

Thermal expansion at the metamagnetic transition in single-crystalline heavy-fermion (Ce,La)Ru₂Si₂

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We present measurements of the volume expansion (α_v) in magnetic fields up to 8.5 T and in the temperature range 1.3 K < T < 20 K on the heavy-fermion compounds Ce_{1-x}La_xRu₂Si₂ ($x = 0$ and 0.05). In reduced scales the two compounds present the same low temperature Grüneisen coefficients. Emphasis is given on the strong field dependence of the low temperature extremum of α_v . The validity of scaling law with pressure is verified. The interest to use a magnetic field to approach a magnetic instability is strongly underlined.

I. INTRODUCTION

In the heavy-fermion systems the understanding of the competition between the local fluctuations and the intersite coupling is still an open problem.¹ In the past few years it has become clear that the proximity of a long-range magnetic instability is a key feature in heavy-fermion systems. In that respect, the compound CeRu₂Si₂ is an exemplary system. It shows a metamagnetic transition at $B^* = 7.8$ T (Refs. 2 and 3) for a field direction along the tetragonal axis in the liquid-helium temperature range. When alloying with La a long-range antiferromagnetic order develops for La concentrations above $\sim 8\%$, with a Néel temperature of 6 K.

Inelastic neutron-scattering experiments⁴ on CeRu₂Si₂ have shown that the metamagnetic transition is related to a field induced change in magnetic correlations. The zero field ground state is expected to be nonmagnetic as the local spin fluctuations are stronger than the intersite fluctuations. In fact, no long-range order has been detected down to 20 mK.⁵

Changing experimental variables such as pressure or applying a magnetic field might enable us to elucidate the magnetic instability. The aim of this paper is to present thermal-expansion measurements in field for heavy-fermion CeRu₂Si₂ and the compound in which 5% of Ce has been replaced by La. In the latter compound B^* has dropped to 5.6 T.⁶ The heavy-fermion behavior in both compounds is well illustrated by the large linear term in the electronic specific heat⁷: $\gamma = 350$ for $x = 0$ and $\gamma = 500$ mJ/mol K² for $x = 0.05$. The measured very large pressure variations of the electronic parameters give an extraordinary large electronic Grüneisen parameter: $\Gamma_{el} \sim 160$ and 220 for CeRu₂Si₂ and Ce_{0.95}La_{0.05}Ru₂Si₂, respectively.⁸

II. EXPERIMENT

Single-crystalline samples of CeRu₂Si₂ and Ce_{0.95}La_{0.05}Ru₂Si₂ were grown by the Czochralski technique. The samples were shaped by means of spark erosion into a parallelepiped with edges ~ 3 –5 mm, respectively. Experimental details and sample preparation have been described elsewhere.³ The coefficient of linear thermal expansion

$\alpha = (1/L)(dL/dT)$ was measured using a sensitive three-terminal capacitance method.⁹ In order to obtain the volume expansion (α_v), the coefficient of the linear thermal expansion has been measured along (α_{\parallel}) and perpendicular (α_{\perp}) to the tetragonal axis with the magnetic field ($B_{max} = 8.5$ T) applied along the c axis: $\alpha_v = \alpha_{\parallel} + 2\alpha_{\perp}$. The data were gathered stepwise, the lowest temperature amounted to 1.3 K.

III. RESULTS

In a previous paper we analyzed the thermal expansion of the CeRu₂Si₂ compound over a wide temperature range: 1.3 K < T < 300 K.³ In this contribution we will focus only on the low temperature regime, $T < 20$ K. In order to relate the volume expansion to the specific heat, we make use of the so-called experimental Grüneisen parameters. The effective (temperature dependent) Grüneisen parameter is given by (see, for instance, Ref. 1)

