Physica B 161 (1989) 324–332
North-Holland, Amsterdam

MAGNETO-VOLUME EFFECTS IN SOME SELECTED HEAVY-FERMION COMPOUNDS

A. de VISSER[a] and J.J.M. FRANSE
Natuurkundig Laboratorium der Universiteit van Amsterdam, Valckenierstraat 65, 1018 XE Amsterdam, The Netherlands

J. FLOUQUET
[a]Centre de Recherches sur les Très Basses Températures, CNRS, BP 166X, 38042 Grenoble-Cédex, France

We present a review of recent magneto-volume experiments, such as thermal expansion and magnetostriction, performed on good-quality single crystals of heavy-fermion systems, like $CeCu_6$, $CeRu_2Si_2$, URu_2Si_2, UBe_{13} and UPt_3. All investigated compounds exhibit large magneto-volume effects at low temperatures, as is reflected by their huge Grüneisen parameters (at least two orders of magnitude larger than in ordinary metals). A combination of specific-heat and thermal-expansion data yields furthermore information on the pressure dependence of the characteristic temperatures, and allows, in particular cases, for a detailed separation of the Kondo, crystal-field, phonon or other contributions to the thermal properties. We also discuss the magneto-volume effect connected with the metamagnetic-like transitions observed at 20 and 8 T (at 4.2 K) for UPt_3 and $CeRu_2Si_2$, respectively.

1. Introduction

Heavy-fermion systems are characterized by a large coefficient (γ) of the linear electronic specific heat that finds its origin in the low-temperature formation of a highly-correlated f-band close to the Fermi level [1]. Within the Fermi-liquid description the effective mass of the highly correlated electrons amounts to 200 times the free electron mass, and, correspondingly, a low Fermi temperature is observed. Heavy-fermion behaviour is mostly found in Ce and U-compounds as a result of the hybridization of the f-electrons with the p or d-orbitals of the ligand atoms.

Measurements of the coefficient of thermal expansion of heavy-fermion systems are of great interest as the volume effects related with the formation of the quasiparticle bands are large ($\Delta V/V \sim 10^{-4}$). These effects are usually described employing a phenomenological Grüneisen parameter, that represents the coupling between the heavy electrons and the lattice, but their origin lies, of course, in the single-ion Kondo or Kondo-lattice effect and the onset of strong (anti)ferromagnetic spinfluctuations, eventually in the presence of – and thus in a possible admixture with – crystalline-electric field effects. Since detailed theoretical expressions for the thermal-expansion coefficients of heavy-fermion systems are not available yet, we concentrate here on the phenomenological approach, using Grüneisen parameters.

For a particular contribution to the thermal properties a physically meaningful Grüneisen relation emerges, when the entropy S_i can be written as $S_i(T/T_i(V))$, where $T_i(V)$ is a volume-dependent characteristic temperature of the entropy term. The coefficient of volume expansion for this contribution is given by:

$$\alpha_V = \kappa \left(\frac{\partial S_i}{\partial V}\right)_T = -\kappa \left(\frac{\partial^2 F_i}{\partial V \partial T}\right), \tag{1}$$

where F_i is the free energy and $\kappa = -1/V\,(\partial V/\partial P)_T$ is the isothermal compressibility. The molar specific heat (at constant volume) is given by:

$$c_V = T\left(\frac{\partial S_i}{\partial T}\right)_V = -T\left(\frac{\partial^2 F_i}{\partial T^2}\right)_V. \tag{2}$$

The Grüneisen parameter for this particular entropy term is then defined by:

$$\Gamma_i \equiv \frac{V_m \alpha_V}{\kappa c_V} = -\frac{\partial \ln T_i(V)}{\partial \ln V} , \quad (3)$$

where V_m is the molar volume. In the case of heavy fermions one usually approximates the low-temperature specific heat and expansion coefficient by $c = \gamma T$ and $\alpha_V = 3aT$, respectively. The resulting heavy-fermion Grüneisen parameter is

$$\Gamma_{hf} = \frac{3aV_m}{\kappa\gamma} = -\frac{\partial \ln T_F}{\partial \ln V} . \quad (4)$$

The value of Γ_{hf} is about 100 times larger than the corresponding one for the linear electronic term in normal metals ($\Gamma_e \sim 2$), indicating a strong volume dependence of the quasiparticle bands. This implies that the expansion coefficient is more enhanced by the heavy-fermions, than the specific heat itself. Thermal-expansion measurements are thus an important tool to study heavy electrons. Besides, a Grüneisen-parameter analysis can be used to separate the various terms that contribute to the thermal properties. A next advantage is that thermal-expansion measurements may serve to study anisotropy effects (that play an important role in non-cubic heavy-fermion systems) in contrast to specific heat which is a bulk property. Furthermore, a combination of specific heat and thermal expansion can be used to determine the pressure dependence of the characteristic temperatures, and may serve as a check of their direct measurements as function of hydrostatic pressure.

