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Thermal expansion and the Grüneisen parameter near the magnetic instability in $Ce_{1-x}La_xRu_2Si_2$

S Kambe[†]||, J Flouquet[†], P Lejay[‡], P Haen[‡] and A de Visser[§]

† Département de Recherche Fondamentale sur la Matière Condensée, SPSMS, CEA-CENG, 38054 Grenoble, France

‡ Centre de Recherches sur les Trés Basses Température¶, CNRS, 38042 Grenoble, France

 \S Van der Waals–Zeeman Institute, University of Amsterdam, 1018 XE Amsterdam, The Netherlands

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Abstract. The low-temperature thermal expansion of the heavy-fermion system with the formula $Ce_{1-x}La_xRu_2Si_2$ (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 α_V and the Grüneisen parameter Ω of the renormalized Fermi temperature around the magnetic instability in Ce_{1-x}La_xRu₂Si₂. The thermal expansion α_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 α_V . The Grüneisen parameter is remarkable because it can show a

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[∥] Author to whom any correspondence should be addressed: Shinsaku Kambe, DRFMC/SPSMS-LCP, CEA-CENG, 38054 Grenoble, France; telephone: +33-4-76889759; fax: +33-4-76885096; e-mail: kambe@drfmc.ceng.cea.fr; on leave from: Institute for Solid State Physics, University of Tokyo, Roppongi, Tokyo 106, Japan.

[¶] Laboratoire associé à l'Université Joseph Fourier, Grenoble, France.



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 δ for the RG model. T_I for the RG model is effectively δT^* (= 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 Ce_{1-x}La_xRu₂Si₂ system, the magnetic–non-magnetic transition appears at around $x_c = 0.08$ [1]. We have measured α_V down to low temperatures (~0.3 K) and analysed Ω for Ce_{0.95}La_{0.05}Ru₂Si₂ (x = 0.05) and CeRu₂Si₂ (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:

$$F = T\Phi(T/T_s) \tag{1}$$

then the effective Grüneisen parameter $\Omega_{\alpha,C} \equiv V_m \alpha_V / \kappa C$ (V_m : unit volume; κ : compressibility $-V^{-1} \partial V / \partial P$; *C*: specific heat) is constant, i.e. α_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

$$F_{SCR} = t \Phi_1(t, y_1, y_0)$$
(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 $T_0 = T_A\Gamma_L\chi_L/\pi$ (Γ_L : local (q = 0) spin-fluctuation energy; χ_L : local spin susceptibility), and T_A is defined as $T_A = Aq_B^2/2$ (A is the strength of the RKKY dispersion around the

| x = 0 | x = 0.05 |
|-------------------|--|
| 0.31 | 0.1 |
| 1.6 | 1.33 |
| 14.1 | 14.7 |
| 4.4 | 1.5 |
| 4.0×10^2 | 8.0×10^2 |
| | |
| x = 0 | x = 0.05 |
| 0.6 | 0.27 |
| 12.6 | 12.6 |
| 7.6 | 3.4 |
| 1.0×10^2 | 2.6×10^2 |
| | x = 0 0.31 1.6 14.1 4.4 4.0 × 10 ² x = 0 0.6 12.6 7.6 1.0 × 10 ² |

Table 1. Parameters for proximity to the magnetic instability [1], and the Grüneisen parameters for T_I for Ce_{1-x}La_xRu₂Si₂.

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

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

with

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

$$t = \frac{T}{T^*}$$
(3b)

where δ , T^* , and u are independent parameters. The parameter δ is the control parameter rat 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 Φ_1 , Φ_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 δ (for the RG model) reaches 0 at T = 0 K. For Ce_{0.95}La_{0.05}Ru₂Si₂ (x = 0.05) and CeRu₂Si₂ (x = 0), the values of y_0 and δ 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_0T_0 for the SCR model and rT^* 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 Ω_{T_I} differs from $\Omega_{\alpha,C}$, in agreement with an observation of a non-constant experimental $\Omega_{\alpha,C}$ for Ce_{1-x}La_xRu₂Si₂. Since the proximity to the instability depends strongly on the volume in heavy-fermion systems, y_0 and δ (measures of the proximity) will have a considerable volume dependence. However, the pressure dependence of T_0 , y_0 , y_1 , T^* , δ , and u is not yet clearly given in the two models, so some assumptions are necessary for calculating the T-dependence of α_V which is no longer proportional to C.

