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Anomalous magnetovolume effects of $CeCu_6$ at low temperatures

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We report on a low-temperature magnetovolume study of single-crystalline heavy-fermion $CeCu_6$. Magnetostriction data ($B \| c$) taken at temperatures below 100 mK reveal two distinct anomalies at $B_1 = 2.2$ T and $B_2 = 4.0$ T, that are rapidly washed out by increasing the temperature. Thermal expansion measurements in fields up to 8 T ($B \| c$) reveal an anomalous field suppression of the term linear in temperature. The fields B_1 and B_2 are likely related to a change in magnetic correlations. Their interrelation is investigated by a scaling approach.

1. Introduction

In the past years the intermetallic compound $CeCu_6$ has been investigated intensively [1,2] because of its unusual f-electron properties. The hybridization of the 4f (Ce) electrons with the Cu ligand 3d or 3p orbitals leads to the formation of heavy-electron bands, as evidenced by the low-temperature Sommerfeld coefficient in the specific heat ($\gamma \simeq 1600$ mJ/mol K^2 [2–4]). When compared to the non-f-electron system $LaCu_6$ ($\gamma = 8$ mJ/mol K^2 [3]), an effective quasiparticle mass of 480 times the free electron mass results, which characterizes $CeCu_6$ as one of the "heaviest" heavy-electron systems. The strong mass renormalization is furthermore confirmed by extensive de Haas–van Alphen studies performed at very low temperatures ($T < 250$ mK) [5]. Interestingly, $CeCu_6$ is one of the few heavy-fermion compounds that remain in a Pauli paramagnetic state, even at the lowest temperatures investigated (experiments extend down to 14 mK [6]).

In general, the strong mass renormalization in heavy-fermion compounds is attributed to the presence of competing electronic interactions: the onsite Kondo screening and the intersite Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction. For exemplary systems like UPt_3 and $CeRu_2Si_2$ a satisfactory qualitative description of the unusual low-temperature thermodynamic, transport and magnetic properties can be given based on such a model [7]. In particular, the occurrence of a metamagnetic-like transition at fairly large magnetic fields (at 20 T for

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UPt_3 [8] and at 8 T for $CeRu_2Si_2$ [9]) can be explained as a quenching of the intersite correlations. Note that we use the term metamagnetic-like because no long-range order with large moments is observed in these compounds. In the case of $CeCu_6$, inelastic neutron-scattering data [10] also reveal two competing electronic interactions below approximately 10 K: (i) a single-site fluctuation of the Kondo-type and (ii) an antiferromagnetic correlation between adjacent Ce^{3+} moments in the *bc*-plane ($CeCu_6$ has an orthorhombic crystal structure, but goes through a weak monoclinic distortion on cooling at about 220 K [11]; we here retain the orthorhombic notation). Inelastic neutron-scattering data in an applied magnetic field ($B \parallel c$, $T<1$ K) yield a progressive suppression of the intersite interactions with field, a threshold field $B^* \sim 2.5$ T (at 60 mK) and a total suppression of the correlations for fields exceeding 4 T [10]. However, the detection of the concurrent metamagnetic-like transition in the macroscopic magnetization is expected to be a difficult task, because of the small weight of the intersite contribution. Indeed, accurate low-temperature magnetization measurements (along the easy-axis for magnetization (*c*-axis)) down to 1.6 K [2] and down to 0.55 K [12] revealed a strictly linear $M=\chi H$ behaviour in the field range of interest. However, in a search for de Haas–van Alphen oscillations in fields up to 4 T ($B \parallel c$) anomalies in $\partial^2 M/\partial H^2$ were observed at dilution refrigerator temperatures [13] and, only very recently, a weak maximum in $\partial M/\partial H$ at a field of 1.7 T ($B \parallel c$) has been reported at $T=0.3$ K [14]. Magnetoresistance measurements revealed a maximum at 1.5 T at very low temperatures ($T<100$ mK) [2,12].

As in heavy-fermion compounds the coupling of the quasiparticles to the lattice is unusually strong, as evidenced by the electronic Grüneisen parameters that are two orders of magnitude larger than in ordinary metals [15,16], large magnetovolume effects are observed at the metamagnetic-like transition. Magnetostriction measurements and thermal expansion measurements in a field are a very sensitive probe to study metamagnetism ($B^*=8$ T) in $CeRu_2Si_2$ [17–19]. Also in the case of UPt_3 a large magnetostriction is observed at the metamagnetic-like transition at 20 T [16]. However, in these compounds the weight of the intersite contribution is considerable as the metamagnetic-like transition is evidenced by large anomalies in the magnetization and the magnetoresistance. At this time the question remains: how do the fore-mentioned low-field low-temperature anomalies for $CeCu_6$ turn up in the magnetovolume data, and how do these effects compare with the volume effects at the metamagnetic-like transition in other heavy-fermion compounds, e.g. $CeRu_2Si_2$? In order to elucidate this issue we here report on magnetostriction and thermal expansion measurements (in a field) of single-crystalline $CeCu_6$ in the temperature range $0.08<T<0.5$ K and field range $B<8$ T.