The aim of this paper is to review recent thermal expansion measurements on some selected heavy-fermion compounds. We focus on $CeCu_6$, $CeRu_2Si_2$, URu_2Si_2, UBe_{13} and UPt_3. Magneto-volume effects in these compounds have also been studied as function of an external magnetic field (except for $CeCu_6$). Of particular interest are the systems UPt_3 and $CeRu_2Si_2$, where a metamagnetic-like transition occurs (at low temperature) at 20 and 8 T, respectively. This transition has been investigated by magnetostriction measurements, and the data have been analysed in a simple scaling model. All measurements have been performed on single-crystalline samples (dimension ~2–5 mm), using

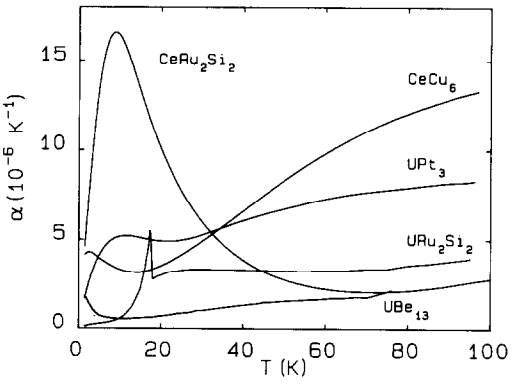

Fig. 1. Coefficient of linear thermal expansion ($\alpha_V/3$) for $CeCu_6$[2], $CeRu_2Si_2$[3], URu_2Si_2[4], UBe_{13}[5] and UPt_3[6].

a sensitive capacitance dilatometer with a detection limit $\Delta L/L \sim 10^{-8}$. An overall picture of the coefficient of volume expansion of the investigated systems is presented in fig. 1. As is clear from fig. 1, several processes (e.g. Kondo-effect, spin fluctuations, crystalline-electric field and phonons) contribute to α. Therefore, it is convenient to define an effective Grüneisen parameter:

$$\Gamma_{eff} = \frac{V_m \alpha_V(T)}{\kappa c(T)} = \sum_i \Gamma_i \frac{c_i(T)}{c(T)} \quad (5)$$

Γ_{eff} is, in general, temperature dependent and can be written as a sum of the products of the individual temperature independent Grüneisen parameters Γ_i and the fractional specific heats $c_i(T)/c(T)$. In fig. 2 we have plotted Γ_{eff} as

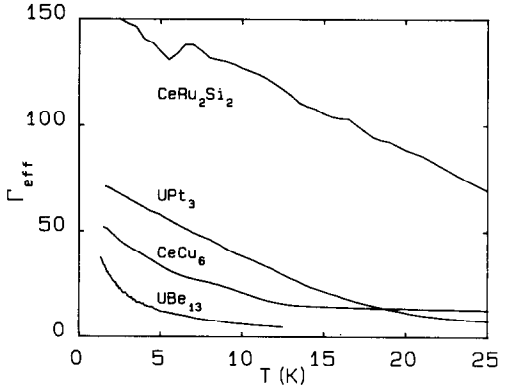

Fig. 2. Effective Grüneisen parameter for $CeCu_6$[2], $CeRu_2Si_2$[3], UBe_{13}[5] and UPt_3[7]. In the case of $CeCu_6$ and $CeRu_2Si_2$ the phonon contribution has been subtracted.

Table I
The coefficient of the linear term in the specific heat (γ) and thermal expansion (a), compressibility (κ), molar volume (V_m), low-temperature Grüneisen parameter (Γ_{ht}) and characteristic temperatures for several heavy-fermion systems.

