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A new scaling analysis of the susceptibility and the specific heat of the non-Fermi liquid system $Ce(Ru_{1-x}Rh_x)_2Si_2$ with x = 0.4 and 0.5

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Abstract. The usual scaling equations at a phase transition, employed out of their usual validity range, with T_c a negative constant, fit properties observed in correlated electron systems with unmatched accuracy. We illustrate this behavior with our data in $Ce(Ru_{1-x}Rh_x)_2Si_2$ for $x = 0.4$ and 0.5 and comment these results and what they imply physically.

PACS. 71.10.Hf Non-Fermi-liquid ground states, electron phase diagrams and phase transitions in model systems – 75.40.Cx Static properties (order parameter, static susceptibility, heat capacities, critical exponents, etc.) – 71.27.+a Strongly correlated electron systems; heavy fermions

The system $Ce(Ru_{1-x}Rh_x)_2Si_2$ has a very rich phase diagram which displays a variety of interesting behaviors [1]. The alloys with $x = 0.4$ and 0.5, in particular, are accepted examples of "non-Fermi liquid" behavior exhibiting such a characteristic signature as a logarithmic divergence of C_p/T when the temperature decreases to zero [2,3]. We have reanalyzed our susceptibility ($2\,\mathrm{K} < T < 300\,\mathrm{K}$) and specific-heat ($0.1\,\mathrm{K} < T < 17\,\mathrm{K}$) data in this system using the expressions

$$\xi = \xi_0 \left(1 - \frac{T_c}{T}\right)^{-\nu} = \xi_0 \left(1 - \frac{T_c}{T}\right)^{-\frac{\theta}{T_c}}$$
$$\chi T = C \left(1 - \frac{T_c}{T}\right)^{-\gamma} = C \left(1 - \frac{T_c}{T}\right)^{-\frac{\gamma\theta}{\nu T_c}}$$
$$C_p T^2 = A \left(1 - \frac{T_c}{T}\right)^{-\alpha} = A \left(1 - \frac{T_c}{T}\right)^{+\frac{d\theta}{T_c} - 2} \qquad (1)$$

proposed by Souletie [4] in an attempt to generalize to non singular situations ($T_c < 0$) the consequences of a renormalization argument which leads to the classical scaling theory in the case where T_c is positive. Here, $\xi(T)$, $\chi(T)$ and $C_p(T)$ are respectively the coherence length, the susceptibility and the specific heat. ξ_0 is an atomic distance, $C = Np_{eff}^2/3k = Ng^2\mu_B^2 S(S+1)/3k$ the usual Curie constant and $A/T_c^2 = (1-\alpha)(2-\alpha)R\ln(2S+1)$ follows if the total entropy between $T = 0$ and $T = \infty$ is $R\ln(2S+1)$. One has recognized the usual scaling relations for a phase transition with the usual definitions of their exponents ν, γ, and α: they will be used here outside of their traditional domain of application by permitting T_c to eventually assume negative values. The expressions in terms of $\theta = \nu T_c$ merely insure that for θ a positive constant

$$\ln(\xi/\xi_0) = -\frac{\theta}{T_c}\ln\left(1 - \frac{T_c}{T}\right) = \frac{\theta}{T} + \frac{\theta T_c}{2T^2} + \frac{\theta T_c^2}{3T^3} + \ldots \qquad (2)$$

is an increasing function of $1/T$ which has everywhere a well behaved expansion in terms of $1/T$ except at $1/T_c$, whatever the sign of T_c. The equation (2) shows that, in the Arrhenius representation of $\ln(\xi/\xi_0)$ *vs.* $1/T$ (see Fig. 1), the sign of T_c determines the curvature in such a way that the Arrhenius law of activation energy θ, which corresponds to the $T_c/T = 0$ limit, separates the singular solutions ($T_c > 0$) from the non-singular solutions: those whose singularity lies on the analytical continuation of the curve on the unphysical side of negative temperatures.

