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DOI

[10.1088/0953-8984/9/23/015](https://doi.org/10.1088/0953-8984/9/23/015)

Publication date

1997

Published in

Journal of Physics-Condensed Matter

[Link to publication](#)

#### Citation for published version (APA):

Kambe, S., Flouquet, J., Lejay, P., Haen, P., & de Visser, A. (1997). Thermal expansion and the Grueneisen parameter near the magnetic instability in  $Ce_{1-x}La_xRu_2Si_2$ . *Journal of Physics-Condensed Matter*, 9, 4917-4924. <https://doi.org/10.1088/0953-8984/9/23/015>

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# Thermal expansion and the Grüneisen parameter near the magnetic instability in $\text{Ce}_{1-x}\text{La}_x\text{Ru}_2\text{Si}_2$

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Received 3 January 1997

**Abstract.** The low-temperature thermal expansion of the heavy-fermion system with the formula  $\text{Ce}_{1-x}\text{La}_x\text{Ru}_2\text{Si}_2$  ( $x = 0$  and  $x = 0.05$ ) close to the magnetic instability is analysed in terms of the renormalization group and self-consistent renormalized spin-fluctuation models. The Grüneisen parameter calculated using the renormalized Fermi temperature is compared with the effective one which is determined from thermal expansion measurements and previously obtained specific heat data.

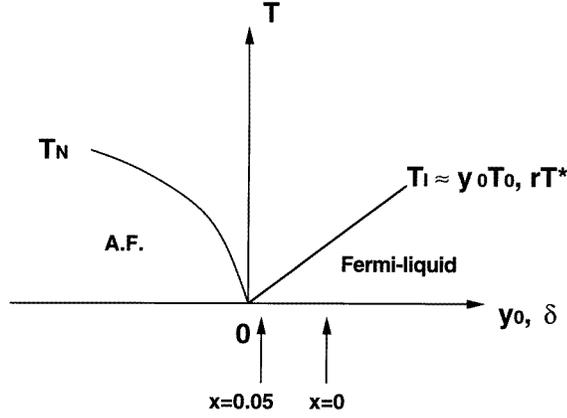
## 1. Introduction

Properties of metallic heavy-fermion systems near a magnetic instability have attracted strong interest, as non-Fermi-liquid behaviour has been expected when a magnetic–non-magnetic transition occurs at  $T \sim 0$  K. In particular,  $\text{Ce}_{1-x}\text{La}_x\text{Ru}_2\text{Si}_2$  [1],  $\text{CeCu}_{6-x}\text{Au}_x$  [2], and  $\text{Ce}(\text{Ni}_{1-x}\text{Cu}_x)_2\text{Ge}_2$  [3], around their critical concentration  $x_c$ , have been extensively studied, and theoretically several approaches have been proposed: the renormalization group (RG) model for quantum critical phenomena [4], the self-consistent renormalized spin-fluctuation (SCR) model [5], the disordered Anderson lattices model [6], etc. Recently [1], the RG and SCR models have been successfully compared with experiments (specific heat, resistivity, thermal expansion, NMR, and inelastic neutron scattering) on  $\text{Ce}_{1-x}\text{La}_x\text{Ru}_2\text{Si}_2$  for  $x = 0, 0.05$  and  $0.75$ . It was concluded that the so-called ‘non-Fermi-liquid behaviours’ can be understood as properties of an extended crossover regime to the Fermi-liquid ground state. It seems that experiments at very low temperature are necessary to distinguish a true non-Fermi-liquid ground state.

The aim of this study is to discuss the volume thermal expansion  $\alpha_V$  and the Grüneisen parameter  $\Omega$  of the renormalized Fermi temperature around the magnetic instability in  $\text{Ce}_{1-x}\text{La}_x\text{Ru}_2\text{Si}_2$ . The thermal expansion  $\alpha_V$  is sensitive to anomalous behaviours because it is a pressure-differential quantity. Thus a crucial comparison between theory and experiment can be expected for  $\alpha_V$ . The Grüneisen parameter is remarkable because it can show a

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**Figure 1.** The phase diagram for antiferromagnetic instability based on the SCR and RG models. The measure for the proximity of the magnetic instability is  $y_0$  for the SCR model and  $\delta$  for the RG model.  $T_I$  for the RG model is effectively  $\delta T^*$  ( $=rT^*$ ) in this study (see section 3.1).

divergent behaviour at the magnetic instability at  $T = 0$  K. Usually, this parameter is estimated experimentally using the specific heat and thermal expansion for convenience; a more precise framework is given based on the RG and SCR models.

