

Journal of Magnetism and Magnetic Materials 226-230 (2001) 252-253



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## Magnetic collapse in $Ce_{1-x}La_xRu_2Si_2$ studied by thermal expansion

P. Haen<sup>a,\*</sup>, S. Kambe<sup>b</sup>, H. Bioud<sup>a,c</sup>, A. de Visser<sup>d</sup>

<sup>a</sup>CRTBT, CNRS and Université Joseph Fourier, BP 166, 38042 Grenoble Cedex 9, France
<sup>b</sup>Advanced Science Research Center, JAERI, Tokai, Ibaraki 319-1195, Japan
<sup>c</sup>Fac. des Sciences, Université Chouaib Doukkali, BP 20, El Jadida, Morocco
<sup>d</sup>Van der Waals-Zeeman Inst., Valckenierstraat 65, 1018 XE Amsterdam, The Netherlands

## Abstract

The variation of  $T_N$  of  $Ce_{1-x}La_xRu_2Si_2$  single crystals with concentrations (x = 0.08, 0.1, 0.13 and 0.2) larger than the critical value  $x_c = 0.075$  for which the system shows an antiferromagnetic ground state is reexamined by thermal expansion experiments. The dependence of  $T_N$  near  $x_c$  is discussed. © 2001 Elsevier Science B.V. All rights reserved.

Keywords: Heavy fermions; Thermal expansion

An interesting question as regards the study of the magnetic collapse in the vicinity of a quantum critical point is whether the (generally antiferromagnetic (AFM)) ordering temperature,  $T_N$ , can reach 0 K at the critical point. However, it might be difficult to answer this question, because  $T_N$  becomes hard to determine when the anomalies associated with the AFM order vanish on approaching the critical point, following the reduction of the magnetic moment. From this point of view, the  $Ce_{1-x}La_{x}Ru_{2}Si_{2}$  system is very interesting, because it is rather well documented. Replacing Ce by La in the heavy fermion compound CeRu2Si2 expands the volume and reduces the Kondo temperature,  $T_{\rm K}$ . Neutron-diffraction experiments have shown that single crystals with x = 0.08, 0.1, 0.13 and 0.2 exhibit a modulated AFM order with a wave vector  $\mathbf{k} = [0.31, 0, 0] [1]$ .  $T_{\rm N}$  and the ordered moment  $M_0$  both show a sharp increase on increasing x to 0.2. The critical concentration in this system is now well known:  $x_c = 0.075$  [2]. Some of these crystals were studied by specific heat, c(T), measurements (x = 0.1 and 0.13 [3], x = 0.075 [4]), and magnetic measurements (x = 0.1 and 0.13 [3], x = 0.2 [5]).

We have performed thermal expansion experiments using a capacitance cell on the same cystals in the case of x = 0.08, 0.1 and 0.13 and on a new x = 0.2 crystal, of which c(T) was also measured [6]. The crystals were spark cut in order to obtain surfaces perpendicular to the tetragonal a- and c-axis, respectively. Here, we report only the variation of the thermal expansion coefficient,  $\alpha(T) = (1/L)(dL/dT)$ , along the *c*-axis (top of Fig. 1). We know from measurements for x = 0.2 [6], that  $\alpha(T)$  along the *a*-axis exhibits exactly the same variation as along *c*, but its value is 2.5 times smaller. For x = 0.2, 0.13 and 0.1,  $\alpha(T)$  shows a drop at  $T_N$  on cooling, with a change of sign. The second drop is seen for x = 0.2, at  $T_{\rm L} = 1.8$  K, which temperature, coincides with an anomaly in the Tdependence of the third harmonic 3k = [0.93, 0, 0] of the AFM ordering vector [1,7], i.e. to some squaring of the modulation.  $T_{\rm L}$  also exists for x = 0.13 (and equals 0.6 K [3]), but has not been detected for lower x. For x = 0.08, the  $\alpha(T)$  curve (measured down to 0.5 K) shows no clear trace of ordering. The maximum near 5K is similar to those occurring at 9 and 6K for non-ordered CeRu<sub>2</sub>Si<sub>2</sub> and the x = 0.05 alloy, respectively [8]. These temperatures scale with  $T_{\rm K}$ . For the x = 0 and 0.05 compounds, the variation of  $\alpha(T)$  as well as that of other properties is quite well accounted for by the SCR model [4,9].

