\text{mol}$  is the molar volume and the compressibility  $\kappa$  is substituted

by  $0.92 \times 10^{-11} \text{ Pa}^{-1}$ , a weighted average for the constituent elements.

The sign of  $\Gamma_{ei}(T)$  is negative in the whole temperature range where  $C_{5f}(T)$  is enhanced [1]. This is a remarkable contrast to the usual heavy-electron materials as well as to the dilute magnetic alloys. Most of them exhibit positive enhancement of  $\Gamma_{ei}$  at low temperatures, known as the Kondo shrink (or collapse) effects: smaller  $f(d)$  atomic volume gains larger  $c$ - $f(d)$  exchange energy. On the other hand, some effects which may lead to the negative  $\Gamma_{ei}$  are also known: (i) the inter-site correlations in the concentrated  $f(d)$  systems [8], (ii) the ferromagnetic Kondo effects in the heavy-rare-earth-doped metals [9], and (iii) the screening effects of the core potential by the Hund's rule  $f$  shells in the mixed valence systems [10]. All of those, however, do not apply to the present system: First, the unusual feature of  $\Delta\alpha_V$  is scaled by  $x$ , similarly to that of  $C$ ,  $\chi$  and  $\rho$ , ensuring that the phenomenon is of single-site origin. Second, the effective single-site correlation estimated from  $\chi^{-1}(T)$  is of antiferromagnetic nature [11]. In addition, the case (iii) compels the unrealistic  $f^3$ - $f^4$  valence mixing to produce the negative sign of  $\alpha_V$ .

We shall now examine the possibility of TCKE: Similarly to the conventional Kondo model,  $\Gamma_{ei} (= -d \ln T_K / d \ln V)$  yields the volume derivative of the effective  $c$ - $f$  exchange parameter  $|J|$ , since  $T_K$  depends on  $|J|$  exponentially [3]. The negative  $\Gamma_{ei}$  therefore corresponds to the depression of  $|J|$  under pressure. Further insight may be given by the Schrieffer-type expression, conceptually written as  $|J| \sim (|V_{12}|^2/E_{12} + |V_{32}|^2/E_{32})$ , where  $V_{nm}$  denote the averaged matrix elements of the  $c$ - $f$  hybridization through the  $f^m$ - $f^n$  virtual processes with the excitation energy of  $E_{nm}$ . TCKE arise from a relevant coupling that links a stable non-Kramers doublet (in  $f^2$ ) to an excited Kramers

doublet (in  $f^1$  or  $f^3$ ). In zeroth-order approximation,  $E_{12}$  and  $E_{32}$  may be written as  $-E_f - U$  ( $> 0$ ) and  $2U + E_f$  ( $> 0$ ) [12], where  $E_f$  ( $< 0$ ) is the energy of the  $5f$  orbital in the atomic potential measured from the Fermi level and  $U$  is the Coulomb repulsion integral between two  $5f$  electrons. If we assume as in the usual dilute alloys that the effect of decreasing the atomic volume is to increase  $|V_{nm}|$  and to decrease  $|E_f|$  [13], then the pressure always enhances the  $f^2$ - $f^1$  process, but may suppress the  $f^2$ - $f^3$  process when  $|dE_f| > |dV_{32}|$ . Thus we may in principle expect the negative sign of  $\Gamma_{ei}$ , if the dominant coupling for TCKE lies in the  $f^2$ - $f^3$  process.

In conclusion, the diluted uranium system  $\text{Th}_{1-x}\text{U}_x\text{Ru}_2\text{Si}_2$  exhibits a negative volume expansion in its low-temperature NFL state, implying that its characteristic energy should be depressed under pressure. Within the context of the two-channel Kondo model, we may attribute the behavior to the dominant  $f^2$ - $f^3$  valence fluctuation.

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