In the  $\text{Ce}_{1-x}\text{La}_x\text{Ru}_2\text{Si}_2$  system, the magnetic–non-magnetic transition appears at around  $x_c = 0.08$  [1]. We have measured  $\alpha_V$  down to low temperatures ( $\sim 0.3$  K) and analysed  $\Omega$  for  $\text{Ce}_{0.95}\text{La}_{0.05}\text{Ru}_2\text{Si}_2$  ( $x = 0.05$ ) and  $\text{CeRu}_2\text{Si}_2$  ( $x = 0$ ), which are in the paramagnetic regime near the magnetic instability (figure 1). The experimental methods are described in reference [7].

## 2. The scaling form of the free energy, and the Grüneisen parameter

It is well known that if the free energy  $F$  is scaled through one parameter  $T_s$  as in the impurity Kondo model:

$$F = T \Phi(T/T_s) \quad (1)$$

then the effective Grüneisen parameter  $\Omega_{\alpha,C} \equiv V_m \alpha_V / \kappa C$  ( $V_m$ : unit volume;  $\kappa$ : compressibility  $-V^{-1} \partial V / \partial P$ ;  $C$ : specific heat) is constant, i.e.  $\alpha_V$  is proportional to  $C$ , and the Grüneisen parameter for  $T_s$ ,  $\Omega_{T_s} \equiv -\partial \ln T_s / \partial \ln V$ , is identical with  $\Omega_{\alpha,C}$ .

In the SCR model [5],  $F$  takes the scaling form

$$F_{SCR} = t \Phi_1(t, y_1, y_0) \quad (2)$$

where

$$t = \frac{T}{T_0}$$

and where  $y_1$  and  $y_0$  are dimensionless independent parameters which can be determined by fitting experimental quantities such as the specific heat. The parameter  $y_1 = 2J_Q / (T_A \pi^2)$  is connected with the RKKY exchange energy  $J_Q$ ,  $y_0$  is proportional to the inverse staggered susceptibility at  $T = 0$  K through the relation  $y_0 = 1 / (2T_A \chi_Q(0))$ ,  $T_0$  is defined as  $T_0 = T_A \Gamma_L \chi_L / \pi$  ( $\Gamma_L$ : local ( $q = 0$ ) spin-fluctuation energy;  $\chi_L$ : local spin susceptibility), and  $T_A$  is defined as  $T_A = A q_B^2 / 2$  ( $A$  is the strength of the RKKY dispersion around the

**Table 1.** Parameters for proximity to the magnetic instability [1], and the Grüneisen parameters for  $T_I$  for  $\text{Ce}_{1-x}\text{La}_x\text{Ru}_2\text{Si}_2$ .

(a) The SCR model		
	$x = 0$	$x = 0.05$
$y_0$	0.31	0.1
$y_1$	1.6	1.33
$T_0$ (K)	14.1	14.7
$T_I = y_0 T_0$ (K)	4.4	1.5
$\Omega_{T_I, SCR}$	$4.0 \times 10^2$	$8.0 \times 10^2$
(b) The RG model		
	$x = 0$	$x = 0.05$
$\delta$	0.6	0.27
$T^*$ (K)	12.6	12.6
$T_I = \delta T^*$ (K)	7.6	3.4
$\Omega_{T_I, RG}$	$1.0 \times 10^2$	$2.6 \times 10^2$

staggered wave vector  $q = Q$ :  $Aq^2 = J_Q - J_{Q+q}$ , and  $q_B$  is the effective zone-boundary vector).

In the RG model [4, 8],  $F$  takes the scaling form

$$F_{RG} = t^{5/2} \Phi_2 \left( \frac{r}{t}, \frac{ut^{3/2}}{r} \right) \quad (3a)$$

