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GRÜNEISEN PARAMETERS OF HEAVY FERMION SYSTEMS

A. de VISSER[a], J.J.M. FRANSE[a], A. LACERDA[b], P. HAEN[b] and J. FLOUQUET[b]
[a]*Natuurkundig Laboratorium der Universiteit van Amsterdam, Valckenierstraat 65, NL-1018 XE Amsterdam, The Netherlands*
[b]*Centre de Recherches sur les Très Basses Températures, CNRS, BP 166X, F-38042, Grenoble Cedex, France*

We discuss recent thermal expansion measurements on single crystalline samples of the heavy fermion compounds $CeCu_6$, $CeRu_2Si_2$, URu_2Si_2, UBe_{13} and UPt_3, and compare the results with existing specific heat data. The resulting Grüneisen parameters ($\Gamma = \alpha_v V_m/\kappa c$) are anomalously large at low temperatures and vary rapidly with temperature. This implies that a single energy parameter can describe the electron–electron interaction only at very low temperatures.

An important issue in the research on heavy fermion systems is the possible existence of a universal low temperature energy scale [1]. In the Kondo lattice picture at least two characteristic temperatures are observed experimentally: (i) the single ion Kondo temperature T_K(~100 K) and (ii) the coherence temperature T_{coh} (of the order of a few K), below which the electron subsystem enters the true Fermi liquid regime ($T \ll T_{coh}$). But also antiferromagnetic intersite correlations (~10 K) and crystal field effects (~100 K) play an important role in heavy fermion systems. Nevertheless, one may expect that in the low temperature limit ($T \to 0$), only one universal energy scale prevails.

An elegant way to probe the presence of such an energy scale is by means of thermodynamic Grüneisen parameters. Assuming that the low temperature entropy term can be written as $S = S(T/T^*(V))$, where $T^*(V)$ is the volume dependent characteristic temperature of the heavy electron liquid, we may define a heavy fermion Grüneisen parameter for $T \to 0$ [2]

$$\Gamma_{hf} = \frac{\alpha_v V_m}{\kappa c} = \frac{3 V_m a}{\kappa \gamma} = -\frac{\partial \ln T^*}{\partial \ln V}, \tag{1}$$

where α_v is the coefficient of volume expansion, c is the molar specific heat, V_m is the molar volume and κ is the isothermal compressibility. As $T \to 0$, we have retained only the linear terms: $c = \gamma T$ and $\alpha_v = 3aT$. Experimentally, it is convenient to define an effective (temperature dependent) Grüneisen parameter

$$\Gamma_{eff}(T) = \frac{\alpha_v(T) V_m}{\kappa c(T)}. \tag{2}$$

One then expects that $\Gamma_{eff}(T)$ levels off at low temperatures and becomes a constant below the critical temperature T^*, where $\Gamma_{eff} = \Gamma_{hf}$.

The aim of this paper is to present thermal expansion data for the heavy fermion systems $CeCu_6$, $CeRu_2Si_2$, URu_2Si_2, UBe_{13} and UPt_3. Combining $\alpha_v(T)$ with existing specific heat data we have calculated the Grüneisen parameters.

The thermal expansion measurements have been performed on single crystalline samples (dimension 2–5 mm), using a sensitive capacitance dilatometer, with a detection limit $\Delta L/L \sim 10^{-8}$. For the non-cubic heavy fermion compounds the coefficients of linear thermal expansion were found to be strongly anisotropic. In this paper we concentrate on the volume effects: $\alpha_v = \alpha_a + \alpha_b + \alpha_c$. The data ($T > 1.5$ K) for non-ordering $CeCu_6$ [3] and $CeRu_2Si_2$ [4, 5], superconducting UBe_{13} ($T_s = 1$ K) [6] and UPt_3 ($T_s = 0.5$ K) [7], and the antiferromagnetic superconductor URu_2Si_2 ($T_s = 1$ K, $T_N = 17.5$ K) [8] are reproduced in fig. 1. As follows from fig. 1 all compounds have a large α_v as the formation of the heavy electron bands sets in. The positive α_v implies that the ground state volume is reduced by the formation of the quasiparticle bands. Qualitatively this can be explained from the fact that the high temperature localized electrons become itinerant at low temperatures and thus contribute to the binding.