$$\Gamma_{eff}(T) = V\alpha_v(T)/\kappa C_p(T), \quad (1)$$

where V is the molar volume, $\kappa = -(1/V)(\partial V/\partial P)$ is the isothermal compressibility,¹⁰ α_v is the volume expansion, and C_p is the molar specific heat at constant pressure. Combining our $\alpha_v(T)$ measurements with existing specific heat data,⁷ we have calculated the effective Grüneisen parameter shown in the inset of Fig. 1. The increase of $\Gamma_{eff}(T \rightarrow 0)$ from 160 (for $x = 0$) to 220 (for $x = 0.05$) is related to the approach of the magnetic instability, notably a long-range magnetic order which occurs for $x \geq 0.08$. However, in reduced units $\Gamma_{eff}(T)/\Gamma(T_m)$ vs T/T_m (T_m is the temperature that indicates the extremum of α_v : $T_m = 9$ K for $x = 0$ and $T_m = 6$ K for $x = 0.05$), the experimental curves for $x = 0$ and 0.05 coincide (see Fig. 1). Thus, the two compounds have a similar behavior on a normalized energy scale. The huge magnitudes of $\Gamma_{eff}(T \rightarrow 0)$ are correlated to the magnetic instability.

In a recent report on the field dependence of thermal expansion in magnetic fields on CeRu₂Si₂,¹¹ our experimental limitation to study the high field regime ($B > B^*$) was attained. As B^* decreases drastically with increasing x , a new similar experiment has been performed for $x = 0.05$ (Fig. 2). As B^* is 5.6 T for $x = 0.05$, the high-field regime can be clearly studied. Qualitatively, the data for the $x = 0.05$ compound are similar to those previously found for

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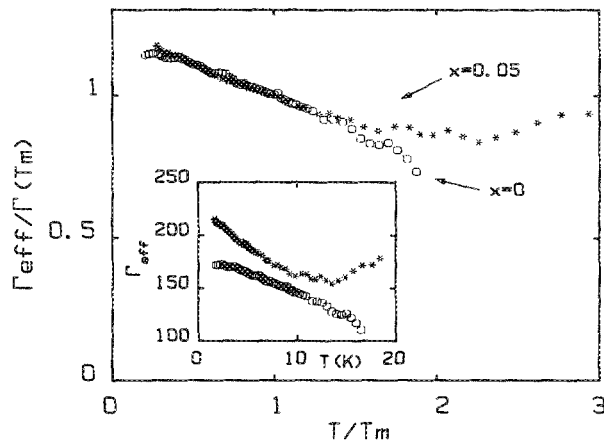


FIG. 1. Scaled Grüneisen parameters: $\Gamma_{\text{eff}}(T)/\Gamma(T_m)$ vs T/T_m for the $\text{Ce}_{1-x}\text{La}_x\text{Ru}_2\text{Si}_2$ system. The inset shows the effective temperature dependent Grüneisen parameters: (O) for $x = 0$ and (*) for $x = 0.05$.

$x = 0$. The change of the sign of the coefficient of the volume expansion at B^* is directly connected to the occurrence of a maximum in the linear temperature coefficient $\gamma(B)$ of the low-temperature specific heat and to the huge pressure variation of B^* .¹² The inset of Fig. 2 shows the field dependence $T_m(B)$ for $x = 0$ and 0.05. It is now confirmed that $T_m(B)$ increases strongly with B for $B > B^*$. Below B^* , $T_m(B)$ seems to qualitatively mimic the boundary of a phase diagram separating a low-field regime, where magnetic correlations (notably antiferromagnetic) dominate, from a high-field phase where only local fluctuations dominate. The quasilinear sharp decrease of T_m as B increases to B^* suggests that T_m may vanish at B^* , i.e., a magnetic transition will exist at $T = 0$ K. In fact, when B reaches B^* , T_m does not vanish but reaches a deep minimum in the vicinity of B^* . In this region ($B \sim B^*$) local fluctuations of very weak energy appear to prevent a real phase transition at 0 K. It is obvious that, further very low-temperature experiments must be performed to study the low-temperature properties for $B \sim B^*$. Experimentally, it is worthwhile to emphasize that the drastic field variation of the energy scale found here