with

$$r = \delta + ut^{3/2} \Phi_{3,2} \left( \frac{r}{t} \right) \quad (3b)$$

$$t = \frac{T}{T^*}$$

where  $\delta$ ,  $T^*$ , and  $u$  are independent parameters. The parameter  $\delta$  is the control parameter  $r$  at 0 K (depending on tuning quantities such as the doping concentration or pressure),  $u$  is the coefficient of the dangerously irrelevant operator, and  $T^*$  is the microscopic characteristic temperature (roughly an effective Fermi temperature  $T_F$ ). The explicit forms of the scaling functions  $\Phi_1$ ,  $\Phi_2$ , and  $\Phi_{3,2}$  are given in references [5] and [8]. As shown in the phase diagram (figure 1), quantum critical phenomena are expected when  $y_0$  (for the SCR model) or  $\delta$  (for the RG model) reaches 0 at  $T = 0$  K. For  $\text{Ce}_{0.95}\text{La}_{0.05}\text{Ru}_2\text{Si}_2$  ( $x = 0.05$ ) and  $\text{CeRu}_2\text{Si}_2$  ( $x = 0$ ), the values of  $y_0$  and  $\delta$  obtained [1] from fits to specific heat data [9] are presented in table 1. In the case of antiferromagnetic interactions, the characteristic temperature for the Fermi-liquid region  $T_I$  is proportional to  $y_0 T_0$  for the SCR model and  $r T^*$  for the RG model. We focus on the Grüneisen parameter for  $T_I$ :  $\Omega_{T_I} \equiv -\partial \ln T_I / \partial \ln V$ .

The one-parameter scaling law is generally fulfilled neither in the framework of RG models nor in that of SCR models, because all of the parameters of the free energy can depend on the volume; thus theoretically  $\Omega_{\alpha, C}$  is not independent of the temperature, and  $\Omega_{T_I}$  differs from  $\Omega_{\alpha, C}$ , in agreement with an observation of a non-constant experimental  $\Omega_{\alpha, C}$  for  $\text{Ce}_{1-x}\text{La}_x\text{Ru}_2\text{Si}_2$ . Since the proximity to the instability depends strongly on the volume in heavy-fermion systems,  $y_0$  and  $\delta$  (measures of the proximity) will have a considerable volume dependence. However, the pressure dependence of  $T_0$ ,  $y_0$ ,  $y_1$ ,  $T^*$ ,  $\delta$ , and  $u$  is not yet clearly given in the two models, so some assumptions are necessary for calculating the  $T$ -dependence of  $\alpha_V$  which is no longer proportional to  $C$ .

### 3. Thermal expansion

#### 3.1. The RG model

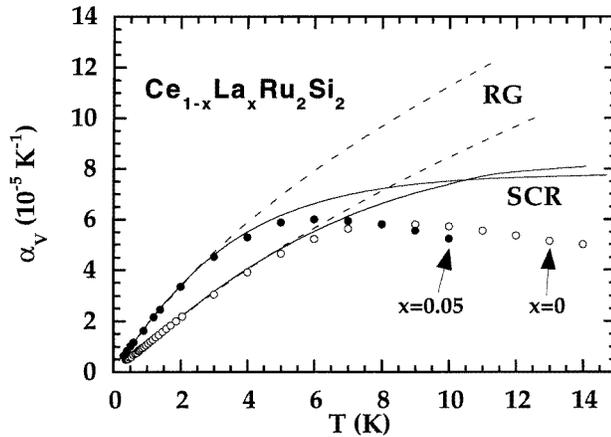
In the calculations of  $\alpha_V$ , we assume that  $u = 0$  (i.e.  $r = \delta$ ) and  $\partial T^*/\partial P = 0$ . It should be noted that  $\partial/\partial P$  means  $\partial/\partial P(P = 0 \text{ atm})$  here, and  $\alpha_V$  is calculated at  $P = 0 \text{ atm}$ . On the basis of the specific heat  $\gamma = C/T$  formula for the crossover region [8] and the thermodynamic relations for the entropy  $S$  ( $S = \int C/T \, dT$  and  $\alpha_V = -V^{-1} \partial S/\partial P$ ), the following equation is obtained:

$$\alpha_V = \frac{T^*}{2V_m} \frac{\partial r}{\partial P} I(r, t) \quad (4)$$

where

$$I(r, t) \equiv c \int_0^\infty d\varepsilon \int_0^1 dt \frac{\varepsilon^2}{\sinh^2 \varepsilon} t^{1/2} \left\{ \frac{r}{t} + \left( \left( \frac{r}{t} \right)^2 + 4\varepsilon^2 \right)^{1/2} \right\}^{-1/2} \\ \times \left\{ \frac{1}{t} + \frac{r}{t^2} \left( \left( \frac{r}{t} \right)^2 + 4\varepsilon^2 \right)^{-1/2} \right\}$$