We believe that we should reserve the name of transition at $T_c = 0$ for the essential singularities which are obtained in the $T_c/T = 0$ limit

$$\xi(T) = \xi_0 \exp\left(\frac{\theta}{T}\right),$$
$$\chi(T) = \frac{C}{T}\exp\left(+\frac{\gamma\theta}{\nu T}\right)$$
$$C_p(T) = \frac{A}{T^2}\exp\left(-\frac{d\theta}{T}\right) \qquad (3)$$

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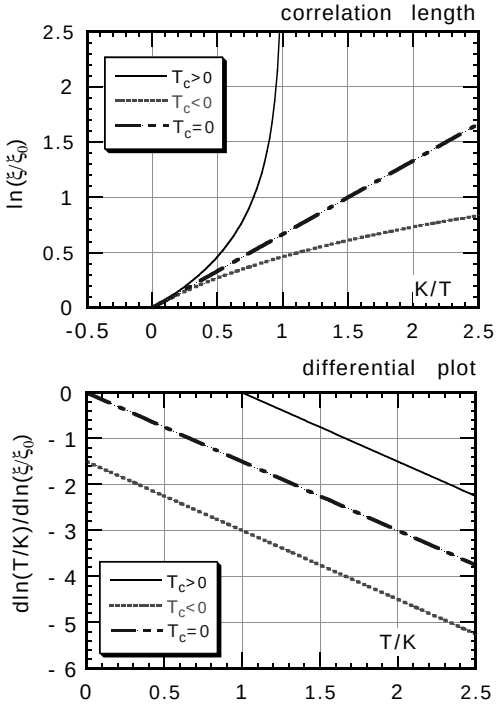

Fig. 1. $\ln(\xi/\xi_0) = (-\theta/T_c)\ln(1 - T_c/T)$ *vs.* $1/T$ and the corresponding $d\ln T/d\ln\xi$ *vs.* T diagram showing the typical information expected with T_c positive, nul or negative (Eq. (5)). We took $\theta = \nu T_c = 2/3$, with T_c 1, 0 and -1.

while the solutions where $T_c < 0$ ($T_c = -T_K$) and where a power law commands the low temperature behavior

$$\frac{\xi(T)}{\xi_0} = \left(1 + \frac{T_K}{T}\right)^{\theta/T_K} \rightarrow \left(\frac{T_K}{T}\right)^{\theta/T_K} \quad \text{when } T \rightarrow 0 \tag{4}$$

belong to paramagnetism. In order to check the validity of the equation (1) and if the sign of T_c is positive or negative, we recommend to determine first the differential form

$$\frac{d\ln T}{d\ln\xi} = -\frac{T - T_c}{\nu T_c} \tag{5}$$

where ξ_0 (or A or C) has been eliminated. By plotting $d\ln T/d\ln(\chi T)$ or $d\ln T/d\ln(C_p T^2)$ *vs.* T, straight lines are obtained, if the approximation is valid, which intersect the temperature axis at T_c and the ordinate at $1/\gamma$ or at $1/\alpha$ respectively (Fig. 1).

Figure 2 shows $d\ln T/d\ln((\chi - \chi_D)T)$ *vs.* T for $Ce(Ru_{1-x}Rh_x)_2Si_2$ with $x = 0.4$. The linear plot extends over all the range up to 180 K although the scatter increases at higher temperatures. The following equation

$$\chi - \chi_D = \chi_{th} = \frac{C}{T}\left(1 + \frac{T_K}{T}\right)^{-\gamma} \tag{6}$$

fits the data of the experimental susceptibility (see Fig. 3) for $x = 0.4$ and 0.5 up to 300 K with C and T_K given in Table 1 which are determined from Figure 2. χ_D was

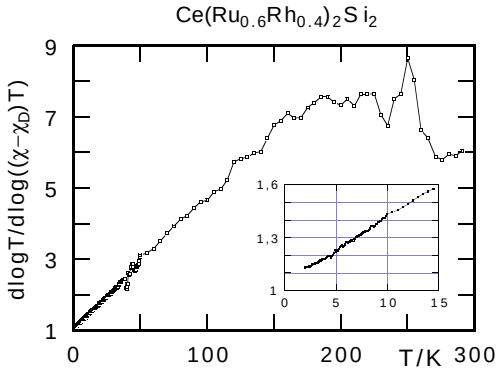

Fig. 2. $d\ln T/d\ln((\chi - \chi_D)T)$ *vs.* T below 300 K in the $x = 0.4$ alloy. Notice that a unique regime, with $T_c < 0$ and γ slightly smaller than one (see insert), describes the situation below 180 K. $\chi_D = -0.000\,85$ e.m.u. is a small diamagnetic correction attributed to the matrix.