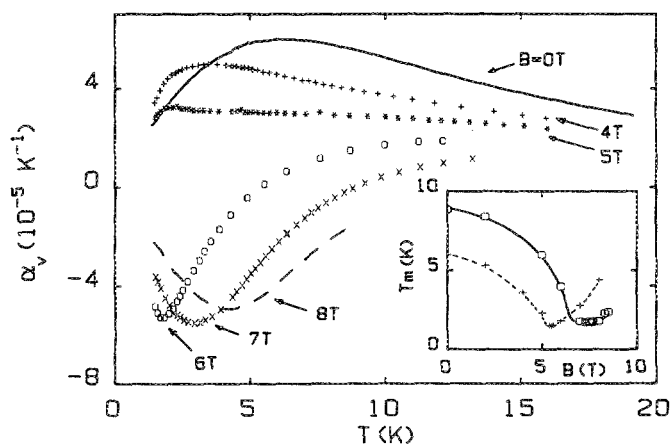


FIG. 2. Coefficient of volume expansion of $\text{Ce}_{0.95}\text{La}_{0.05}\text{Ru}_2\text{Si}_2$ for magnetic fields as indicated. The inset shows the field dependence of the extremal temperature T_m , for $x = 0.05$ (dashed line), and for $x = 0$ (solid line).

(T_m) is not obvious in the direct specific-heat measurements. The γ term of the specific heat only shows an increase of 30% at B^* by comparison to the $\gamma(B = 0)$ value.⁷ Thermopower measurements sensitive to the energy derivative of the density of state also show a huge anomaly at B^* .¹³ A very low energy scale is also detected in the low-temperature dependence ($T \sim 1.3$ K) of the differential susceptibility $\chi = \partial M / \partial B$ at B^* , which follows a Curie-Weiss law with a vanishing θ value.¹¹

The measurements of the thermal expansion at different fields can be used to test the validity of the scaling ansatz (SA) found in CeRu_2Si_2 , i.e., an entropy of the form:

$$S = S\left(\frac{T}{T_s(P)}, \frac{B}{B_s(P)}\right). \quad (2)$$

We will assume that only one single pressure dependent energy scale exists. This energy scale is indifferently expressed in temperature $T_s(P)$ or field $B_s(P)$. In this approach, the linear terms of the volume expansion ($a = \alpha_V/T$) and molar specific heat ($\gamma = C_p/T$) are related by the following differential equation¹¹:

$$a(B) = \frac{\Omega}{V} \left(\gamma + B \frac{\partial \gamma}{\partial B} \right), \quad (3)$$

where V is the molar volume. To characterize the huge magnetoelastic coupling, we define

$$\Omega = \frac{d \ln T_s}{dP} = \frac{d \ln B_s}{dP} = 160 \text{ Mbar}^{-1}$$

(for the CeRu_2Si_2 compound).¹¹ In the same temperature range, Maxwell's relation implies: $(\partial M / \partial T)_{P,B} = T(\partial \gamma / \partial B)_P$. Thus, low-temperature ($1.3 \text{ K} > T > 2.0 \text{ K}$) magnetization measurements have been performed on the same single-crystalline samples as used for the thermal-expansion experiments. Using the initial value $\gamma = 350 \text{ mJ/mol K}^2$ (Ref. 7) at $B = 0$, we are able to compare the prediction of Eq. (3) to the experimental values of $a(B)$. Our result for the CeRu_2Si_2 compound is reported in Fig. 3. It is readily seen that the overall agreement is excellent: Eq. (3) implies that $a(B)$ changes sign slightly above the maximum in $\gamma(B)$. However, very close to 8 T the predicted val-

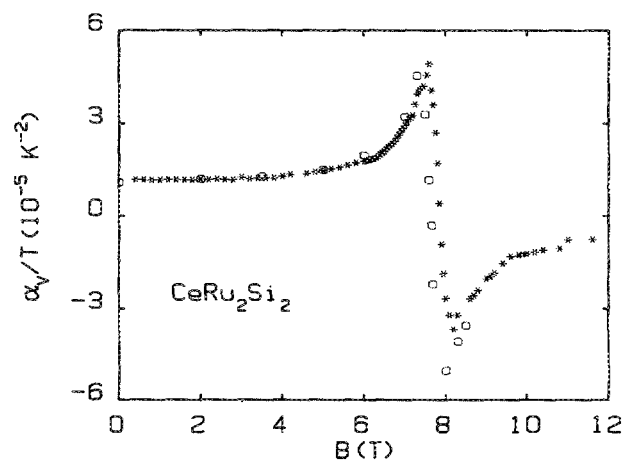


FIG. 3. Field dependence of the linear term of the volume expansion $a = \alpha_V/T$, for CeRu_2Si_2 . (O) as measured; (*) as calculated using Eq. (3).