where the same notation as in equation (3) is used;  $c$  is a constant related to the microscopic length, and  $r$  is related to  $x$  by  $r = \delta = (x_c - x)/x^*$  ( $x_c$  and  $x^*$  being the critical and scaling concentrations) [8]. The parameters  $c$ ,  $T^*$ ,  $x_c$ , and  $x^*$  are assumed to be independent of  $x$  around  $x_c$ , and the estimates used are those obtained previously [1] by fitting the specific heat data [9] down to 100 mK for the  $\text{Ce}_{1-x}\text{La}_x\text{Ru}_2\text{Si}_2$  series;  $c = 0.32 \text{ J K}^{-2}/\text{mol Ce}$ ,  $T^* = 12.6 \text{ K}$ ,  $x_c = 0.09$ , and  $x^* = 0.15$ .



**Figure 2.** Experimental values of the thermal expansion  $\alpha_V$  are compared for the RG and SCR models. Experiments: closed circles:  $x = 0.05$  [7]; open circles:  $x = 0$ . Fitting curves: solid lines: the SCR model; dashed lines: the RG model.

Since  $\partial T^*/\partial V = 0$ , the Grüneisen parameter for  $T_I (= \delta T^*)$ ,  $\Omega_{T_I, RG}$ , is proportional to  $\partial \delta/\partial P$ :

$$\Omega_{T_I, RG} = -\frac{V}{T_I} \frac{\partial T_I}{\partial V} = \frac{1}{\kappa \delta} \frac{\partial \delta}{\partial P}. \quad (5)$$

Now let us compare the RG model with experiments.  $I(r, t)$  can be calculated explicitly using the parameters estimated previously from the specific heat measurements, while  $\partial r/\partial P = \partial \delta/\partial P$  is a  $T$ -independent adjustable parameter in equation (4). If we optimize  $\partial \delta/\partial P$ , an agreement between the model and experiments for  $\alpha_V$  (figure 2) is obtained considerably below  $T_I (= \delta T^*)$  for both  $x = 0.05$  ( $T_I = 3.4$  K) and for  $x = 0$  ( $T_I = 7.6$  K). The deviation for  $T_I < T < T^*$  may reflect the simplicity of our assumptions, while above  $T^*$  the RG model does not describe the excitations correctly. The value of  $\Omega_{T_I, RG}$  is estimated as  $1.0 \times 10^2$  for  $x = 0$  and  $2.6 \times 10^2$  for  $x = 0.05$  from equation (5), using  $\kappa = 0.95 \times 10^{-11} \text{ m}^2 \text{ N}^{-1}$  and  $V_m = 5.19 \times 10^{-5} \text{ m}^3 \text{ mol}^{-1}$ . As expected,  $\Omega_{T_I, RG}$  increases as the magnetic instability is approached.

### 3.2. The SCR model

The characteristic quantities of the SCR model [5] are the renormalized inverse susceptibility at  $q = Q$ :  $y \equiv 1/(2T_A \chi_Q)$ , and the parameters in equation (2). Previously, the values of  $T_A$ ,  $T_0$ ,  $y_0$ , and  $y_1$ , and the  $T$ -dependence of  $y$  have been determined [1] for  $\text{Ce}_{1-x}\text{Ru}_x\text{La}_2\text{Si}_2$  from specific heat and susceptibility measurements [1, 9]. On the basis of these parameters,  $\alpha_V$  has been fitted with the SCR model [1] using the following relations [10]:

$$\begin{aligned} \omega_m &= \frac{\Delta V}{V} = \frac{3 D_Q \kappa}{5 F_s \chi_Q} \\ \alpha_V &= \frac{1}{V} \frac{dV}{dT} = \frac{d\omega_m}{dT} = 3 \frac{\pi^2 D_Q \kappa}{J_Q} \frac{dy}{dt} \end{aligned} \quad (6)$$

where  $D_Q$  and  $F_s (= 2J_Q T_A / 5\pi^2 T_0)$  are the magnetovolume constant at  $q = Q$  and the mode-mode coupling constant, respectively.