introduced to account for a lattice contribution in addition to the contribution from the Ce atoms. Here χ_D is a small diamagnetic term much the same for $x = 0.4$ and 0.5 and for other concentrations x which we have measured. The incidence of this contribution is better seen at high temperatures where $\chi_D T$ dominates the temperature dependence of χT. On the other hand, χ_D is negligible at lower temperatures where C, T_K and γ are actually determined. The asymptotic Curie constant C is estimated to be 1.237 e.m.u. in the limit $T \rightarrow \infty$ and is consistent with an effective moment $p_{eff} = 3.15\,\mu_B$ which is larger than 2.5 μ_B the theoretical value for ionic Ce^{3+}. A fascinating point is the fact that our exponent differs from 1 by an amount which is small, but larger than the experimental error (see insert Fig. 2). As a result, our model proposes an expression for the susceptibility which actually diverges with a very small exponent

$$\chi - \chi_D = \frac{C}{T_K}\left(1 + \frac{T}{T_K}\right)^{-1+\epsilon}\left(\frac{T}{T_K}\right)^{-\epsilon} \tag{7}$$

instead of reaching a constant in the limit where $T = 0$ as would be the case with $\gamma = 1$.

Most of what we said for $(\chi - \chi_D)T$ can be repeated for $(C_p - C_{latt})T^2$. We approximate the contribution of the lattice by $C_{latt}/(\mathrm{J/molK}) = 6.07 \times 10^{-3}\ \mathrm{T/K} + 2.66 \times 10^{-4}\ (\mathrm{T/K})^3$ which fits the measured heat capacity of the non-magnetic isoelectronic system $La_2Ru_2Si_2$ in this range. For obvious reasons, though, the accuracy on $d\ln T/d\ln[(C_p - C_{latt})T^2]$ is not comparable to that obtained on the $d\ln T/d\ln[(\chi - \chi_D)T]$. It is sufficient however to observe that there is a linear regime for T <9 K with a (wide) range of possible values for T_K which includes 28 K deduced from the susceptibility and an equally wide range of acceptable values of α all close to 3 but actually slightly smaller than 3 (notice that the usual restriction α <1 which insures the finitude of the entropy when $T_c > 0$ does not hold anymore when $T_c < 0$ [4]). An optimization, restricted to the data below 9 K and with

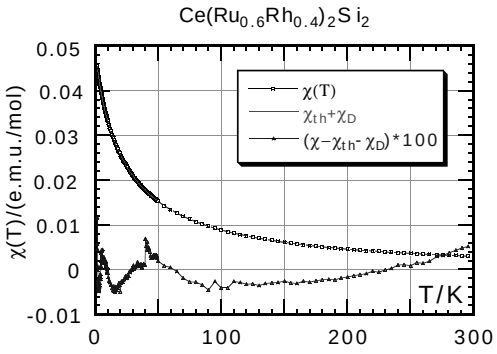

Fig. 3. The antiferromagnetic character of the interactions is insufficient to prevent the susceptibility to diverge but it diverges with a small exponent. The fit to equation (6) accounts for a small diamagnetic contribution $\chi_D = -0.000\,85$ e.m.u. of the lattice. The error is everywhere smaller than 1% of the smallest susceptibility measured.

Table 1. We show lines 1 and 2 for the $x = 0.4$ and the $x = 0.5$ alloys respectively the parameters C, γ, T_K and χ_D derived from our fit to the susceptibility data and the additional two parameters A and α deduced from our fit to the specific heat (columns 1 to 7).

x	$\frac{C}{\text{e.m.u./mol}}$	γ	$\frac{T_K}{K}$	$\frac{\chi_D}{\text{e.m.u./mol}}$	$\frac{A}{\text{J K/mol}}$	α
0.4	1.24	0.96	28.4	−0.00085	9906	2.86
0.5	1.19	0.89	29.5	−0.00065	8310	2.75

$T_K = 28$ K imposed, leads to the following expression (see Fig. 4):

$$C_{th}(T) = \frac{A}{T^2}\left(1 + \frac{28\ K}{T}\right)^{-\alpha} \tag{8}$$

with A and α given in Table 1. From the value of A we calculate an asymptotic value for the entropy between $T = 0$ and $T = \infty$ of $1.33R\ln 2$. At the same time the difference $C_p - C_{latt} - C_{th}$ becomes sizeable over 9 K giving some support to the speculation that there might be a second anomaly of magnetic origin, centered at higher temperature and whose position is determined by the splitting of the crystalline field as is often the case with cerium.