Our previous dilatation study of $CeCu_6$ in zero field ($0.02<T<100$ K) has revealed that the coefficients of linear thermal expansion are strongly anisotropic [20]. At low temperatures the volume expansion (α_V) receives its main contribution from the expansion along the *c*-axis. A pronounced maximum in $\alpha_V(T)$ appears at 2.5 K, which has been attributed to the Kondo-lattice effect. The low-temperature Grüneisen parameter for the heavy-fermion contribution, $\Gamma_{hf} = -\partial \ln T^*/\partial \ln V$, amounts to the large value of 57.

2. Experimental details

The single-crystalline $CeCu_6$ sample was pulled [21] from a tungsten crucible using the Czochralski technique. As starting materials served 99.99% pure Ce (Rare Earth Products Ltd.) and 99.999% pure Cu (Koch Light Ltd.). No additional annealing was performed. The sample was shaped by means of spark erosion in the form of a parallelepiped with dimensions $3.8\times5.0\times3.8$ mm^3.

The linear magnetostriction, $\lambda=[L(B)-L(0)]/L(0)$, and the coefficient of linear thermal expansion, $\alpha=L^{-1}\mathrm{d}L/\mathrm{d}T$, were measured using a sensitive three-terminal capacitance method with a detection limit for length changes of 0.01 Å. The sample was placed in a parallel-plate capacitance cell, machined of oxygen free high conductivity copper [22]. Measurements were performed in a dilution refrigerator equipped with a superconducting magnet. The cell was thermally anchored to the copper tail of the mixing chamber. A compensated zero-field region was available at the level of the mixing chamber, allowing for accurate thermometry. The magnetostriction measurements were performed by slowly sweeping the field, meanwhile recording the capacitance signal. The coefficient of thermal expansion was obtained by using a temperature modulation technique.

3. Experimental results

The parallel magnetostriction along the c-axis (i.e. $\Delta L \| c$ and $B \| c$) has been measured at temperatures of 80, 100, 150, 200, 300 and 350 mK. Some typical λ versus B curves are shown in fig. 1. The most striking results are obtained at the lowest temperature (80 mK): after a rapid rise at low fields a distinct kink appears at $B_1 = 2.2$ T and a sharp maximum at $B_2 = 4.0$ T. B_1 and B_2 are only weakly temperature dependent, however, by increasing the temperature the anomalies are rapidly washed out and a weak maximum remains for fields slightly exceeding B_2.

The thermal expansion data in a constant applied field are shown in fig. 2 (here also $\Delta L \| c$ and $B \| c$). In zero field the coefficient α_c is large and positive. At applying a magnetic field α_c is gradually suppressed but changes sign between 3 and 5 T. Retaining only the term linear in temperature (for $T \to 0$) $\alpha_c = a_c T$, the field suppression of α_c can be expressed by the field variation of a_c as shown in fig. 3.

Note that the experimental results presented in figs. 1–3 only relate to the strain along the field direction. In order to perform a more complete analysis the total volume effect should be considered and therefore also the strains perpendicular to the magnetic field ($\Delta L \| a$ and $\Delta L \| b$) should be measured, as

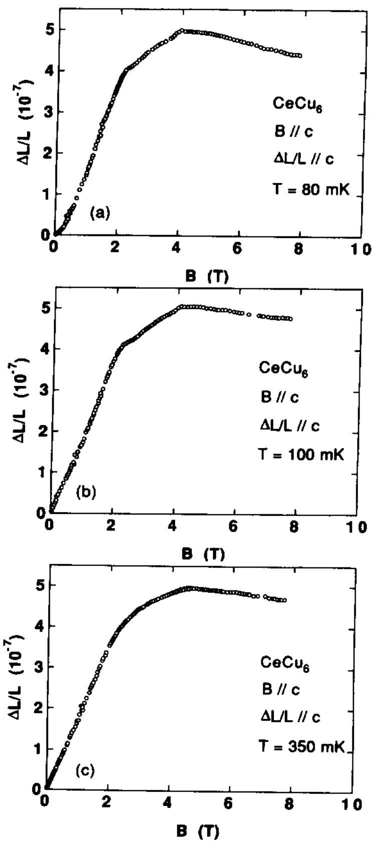

Fig. 1. Magnetostriction of $CeCu_6$ ($B \| c$, $\Delta L \| c$) at temperatures of (a) 80 mK, (b) 100 mK and (c) 350 mK.