However, a different expression for  $\alpha_V$  can be deduced by integrating  $C/T$  from the SCR model [5]:

$$\frac{C}{T} = \frac{3}{\pi} \frac{\partial^2}{\partial T^2} \sum_q T \int_0^\infty d\lambda \frac{1}{e^\lambda - 1} \tan^{-1} \frac{\lambda T}{\Gamma_q} \quad (7)$$

where

$$\Gamma_q = 2\pi T_0 (y + x^2) \quad x = q/q_B$$

and

$$\alpha_V = -\frac{1}{V} \frac{\partial S}{\partial P} = -\frac{1}{V} \frac{\partial}{\partial P} \int_0^T \frac{C}{T} dT = \frac{6}{V_m T_0 y_1} \left( \frac{\partial^2 (T_0 y)}{\partial t \partial P} (y - y_0) + \frac{\partial (T_0 y)}{\partial P} \frac{dy}{dt} \right). \quad (8)$$

In the calculation of  $\alpha_V$ , we assume here  $\partial T_0/\partial P = 0$ , and that  $\partial y/\partial P$  is independent of the temperature, i.e.  $\partial y/\partial P = \partial y_0/\partial P$ . On these assumptions, the first term of equation (8) vanishes, leading to an expression similar to equation (6):

$$\alpha_V = \frac{6}{V_m y_1} \frac{\partial (y_0)}{\partial P} \frac{dy}{dt}. \quad (9)$$

The comparison of equations (6) and (9) leads to the following relation:

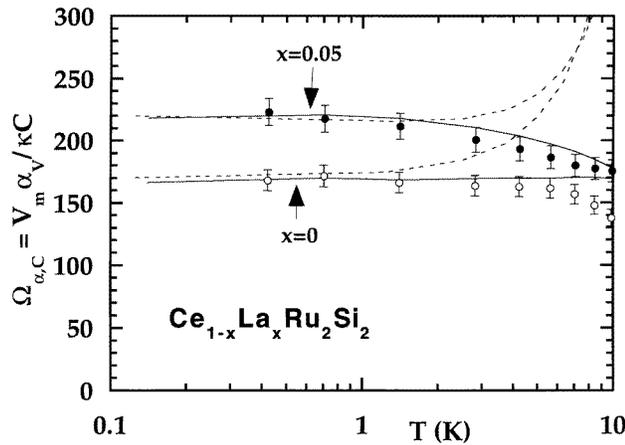
$$D_Q = -T_A \frac{\partial (y_0)}{\partial V}. \quad (10)$$

The Grüneisen parameter for  $T_I = y_0 T_0$ ,  $\Omega_{T_I, SCR}$ , is given by

$$\Omega_{T_I, SCR} \approx -\frac{V}{y_0} \frac{\partial (y_0)}{\partial V} = \frac{1}{\kappa y_0} \frac{\partial (y_0)}{\partial P}. \quad (11)$$

That is, the assumption of a  $T$ -independent  $\partial y/\partial P$  corresponds to the case of a  $T$ -independent  $\Omega_{T,SCR}$ .

The fit of  $\alpha_V$  to equation (9) is obtained using  $dy/dt$  determined from the specific heat data, and an optimized  $\partial y_0/\partial P$  [1]. As shown in figure 2, a good agreement with experiment is obtained below  $T_I$  ( $=y_0T_0$ : 1.5 K for  $x = 0.05$ , and 4.4 K for  $x = 0$  [1]). The corresponding  $\Omega_{T,SCR}$  is estimated as  $4.0 \times 10^2$  for  $x = 0$  and  $8.0 \times 10^2$  for  $x = 0.05$ . These values are somewhat larger than those of the RG model, because the proximity is regarded as stronger in the SCR model, i.e.  $y_0 < \delta$ . The better representation of the  $\alpha_V$ -data in the SCR approach than in the RG approach may reflect the differences in the respective assumptions made in the calculations of  $\alpha_V$ . Therefore it is difficult to decide on the validity of the two models at the present stage.



**Figure 3.** Comparisons of the effective Grüneisen parameter. Experiments: closed circles:  $x = 0.05$ ; open circles:  $x = 0$ . Calculated curves: solid lines: the SCR model; dashed lines: the RG model.

#### 4. Discussion

The present analysis treats only the intersite effect, since it dominates the thermal properties near the instability. The local characteristics such as  $T^*$  and  $T_0$  are assumed to be independent of pressure; however, these assumptions are rather imprecise because usually local (intrasite) properties such as the Kondo effect depend on the pressure and doping concentration. Obviously a model which includes valence fluctuations is necessary for treating a modification of the local characteristics. Although  $T_0$  has been found to be independent of  $x$ , the difficulty of the constant  $T^*$  for a wide range of  $x$  in  $\text{Ce}_{1-x}\text{La}_x\text{Ru}_2\text{Si}_2$  has already been mentioned [1], implying that the assumption that  $\partial T^*/\partial P = 0$  is suspect as compared to the assumption that  $\partial T_0/\partial P = 0$ . This difference may explain the better representation of  $\alpha_V$  in the SCR model.