The bottom part in Fig. 1 represents the derivatives  $d\alpha/dT$  of the curves plotted in the top panel. The

<sup>\*</sup>Corresponding author. Tel.: + 33-0-4-76-52-10-66; fax: + 33-0-4-76-87-50-60.

E-mail address: haen@labs.polycnrs-gre.fr (P. Haen).



Fig. 1. Top: Temperature dependence of the thermal expansion coefficient  $\alpha(T) = (1/L)(dL/dT)$  along the *c*-axis of Ce<sub>1-x</sub>La<sub>x</sub>Ru<sub>2</sub>Si<sub>2</sub> single crystals with x = 0.08, 0.1, 0.13 and 0.2. Bottom: Temperature dependence of the derivatives  $d\alpha/dT$  of the curves shown in the upper part.

well-defined peaks seen for x = 0.2 and 0.13 become broad maxima for x = 0.1 and 0.08. This smearing of the  $d\alpha/dT$  peak when x is lowered can be attributed to weak crystalline disorder. A relative uncertainty of 3% in the value of x is sufficient to account for the tiny residual static moment measured for the  $x_c = 0.075$  crystal [2]. Similar weak disorder can be expected for all the crystals studied here, but its effect is most important for small x.

The temperatures of the peaks or maxima in  $d\alpha/dT$ can be taken to represent  $T_{\rm N}$  (and  $T_{\rm L}$  for x = 0.2). The  $T_{\rm N}$  values are plotted in Fig. 2. This figure also shows the temperatures of the c(T) peaks, when they exist, which are slightly lower. In fact, for x = 0.2 and 0.13, the c(T)peak temperatures coincide with the  $\alpha(T)$  minima. For all concentrations, the  $d\alpha/dT$  maximum corresponds to the inflection of the jump of c(T)/T on cooling. This jump is also smeared on decreasing x (see Ref. [3]). We also report in Fig. 2 T<sub>N</sub> values derived from magnetic measurements for x = 0.13 and 0.1. In these cases, the initial susceptibility,  $\chi(T)$ , shows no peak, but  $T_N$  could be defined as the temperature above which no steps (resulting from the crossing of metamagnetic lines) appear in the magnetization curves [3,5]. These  $T_N$  values are in excellent agreement with those of the maxima in  $d\alpha/dT$ . Metamagnetic steps have not been detected for x = 0.08, even not at 0.1 K. The x = 0.2 alloy shows a  $\chi(T)$  peak but, according to the data in Ref. [5], this peak is located a bit lower than the present  $d\alpha/dT$  peak.



Fig. 2. Experimental  $T_N$  vs. x variation in  $Ce_{1-x}La_xRu_2Si_2$ (dashed line), as determined from the temperatures of the  $d\alpha/dT$ maxima in the bottom panel of Fig. 1 (O) and, for x = 0.1 and 0.13 from magnetic measurements ( $\Box$ , [3]). For comparison: peak temperatures of c(T) (V) for x = 0.1, 0.13 [3] and 0.2 [5]. Dotted line: calculated  $(x - x_c)^{2/3}$  variation adjusted on the  $T_N$  value for x = 0.1.

The  $T_N$  values derived from neutron-diffraction experiments are not reported in Fig. 2 because large error bars should be associated with these values, especially for low x due the existence of a tail near  $T_N$  in the magnetic intensity.

The  $T_N$  variation in Fig. 2 seems to scale with the power  $\frac{2}{3}$  predicted by theoretical models [10]. (This contrasts with the almost linear variation observed in CeCu<sub>6-x</sub>Au<sub>x</sub> for  $0.1 \le x \le 1$  [11] which is also theoretically expected.) However, it appears that  $T_N$  in Fig. 2 cannot be accounted for by a simple  $(x - x_c)^{2/3}$  variation, except in a quite narrow concentration range  $(x \le 0.1)$ . A correction can be made for the Ce concentration, but this does not ameliorate the fits. However, replacing the true control parameter considered in the models by  $(x - x_c)$  is an oversimplification which might be valid close to  $x_c$  only. The study of other crystals with  $x_c \le x \le 0.13$  should give a more detailed  $T_N$  variation in this range, and allow to improve the comparison with theoretical models.

We thank Dr. J. Flouquet for fruitful discussions.

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