We have already stressed that the exponent α of C_pT^2 in equation (8) is very close to 3 but still smaller than 3. For this reason C_p/T diverges with a very small exponent instead of going to a constant:

$$\frac{C_p(T)}{T} = \frac{A}{T_K^3}\left(1 + \frac{T}{T_K}\right)^{-3+\epsilon'}\left(\frac{T}{T_K}\right)^{-\epsilon'} = \frac{A}{T_K^3}\left(1 + \frac{T}{T_K}\right)^{-3+\epsilon'}\exp\left(-\epsilon'\ln\frac{T}{T_K}\right). \tag{9}$$

Because ϵ' is so small, the exponential in the second member can be developed as $1 + \epsilon'\ln T_K - \epsilon'\ln T$ over a wide range of temperatures: actually if $\ln(T/T_K) \ll 1/\epsilon'$. This

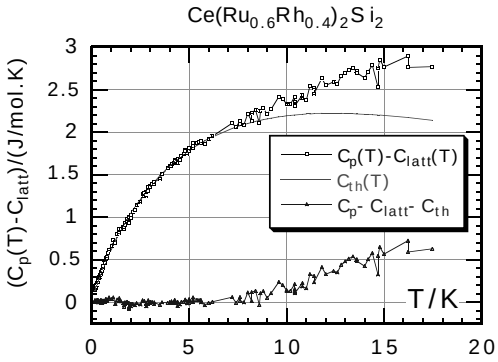

Fig. 4. The magnetic contribution to the specific heat below 9 K is fitted by the equation $C_{th}(T) = (A/T^2)\,(1 + (28\text{ K})/T)^{-\alpha}$. The amplitude of A with $\alpha = 2.86$ (Tab. 1) leads to an entropy $S = 1.33\,R\ln 2$ between $T = 0$ and T_∞. The excess contribution over 10 K signals, possibly, a second magnetic anomaly, centered at a higher temperature as is usual with Ce.

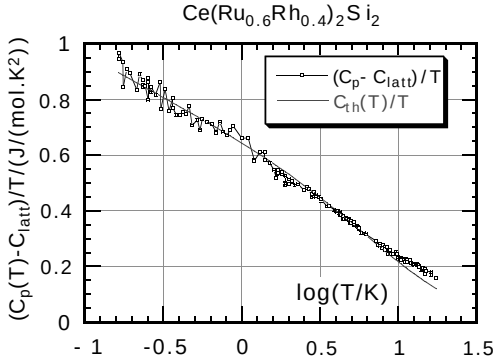

Fig. 5. The success of the semilogarithmic representation of C_p/T *vs.* $\ln T$ results from the fact that $C_p(T)/T$ diverges with a very small exponent (Eq. (9)) because the exponent α of C_pT^2 is very close to 3.

is the reason for the success of the semilog representation which is currently considered as the signature of the "non-Fermi liquid" behavior (see Fig. 5). As for $T_K = 28$ K which is our unique temperature scale it is connected by our model to the temperature T_{max} of the maximum of the specific heat by the relation $T_K = 2T_{max}/(\alpha - 2)$. It is therefore related to the usual Kondo temperature $T_K^* = 2.2T_{max}$ by the equation $T_K^* = 1.1(\alpha - 2)T_K$.

Our approach has been motivated by the simple idea that the coherence length could be constructed by steps in a hierarchical way, with the same recipe relating one stage with the other of the renormalization procedure. Then, the following recipe

$$\xi_{n+1} = a\xi_n$$
$$\frac{J}{T_{n+1}} - \frac{J}{T_c} = a^{-\frac{1}{\nu}}\left(\frac{J}{T_n} - \frac{J}{T_c}\right) \tag{10}$$

leads, if we iterate, to equation (1) and to critical scaling if $T_c > 0$. But if we look for non-singular solutions we have no reason to ignore those which are generated if $T_c < 0$.

