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# ANALYSIS OF THERMODYNAMIC PROPERTIES OF $UPt_3$ BY MEANS OF GRUNEISEN RELATIONS

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Specific heat and thermal expansion data of $UPt_3$ are analyzed by means of Grüneisen relations over a wide temperature range. Two contributions to both quantities are distinguished: a heavy-fermion contribution with a large value of 73 for the corresponding Grüneisen parameter and an associated entropy of $R$ ln 2, and a second contribution that is dominated by phonons.

The intermetallic compound $UPt_3$ belongs to the small group of heavy-fermion superconductors. Amongst the $UX_3$-compounds, with X a 4d- or 5d-metal or an element from group III or IV, $UPt_3$ is the only compound that crystallizes in the hexagonal $MgCd_3$-type of structure. Due to the absence of a non-magnetic analog system of $UPt_3$ the analysis of its properties is hampered. Usually, the analysis of specific heat or resistivity data, for instance, starts with eliminating the contribution connected with phonons by subtracting the data as measured on the non-magnetic analog system.

For $UPt_3$, however, the phonon contribution to the specific heat can only be determined by detailed measurements of phonon dispersion curves. This has been done by Renker et al. [1] by means of inelastic neutron-scattering experiments. Felten [2] used this information to determine from the specific heat that part that has an electronic origin and proposed in the low-temperature region two different electronic contributions. The first one, that leads to the high $\gamma$-value characteristic for heavy-fermions, is associated with the Kondo-effect. Within a single-ion $S = 1/2$ Kondo model one deduces $T_K = 13.5$ K from the $\gamma$-value. The second one, with a peak at 23 K, is of the Schottky-type connected with electronic levels that are separated by roughly 50 K.

In the present paper we follow an entirely different line in analyzing the specific heat by means of Grüneisen relations.

Physically meaningful Grüneisen relations emerge when a part of the entropy can be written as $S_i = S_i(T/T_i(V))$, where $T_i(V)$ is a (volume dependent) characteristic temperature. The dimensionless Grüneisen parameter is defined as $\Gamma_i = -\partial \ln(T_i(V))/\partial \ln V$. Employing thermodynamic relations, one can express $\Gamma_i$ in the linear thermal expansion coefficient $\alpha_i$ and the specific heat $c_i$ connected with the entropy part $S_i$ : $\Gamma_i = 3V_m\alpha_i/\kappa c_i$, where $\kappa$ is the compressibility and $V_m$ the molar volume. In general, several processes contribute to $c$ and $\alpha$, leading to an effective Grüneisen parameter: $\Gamma_{eff} = 3V_m\alpha/\kappa c = \Sigma\Gamma_i c_i/c$. In contrast to the $\Gamma_i$, $\Gamma_{eff}$ is, in general, temperature dependent and can not be related to the volume derivative of a single characteristic temperature. In case only two contributions are present in $c$ and $\alpha$, the combination of the $c$- and $\alpha$-data allows the splitting of both quantities into the two original contributions, as has been shown for different systems [3,4].

To start our analysis we have used the $c$ and $\alpha = (\alpha_a + \alpha_b + \alpha_c)/3$ values reported by de Visser et al. (see for a compilation [5]) below 32 K to calculate $\Gamma_{eff}$, see fig. 1. The strong temperature dependence suggests that at least two contribu-

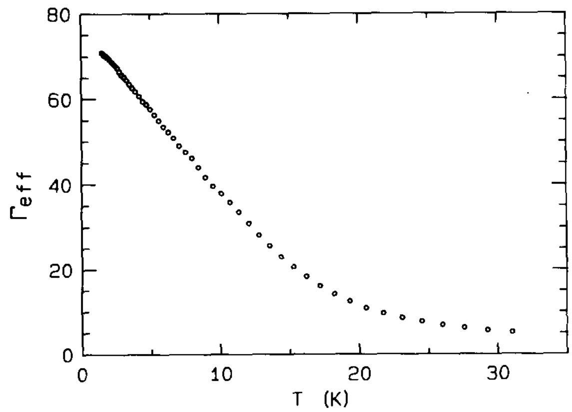

Fig. 1. Temperature dependence of $\Gamma_{eff} = 3V_m\alpha/\kappa c$.

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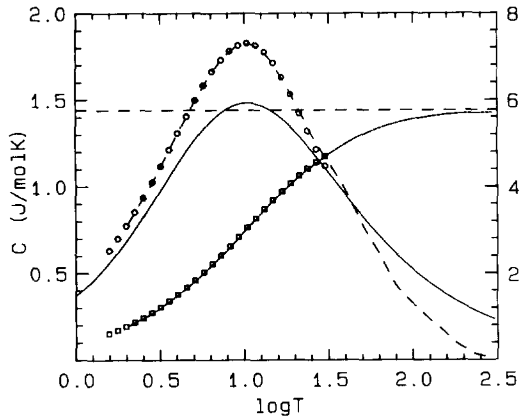