In the SCR model [5], the pressure dependence of  $y_0$  is related to the resistivity  $\rho$ , since the Fermi-liquid-contribution term  $I$  in the resistivity ( $\rho = \rho_0 + IT^2$ ) varies as  $I \sim y_0^{-0.5}$  around the magnetic instability, and in addition a recent calculation [11] predicts a weaker  $y_0$ -dependence of  $I$  in an off-critical case like  $\text{CeRu}_2\text{Si}_2$ . However, the Grüneisen parameter  $\Omega_I \equiv -\partial \ln(I^{-1/2})/\partial \ln V$  for  $\text{CeRu}_2\text{Si}_2$  is found to be 180, and  $P$ -independent up to 6 kbar

[12], which leads to a stronger  $y_0$ -dependence of  $I$ :

$$I \sim y_0^{-2\Omega_I/\Omega_{T_i,SCR}} = y_0^{-0.9}. \quad (12)$$

The dominant mechanism here may be the variation of the local Kondo temperature, which will exponentially change with the pressure according to the linear shift of the 4f level relative to the Fermi level. Obviously more experiments in the vicinity of the critical point are needed.

Let us compare the experimental results with the calculated  $\Omega_{\alpha,C}$  using the values of  $\alpha_V$  determined (figure 3). As shown in figure 2, the behaviour of the calculated  $\Omega_{\alpha,C}$  at high temperatures is quite different for the two models; however, this feature depends strongly on the assumptions for  $T^*$ ,  $T_0$ ,  $\partial r/\partial P$ , and  $\partial y/\partial P$ . At low temperatures, the observed  $\alpha_V$  and  $\Omega_{\alpha,C}$  can be well fitted using the assumption of the finite  $T$ -independent  $\Omega_{T_i,RG}$  or  $\Omega_{T_i,SCR}$ , indicating that these compounds are still described by the Fermi-liquid picture in agreement with the previous analysis [1]. More theoretical investigations for the  $T$ -dependence of  $\partial r/\partial P$  and  $\partial y/\partial P$  are needed to discuss  $\alpha_V$  in the crossover regime.

When  $\delta$  or  $y_0$  approaches 0 as  $T \rightarrow 0$  K,  $\Omega_{T_i}$  and  $\Omega_{\alpha,C}$  are related through the following approximations respectively for the RG and SCR models:

$$\Omega_{\alpha,C} \approx n\delta^{1/2}(1 - (t/\delta)^2)^{-1}\Omega_{T_i,RG} \quad (13)$$

where

$$n = 6\gamma_0 T^* N^{-1} \approx O(1)$$

and

$$\Omega_{\alpha,C} \approx y_0 y^{-1/2} \Omega_{T_i,SCR} \quad (14)$$

where  $\gamma_0 = C/T$  ( $T \rightarrow 0$  K), and  $N$  is the number of components for the ordering field (the constant  $c$  is proportional to  $N$ ) [8]. Therefore, at the critical point,  $\Omega_{T_i} \sim y_0^{-1}$ ,  $\delta^{-1}$  and  $\Omega_{\alpha,C} \sim y_0^{-1/2}$ ,  $\delta^{-1/2}$  are expected to diverge, while  $C/T$  remains finite and  $\alpha_V \rightarrow 0$  as  $T \rightarrow 0$  K for both models. Further studies concentrating on the divergent behaviour of  $\Omega_{T_i}$  at the critical pressure or critical concentration at very low temperatures will be quite useful for clarifying the critical behaviours. It is expected that  $\Omega_{\alpha,C}$  will be quite sensitive to Kondo disorder, and it may be suspected that the presence of disorder could prevent the divergence of  $\Omega_{\alpha,C}$  at the magnetic instability. It is worth noting that the so-called metamagnetic phenomenon for  $\text{CeRu}_2\text{Si}_2$ , which is accompanied by a large volume effect, is strongly smeared out by the substitution of La or Ge [13]. Finally, it should be noted that the present considerations can be applied to ultrasound measurements such as were recently performed on  $\text{CeCu}_{6-x}\text{Au}_x$  [14].

## Acknowledgment

One of the authors (SK) thanks the Van der Waals–Zeeman Institute, University of Amsterdam, for kind hospitality.

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