These solutions fit our experiments with the $x = 0.4$ and 0.5 samples down to $T_K/10$ for the susceptibility and to $T_K/100$ for the specific heat with an accuracy such that any low temperature trend which would eventually drive χ and C_p/T to a constant, would appear as the result of a crossover induced by new circumstances. In the insert of Figure 2 the data below 2 K would then align on a distinct line aiming 1 at $T = 0$ and which we would associate with the new regime. For ϵ and ϵ' finite this raises a difficulty as we predict a divergence of both χ and C/T in distinction with the current renormalization group theory [5] which predicts that both quantities reach a finite limit when T cancels in 3d-antiferromagnetic itinerant fermion systems even in non-Fermi liquid state.

This leads us to the most intriguing question perhaps that this analysis raises: why do we have these special values of the exponent like $\gamma = 1$ and $\alpha = 3$ and why not exactly these values? If it is confirmed that some particular values exist this opens the possibility to create a classification, based upon numbers, of subtle qualitative differences which are reported in these systems. Would this mean that we have "universality classes" when $\xi(T)$ diverges as this is the case with standard phase transitions in the vicinity of T_c? As for the reasons why our exponents are close to but distinct from integers ... ? Like with the phase transitions, the problem of disorder and of its pertinence will have to be raised, specially because these behaviours are often observed in alloys as this is the case in this paper. Would some Harris criterion maintain us apart from plain universal behaviour? We notice, in support of this interpretation that the ϵ are not the same for $x = 0.4$ and $x = 0.5$ as if the disorder was introducing some kind of distortion of the dimensionality. Notice that if ϵ and ϵ' cancel (in the limit of zero disorder for example), our model predicts a constant $T = 0$ limit in agreement with the renormalization group theory [5]: the disagreement however persists at higher order because the theory predicts deviations to this constant limit which are proportional to $T^{1/4}$ for χ and to $T^{1/2}$ for C/T (for $z = 2$ at $d = 3$) while our expansions (Eqs. (7, 9)) are analytical in T.

Because the equations (1) are exactly the same scaling equations than in critical scaling, it is difficult to ignore some problems that this discussion may arise among phase transition experts. The great extension of the temperature range itself, may appear as a surprise to many as it is generally considered that the validity of the equations (1) is restricted to a narrow "critical regime" near T_c in the case where T_c is positive. We want to stress again that, often, the appreciation of what the extension of this range should be is based upon an artefact: people reason that since we are to work close to T_c, it is acceptable to identify T with T_c when it is not in a difference $T - T_c$; they thus artificially generate a regime near T_c where it becomes necessary to stay. If one uses for example $(T-T_c)^{-\gamma}$ rather than equation (1), one loses the high temperature Curie law. Equation (1), by contrast, works well up to the highest temperatures and has in particular the correct Curie-Weiss limit $C/(T - \theta_{CW})$ with $\theta_{CW} = \gamma T_c$ [6,7].

We have been nevertheless surprised by the Figure 2 and the fact that a unique regime is obtained between 2 K and 180 K. Not only we do not see any sign of a critical regime but neither do we see any sign of a "Griffith's temperature" over which the Curie law is strictly obeyed because, say, the temperature is larger than the largest interaction and the system is not capable to sustain any correlation at any range. Even the flattening observed over 180 K for both samples, if we are to take it seriously, is not the sign of the Curie law. It is the sign of a power law $\chi T \sim AT^{1/7.5} = AT^{0.13}$ and the power which governs the divergence of the susceptibility markedly differs from one. By the way, one astonishing fact perhaps in these systems is that the Curie law remains an asymptotic limit. Nowhere at finite temperature, it is actually observed.

Of course, it would be extremely interesting to know if this low temperature scaling which we have described in non-Fermi systems signals or not the universal "cross over" towards Fermi-liquid behaviour which some models anticipate [8]. In order to obtain some answer we have extended our analysis to other cases which show standard Fermi-liquid behaviour. Preliminary results are shown in reference [9]. A complete discussion is under preparation.

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