Fig. 2. Part of the specific heat and entropy that is associated with $\Gamma_1 = 73$. The circles and the dashed line through the circles represent results originating from the low- and high-temperature data, respectively. The full line shows the theoretical $S = 1/2$ single-ion Kondo peak with $T_K = 15.5$ K. The squares show the entropy originating from the low temperature data. The entropy at high temperatures (as denoted by the line through the squares) shows that the $R$ ln 2-level (indicated by the horizontal dashed line) is approached.

tions with different $\Gamma$-values are present. By extrapolating $\Gamma_{\mathrm{eff}}$ to 0 K, we derive for one of the Grüneisen parameters the value of 73, whereas for the second one we take a standard value of approximately 2, in this case 2.35. Using $\Gamma_1 = 73$ and $\Gamma_2 = 2.35$ we can split the specific heat into two contributions (fig. 2 and 3). The second contribution is dominated by the phonon term, whereas the first contribution with the exceptionally large $\Gamma$-value is thought to be related to the heavy-fermion behaviour. Evaluating the entropy of $c_1$ we notice that a value of $R$ ln 2 is approached whereas $c_1$ plotted vs. log $T$ reminds to a single-ion Kondo contribution. For this purpose the theoretical $S = 1/2$ Kondo-peak [6] with $T_K = 15.5$ K is also plotted in fig. 2; this $T_K$-value was taken to get the theoretical peak at the peak position of $c_1$. We note that the term $c_1$ in fig. 2 deviates from a spin 1/2 Kondo term by its enhanced maximal value and its reduced width. In fig. 3 we compare $c_2$ with the phonon contribution as deduced from the neutron data of Renker. It is clear that there is an additional contribution which, in the interpretation scheme of Felten, is associated with a crystal field contribution. The present analysis, however, does not confirm the crystal field nature of this contribution since the shape does not resemble a standard Schottky curve.

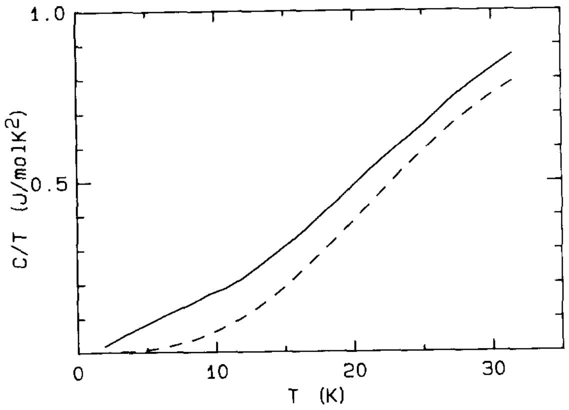

Fig. 3. Part of the specific heat that is associated with $\Gamma_2 = 2.35$ (full line). The dashed line shows the phonon contribution.

The analysis can be extended by combining high-temperature data measured on different samples by different groups (i.e. the specific heat data of Felten and the thermal expansion data of van Sprang [7]). In this case we analyse the data after subtracting the phonon contribution. In case of the thermal expansion, the phonon contribution has been determined by means of the Debye-function with $\theta_D = 200$ K and $\Gamma_{\mathrm{ph}} = 2.35$. Again splitting the remaining terms, with in this case values for the Grüneisen parameters of 73 and 2 (a common value for the normal electron gas), results in the high-temperature part of $c_1$ (fig. 2). We notice that in the overlapping temperature region, the results coincide with data deduced from the measurements below 32 K. The entropy of the first term also coincides with our previous analysis and almost exactly approaches a value of $R$ ln 2 near room temperature.

The present analysis of the specific heat deviates from our previous approach in which a spin-fluctuation model was employed including a $T^3 \ln T/T^*$ term in the temperature range up to 20 K. As earlier remarked, the spin-fluctuation description could be extended to temperatures close to the characteristic temperature $T^*$, whereas theory justifies this logarithmic term only for temperatures much smaller than $T^*$. For that reason we did not exclude in previous publications [8] that the good quality of the specific heat fit with the $T^3 \ln T/T^*$ term up to such high temperatures is accidental. In the low-temperature range (below 5 K), however, the term $c_1$ can still be represented by the sum $\gamma^* + \delta T^3 \ln T/T^*$ with values for $\gamma^*$ of 433 (422) mJ/molK$^2$, for $\delta$ of 5.88 (1.38) mJ/molK$^4$ and for $T^*$ of 12.4 (27) K, where numbers between brackets refer to the previous analysis in the temperature region below 20 K [5].

A few more points are worth mentioning. The large value for $\Gamma$, characteristic for the heavy-fermion state, can also be derived from the volume derivative of other parameters. Some of these are evaluated at elevated temperatures, like $\Gamma_1 = -\partial \ln T_{\max}/\partial \ln V = 63 \pm 13$ where $T_{\max}$ is the temperature ( ~ 17 K) where the susceptibility has a maximum. Apparently, the Grüneisen parameter $\Gamma_1$ is not changing rapidly with temperature indeed. Further support for the present analysis stems from the fact that one finds almost identical results for $c_1$, whether one starts from the total $c$- and $\alpha$-data or from the phonon-corrected data. The analysis of $c_2$, however, depends very sensitively upon the phonon contribution so that the interpretation in terms of crystal field contributions remains difficult. Further details will be published elsewhere.

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