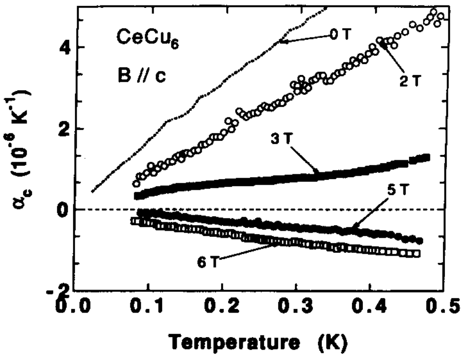

Fig. 2. The coefficient of thermal expansion of $CeCu_6$, $\alpha_c(T)$, for a field and dilatation direction along the c-axis, in magnetic fields as indicated.

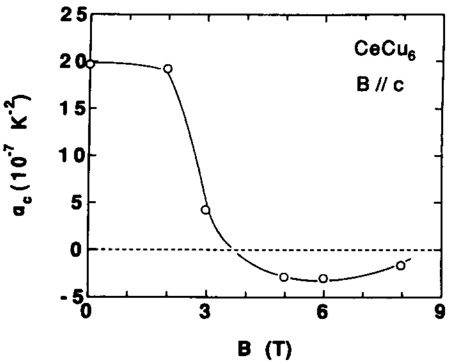

Fig. 3. Field variation of the coefficient of the linear term, $a_c = \alpha_c / T$, in the thermal expansion of $CeCu_6$ along the c-axis ($B \| c$).

$\alpha_V = \alpha_a + \alpha_b + \alpha_c$. However, such measurements have not been performed thus far and we concentrate in the following on a qualitative analysis.

4. Discussion

The data in figs. 1–3 convincingly show that $CeCu_6$ at very low temperatures exhibits at least three different field regimes: (i) up to 2.2 T, λ increases rapidly, while a_c is rather field insensitive, (ii) for $2.2 < B < 4$ T a much weaker field dependence of λ is observed, while a_c displays a precipitous drop, and (iii) for fields exceeding 4 T, λ starts to decrease and a_c becomes negative. The strain effects are such that: in the low-field region a strong positive dilatation and magnetostriction is observed, in the intermediate-field range the effects become much smaller, whereas above 4 T, by further increasing the magnetic field, the contrary takes place, namely a contraction of the sample with field and temperature. As the fields B_1 and B_2 are comparable to the characteristic fields that are observed in the ineleastic neutron-scattering data [10] they are likely related with changes in magnetic correlations. Interestingly, three different regimes are also observed in the field dependence of the elastic constants as probed by sound velocity measurements [23]. In particular the c_{33} mode reveals oscillatory behaviour, with sharp dips at 2.2 and 4 T. This seems to indicate that the changes in magnetic correlations are accompanied by modifications of the Fermi surface at 2.2 and 4 T. The anomalies observed below 4 T in $\partial^2 M/\partial H^2$ in the de Haas–van Alphen experiments [13] have been interpreted in a similar way.

In the compounds $CeRu_2Si_2$ and UPt_3 the metamagnetic-like transition turns up as a sharp inflection point at B^* in the magnetostriction curve [16,17]. At first sight such an effect is not observed for $CeCu_6$. However, a closer inspection of the data in fig. 1 does indeed reveal an inflection point, albeit rather weak, near $B^* = 1.5$ T. This value corresponds quite well with the B^*-values reported from the magnetoresistance (1.5 T [2,12]) and magnetization (1.7 T [14]) data. Note that a precise determination of B^* (~ 2.5 T) by the inelastic neutron-scattering data [10] is beyond the experimental limits. Thermal expansion measurements in a field on $CeRu_2Si_2$ [18,19] have shown that the coefficient of the linear temperature term, $\alpha = aT$, rises rapidly for $B \to B^*$, while it changes sign at B^*. As for $CeCu_6$ the contribution of the intersite interactions to the magnetic properties is very small we do not expect to observe a large anomaly in a_c at B^* (fig. 3), at least not in the measured temperature range ($T > 0.08$ K). The measured size of the magnetovolume effect for $CeCu_6$ up to 8 T is roughly three orders of magnitude smaller than observed for $CeRu_2Si_2$ at $B^* = 8$ T.