Next we combine the thermal expansion data with specific heat data taken from the literature ($CeCu_6$ [9], $CeRu_2Si_2$ [10], UBe_{13} [11], UPt_3 [12]). The deduced Grüneisen parameters are shown in fig. 2. For $CeCu_6$ and $CeRu_2Si_2$ the phonon contribution to Γ_{eff} has been

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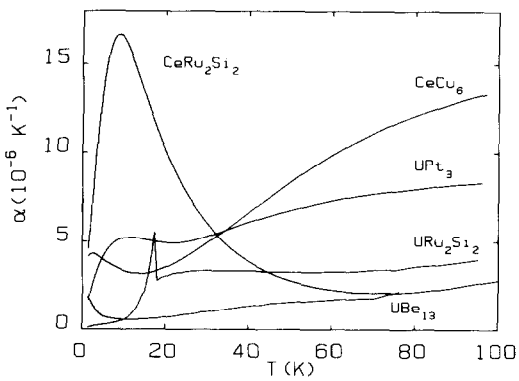

Fig. 1. Coefficient of volume expansion, α_v, as function of temperature for some heavy fermion compounds.

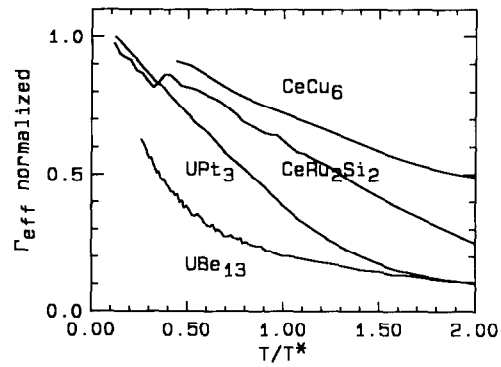

Fig. 2. Grüneisen parameter normalized to 0 K, as function of the reduced temperature T/T^* (see text). In the case of the Ce compounds, the phonon contribution of Γ_{eff} has been separated out.

separated out, whereas for UBe_{13} and UPt_3 this is not the case, since the phonon part of α_v is not known. However, the phonon contribution to Γ_{eff} can readily be neglected at low temperatures. Note, furthermore, that only in the case of $CeCu_6$ and UPt_3, $\alpha_v(T)$ and $c(T)$ have been obtained on the same sample. The data in fig. 2 show that Γ_{eff} varies rapidly with temperature. When extrapolated to 0 K (from temperatures above 1.5 K) we find values for Γ_{hf} of 57 ($CeCu_6$), 160 ($CeRu_2Si_2$; see also ref. [13]), 25 (URu_2Si_2), 60 (UBe_{13}) and 71 (UPt_3; see also ref. [13]). In order to look for a universal behaviour of $\Gamma_{eff}(T)$, we have plotted Γ_{eff}/Γ_{hf} as function of T/T^*, where we took, tentatively, $T^* = 0.68R/\gamma$ (= T_K in the model of Andrei et al. [14]; R is the gas constant). Values for T^* amount to 3.4 ($CeCu_6$), 17.1 ($CeRu_2Si_2$), 5.1 (UBe_{13}) and 13.1 K (UPt_3). As follows from fig. 2, Γ_{eff} still increases, even at the lowest temperatures investigated, and has an almost linear temperature dependence up to $\sim T^*$, except for UBe_{13}. In the latter case, however, it is very difficult to determine (the normal state) γ, as superconductivity sets in at 1 K. The choice $\gamma = 1100$ mJ/mol K^2 might lead to a too large value for T^*. From fig. 2 we conclude that measurements below $T = 1.5$ K are needed, for all compounds, in order to find a temperature independent Γ_{eff}.

Ideal systems to investigate the presence of a single energy scale are $CeCu_6$ and $CeRu_2Si_2$, because these systems do not show order phenomena, as followed from investigations down to 20 mK. In fig. 3 we present thermal expansion data of $CeCu_6$, taken in a dilution refrigerator in the temperature interval 20 mK $< T <$ 1500 mK, on the same sample as used for the high temperature data ($T > 1.5$ K) [3]. The agreement between α_c in fig. 3 and previous data [3] is perfect, however, the present values for α_a and α_b differ somewhat from previous data, leading to a 20% larger value for α_v at 1.5 K. This discrepancy is probably connected with irreversibilities observed in $\alpha_a(T)$ [3], but might also be related to the orthorhombic–monoclinic phase transition at 220 K [15]. Furthermore, the data in fig. 3 have not been corrected for the cell effect (i.e. signal of the dilatometer with a copper sample). The extrapolation from $T > 1.5$ K suggests that this correction increases with decreasing temperature, and might become of the order of several times 10^{-7} K^{-1} in the low temperature limit. The data in fig. 3 show that the thermal expansion of $CeCu_6$ is strongly anisotropic (see ref. [3] for a discussion). Rather weak extrema are observed at 0.5 and 0.7 K, for the a- and b-axis, respectively. In fig. 4 we