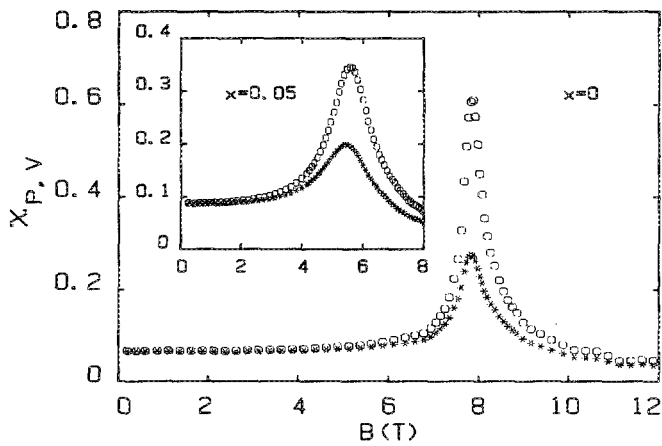


FIG. 4. χ_P (○) and χ_v (*) (in units of μ_B/T) for the $Ce_{1-x}La_xRu_2Si_2$ system with $x = 0$ and 0.05 (inset).

ues of $\alpha(B)$ from Eq. (3) are smaller than the experimental values. This may be due to the fact that in this region $T_m(B)$ is a little bit larger than the temperature range where we evaluated α_v/T and $\partial M/\partial T^2$.

Recently, it has been pointed out¹⁴ that the huge increase of $\chi_P = (\partial M/\partial B)_P$ in the $CeRu_2Si_2$ compound may be due to the big volume change around B^* . For example, at 1.3 K, $\chi_P(B^*)/\chi_P(B=0)$ is near 10; for comparison $\gamma(B^*)/\gamma(B=0)$ is only 1.3. The huge maxima of χ_P/γ at B^* was taken as an indication that the ferromagnetic correlation (i.e., the low energy response) plays a dominant role at B^* . However, it has been pointed out that the huge increase of $\chi_P(B^*)$ may be mostly due to the large magnetostriction effect. Indeed the question is: would the system present a sharp maximum in $\chi_v = (\partial M/\partial B)_v$ in a fixed volume isotherm? By standard thermodynamics the difference $\chi_P - \chi_v$ has been evaluated:

$$\chi_P - \chi_v = \frac{V}{\kappa} \left(\frac{\partial V}{\partial B} \right)_{P,T}^2. \quad (4)$$

In this equation, the magnetostriction $(\partial V/\partial B)$, compressibility (κ) and χ_P are known quantities. For the $CeRu_2Si_2$ compound χ_P , $(\partial V/\partial B)$ and κ have been measured.^{3,15} Since neither χ_P nor κ has been measured yet for our $Ce_{0.95}La_{0.05}Ru_2Si_2$ compound, these quantities have been determined from SA.^{16,17} We have evaluated χ_v and χ_P at $T = 1.3$ K and report the results in Fig. 4. It is readily seen that: (i) the increase in χ_P is partially due to the volume

change in isobaric isotherms and (ii) a sharp maximum with an overall increase of a factor of 5 (for $x = 0$) is still present in χ_v at B^* . We expect that the metamagnetic transition in this compound is thus an intrinsic property of the heavy electron wave function and that it is the cause of the huge volume change at B^* and not the contrary. The Wilson ratio (χ/γ) when calculated with χ_v will show a maximum at B^* .

To summarize, we have presented extensive data on the thermal expansion in magnetic fields of the single-crystalline heavy-fermion $CeRu_2Si_2$ and $Ce_{0.95}La_{0.05}Ru_2Si_2$. The results show that such experiments can be very powerful tools for studying systems close to a magnetic instability.

ACKNOWLEDGMENTS

A. L. is supported by CNPq (Conselho Nacional de Desenvolvimento Científico e Tecnológico, Brazil). The work of A. d. V. has been made possible by a fellowship of the Royal Netherlands Academy of Arts and Sciences.

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