3. Thermal expansion

3.1. The RG model

In the calculations of α_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 α_V is calculated at P = 0 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:

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

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 Ce_{1-x}La_xRu₂Si₂ series; c = 0.32 J K⁻²/mol Ce, $T^* = 12.6$ K, $x_c = 0.09$, and $x^* = 0.15$.



Figure 2. Experimental values of the thermal expansion α_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 (= δT^*), $\Omega_{T_I,RG}$, is proportional to $\partial \delta / \partial P$:

$$\Omega_{T_I,RG} = -\frac{V}{T_I} \frac{\partial T_I}{\partial V} = \frac{1}{\kappa \delta} \frac{\partial \delta}{\partial P}.$$
(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 α_V (figure 2) is obtained considerably below T_I (= δT^*) 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 1.0×10^2 for x = 0 and 2.6×10^2 for x = 0.05 from equation (5), using $\kappa = 0.95 \times 10^{-11}$ m² N⁻¹ and $V_m = 5.19 \times 10^{-5}$ m³ 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 Ce_{1-x}Ru_xLa₂Si₂ from specific heat and susceptibility measurements [1, 9]. On the basis of these parameters, α_V has been fitted with the SCR model [1] using the following relations [10]:

$$\omega_m = \frac{\Delta V}{V} = \frac{3}{5} \frac{D_Q \kappa}{F_s \chi_Q}$$

$$\alpha_V = \frac{1}{V} \frac{\mathrm{d}V}{\mathrm{d}T} = \frac{\mathrm{d}\omega_m}{\mathrm{d}T} = 3 \frac{\pi^2 D_Q \kappa}{J_Q} \frac{\mathrm{d}y}{\mathrm{d}t}$$
(6)

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

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

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

where

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

and

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

In the calculation of α_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):

$$\alpha_V = \frac{6}{V_m y_1} \frac{\partial(y_0)}{\partial P} \frac{\mathrm{d}y}{\mathrm{d}t}.$$
(9)

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

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

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

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

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

The fit of α_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 $\Omega_{T_I,SCR}$ is estimated as 4.0×10^2 for x = 0 and 8.0×10^2 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 α_V -data in the SCR approach than in the RG approach may reflect the differences in the respective assumptions made in the calculations of α_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 Ce_{1-x}La_xRu₂Si₂ 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 α_V in the SCR model.

In the SCR model [5], the pressure dependence of y_0 is related to the resistivity ρ , 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 CeRu₂Si₂. However, the Grüneisen parameter $\Omega_I \equiv -\partial \ln(I^{-1/2})/\partial \ln V$ for CeRu₂Si₂ is found to be 180, and *P*-independent up to 6 kbar [12], which leads to a stronger y_0 -dependence of I:

$$I \sim y_0^{-2\Omega_I / \Omega_{T_I,SCR}} = y_0^{-0.9}.$$
 (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 α_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 α_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 α_V in the crossover regime.

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

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

where

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

and

$$\Omega_{\alpha,C} \approx y_0 y^{-1/2} \Omega_{T_I,SCR} \tag{14}$$

where $\gamma_0 = C/T$ ($T \to 0$ 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, $\Omega_{T_I} \sim y_0^{-1}$, δ^{-1} and $\Omega_{\alpha,C} \sim y_0^{-1/2}$, $\delta^{-1/2}$ are expected to diverge, while C/T remains finite and $\alpha_V \to 0$ as $T \to 0$ K for both models. Further studies concentrating on the divergent behaviour of Ω_{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 CeRu₂Si₂, 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 CeCu_{6-x}Au_x [14].

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