Summarizing, $CeCu_6$ seems to have three characteristic magnetic fields at low temperatures. A metamagnetic-like transition at $B^* \sim 1.5$ T, followed by two other transitions at $B_1 = 2.2$ T and $B_2 = 4$ T. However, as the weakness of the effects may lead to an artificial distinction of B^* and B_1, one cannot exclude that B^* and B_1 indicate but one single cross-over field. From the inelastic neutron-scattering data [10] it is concluded that the intersite correlations are completely suppressed for fields exceeding 4 T ($= B_2$). Therefore, the suppression of the intersite correlations seems to take place in two steps with characteristic fields B_1 (or B^*) and B_2. This reminds one of the field–temperature phase diagram of a weakly anisotropic long-range ordered antiferromagnet, where the suppression also takes place in a two step process (via a spin-flop phase) [24]. In which sense these effects are related with changes of the Fermi surface remains unclear at present.

In the case of $CeRu_2Si_2$ the low-temperature properties can be scaled extremely well, by the use of one single temperature (T^*) and field scale (B^*) [17–19]. The characteristic energy here is related to the strength of the intersite correlations that largely dominate the field dependence of the magnetization (M), the magnetostriction (λ), the specific heat ($\gamma(B)$) and the thermal expansion ($a(B)$). In particular, we demonstrated that M scales with λ [17], and that $\gamma(B)$ scales with $a(B)$ [18,19]. As in $CeCu_6$ both $\gamma(B)$ and $a(B)$ have a rather strong field dependence it is of interest to investigate their relation via the scaling expression [18]

$$a(B) = \frac{\kappa}{V_m}\left(\Gamma_T \gamma(B) + \Gamma_B B \frac{\partial \gamma}{\partial B}\right), \quad (1)$$

where κ is the compressibility and V_m is the molar volume. The thermal (Γ_T) and magnetic (Γ_B) Grüneisen parameters express the volume dependence of the characteristic temperature and field. We here assume $\Gamma_T = \Gamma_B$ [17–19]. In fig. 4 we compare the experimental values of $\gamma(B)$ [2] with the ones calculated from $a_c(B)$ (fig. 3) using eq. (1). It is readily seen that the scaling ansatz gives a quite satisfactory result, except above 6 T. Note that the comparison made in fig. 4 can only be relative ($\gamma(0)$ is normalized to 1), because the dilatation has only been measured *along* the field so far. We furthermore assumed that the coefficients a_a and a_b (dilatation directions perpendicular to the field direction) have an identical or much weaker field variation compared to a_c. It is tempting to identify the relevant characteristic energy scale with B^*, as in the case of $CeRu_2Si_2$. The success of the scaling over a wide field range up to 6 T, implies that B_1 (or B^*) and B_2 are closely connected, at least their Grüneisen parameters must be nearly equal.

For the compounds $CeRu_2Si_2$ and UPt_3 a considerable enhancement of the quasiparticle mass at B^* is observed for $T \to 0$: $m_{eff}(B^*)/m_{eff}(0)$ amounts to 1.77 at $B^* = 8$ T in the case of $CeRu_2Si_2$ [19] and to 1.44 at $B^* = 20$ T in the case of UPt_3 [25]. A clear-cut field mass enhancement has not been observed in the case of $CeCu_6$ thus far (fig. 4). Nevertheless, metamagnetism seems to be a common feature of these materials, although the energy scale is much smaller in the case of $CeCu_6$. Concurrently, in $CeCu_6$ the contribution from metamagnetism to the magnetic and thermal properties is extremely weak.

In summary, our magnetovolume study of $CeCu_6$ reveals two threshold fields, at $B_1 = 2.2$ T and at $B_2 = 4$ T, at very low temperatures. These characteristic fields likely indicate a two step process in the suppression of the intersite correlations.

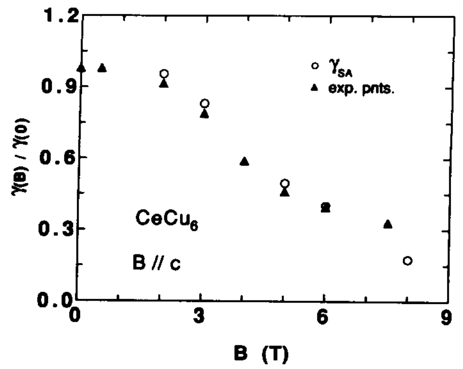

Fig. 4. The relative field variation of γ (normalized to 1 at $B = 0$ T), as measured (▲) [2] and as calculated (○) by the scaling ansatz using eq. (1).

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