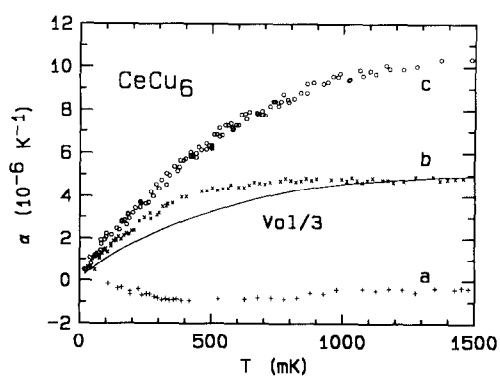

Fig. 3. Coefficients of linear expansion of $CeCu_6$ along the a (+), b (×) and c (○) axis. The solid line represents $\alpha_v/3$.

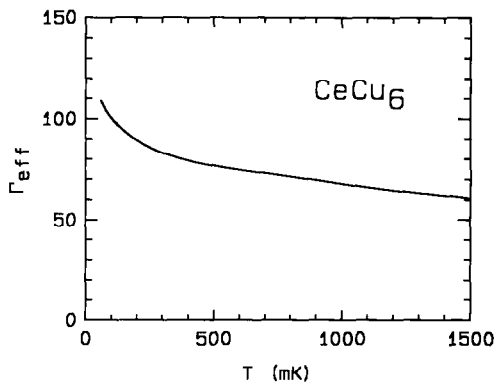

Fig. 4. Effective Grüneisen parameter ot $CeCu_6$ as function of temperature.

have plotted $\Gamma_{\rm eff}(T)$ for $T < 1.5$ K, using the specific heat data of Amato et al. [9]. Note that due to the fore-mentioned larger value of α_v, we obtain a correspondingly larger value of $\Gamma_{\rm eff}$. For $1\,{\rm K} < T < 1.5\,{\rm K}$ $\Gamma_{\rm eff}$ increases linearly with temperature and extrapolates to a value of 80 ($T \rightarrow 0$ K). Below 1 K $\Gamma_{\rm eff}$ does not level off, but rises again. The low temperature upturn must partly be ascribed to the cell effect, although we cannot exclude that another energy scale is entered at very low temperatures. It is interesting to note that Hall constant data [16] have been interpreted as revealing two low temperature energy scales, in the order of several hundreds and several tens of a mK. Further support for a second low temperature energy scale comes from the thermopower measurements [9]. From fig. 3 and the specific heat data [9] it follows that the linear terms are attained only below 100 mK, leading to a $\Gamma_{\rm hf}$ of 130.

Another heavy fermion compound for which low temperature thermal expansion data exist is $CeAl_3$ [17]. Here $\Gamma_{\rm eff}$ amounts to 25 at 1 K and then drops rapidly to about -200 at 100 mK, which was long thought to be a sign of coherence in the Kondo lattice. Extrapolation to zero temperature of our data in fig. 1 does not suggest a negative $\Gamma_{\rm eff}$ at low temperatures for the investigated heavy fermion systems. Therefore, a negative $\Gamma_{\rm eff}$ is not necessarily a sign of coherence, but is likely related with sharp features in the band structure, that lead to an initial increase of the density of states with pressure. However, more likely is that the negative $\Gamma_{\rm eff}$ in $CeAl_3$ is a consequence of the recently discovered long range magnetic order below 1.2 K [18], as is also suggested by recent thermal expansion data on doped ordered $U(Pt, Pd)_3$ [19] and $(Ce, La)Ru_2Si_2$ [20].

In conclusion, we have investigated the presence of a universal low temperature energy scale in the heavy fermion systems $CeCu_6$, $CeRu_2Si_2$, UBe_{13} and UPt_3, by means of thermodynamic Grüneisen parameters. The variation of $\Gamma_{\rm eff}$ with T suggests that a single energy parameter, describing the heavy electron liquid, might be present only at very low temperatures ($T < 100$ mK).

Acknowledgements

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