Compound	Structure	γ (mJ/mol K^2)	a (10^{-6} K^{-2})	κ (10^{-11} m^2/N)	V_m (10^{-6} m^3/mol)	$\Gamma_{ht} = \frac{3aV_m}{\kappa\gamma}$	Characteristic temperatures	References
$CeCu_6$	Orthorhombic	1670	600	1.21	6.33	57	T_K = 3.9 K, Δ_1 = 64 K, Δ_2 = 128 K	2,8,9
$CeRu_2Si_2$	Tetragonal	330	320	1	5.19	150	T_K = 24 K, Δ_1 = 220 K, Δ_2 = 1000 K	3,10,11
URu_2Si_2	Tetragonal	64	8	0.73**	4.9	25	T_S = 1 K, T_N = 17.5 K, Δ_1 = 40 K	4,12
UBe_{13}	Cubic	780*	119*	1.08***	8.13	34*	T_S = 0.9 K, T_K = 8 K	5,13,14
UPt_3	Hexagonal	430	116	0.48	4.24	71	T_S = 0.5 K, T_A = 12 K	6,7

*at 1.5 K **estimated value ***at 10 K

function of temperature for some systems. At low temperatures Γ_{eff} varies rapidly with T. Only in the low-temperature limit the linear terms in c and α are attained and the equality $\Gamma_{eff} = \Gamma_{ht}$ holds. This implies that a scaling law with one single energy parameter can only be valid at low temperatures. In table I and table II we have collected some relevant parameters for the investigated systems.

2. $CeCu_6$

The compound $CeCu_6$ crystallizes in the orthorhombic structure. It has the largest γ-value among the studied compounds: $\gamma = 1670$ mJ/mol K^2 [8] (table I). No superconductivity or magnetic order is observed down to 18 mK. The coefficients of linear thermal expansion along the three crystallographic directions, α_a, α_b and α_c, have been measured in the temperature range 1.4–100 K [2] (see fig. 3) and between 6 and 200 K [16]. As follows from fig. 3, α is strongly anisotropic. With raising temperature the unit cell expands along the b and c-axis, while it shrinks along the a-axis up to 17 K. Above 19 K the c-axis begins to shrink till it expands again above 31 K. This complex behaviour is reflected in the various extrema observed in the volume expansion $\alpha_V = \alpha_a + \alpha_b + \alpha_c$ (fig. 1). When the phonon contribution, obtained on single-crystalline $LaCu_6$ [16], is subtracted, two f-electron

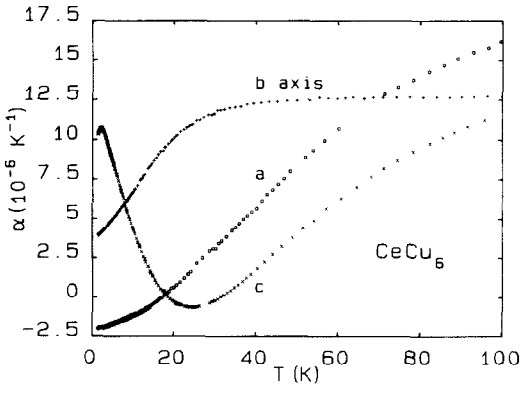

Fig. 3. Coefficient of linear thermal expansion of single-crystalline $CeCu_6$ along the $a(\circ)$, $b(+)$ and $c(\times)$ axis (after ref. [2]).

Table II
Comparison of Grüneisen parameters for heavy-fermion systems as obtained from the pressure dependence of γ [15], $\Gamma^{\gamma}_{meas} = \partial \ln \gamma / \partial \ln V$, or χ [23, 28, 32], $\Gamma^{\chi}_{meas} = \partial \ln \chi(4.2\,K)/\partial \ln V$, and as calculated from a combination of c and α, according to $\Gamma^{\gamma}_{calc} = 3aV_m/\kappa\gamma$, or from a combination of λ and χ (using eq. (6)).

Compound	Γ_{meas}	Γ^{γ}_{calc}	Γ^{χ}_{meas}	Γ^{χ}_{calc}
$CeCu_6$	72	57	—	—
$CeRu_2Si_2$	—	150	171 (B‖c)	188 (B‖c)
URu_2Si_2	—	25	40 (B‖c)	21 (B‖c)
UBe_{13}	26*	34*	—	7.4 (B‖a)
UPt_3	55	71	58 (B⊥c)	46 (B⊥c)
			10 (B‖c)	6 (B‖c)

*at T = 1.5 K

contributions remain: (i) a low-temperature anomaly centered at 2.5 K, attributed to the Kondo-lattice effect ($T_K = 3.9$ K), and (ii) a broad high-temperature contribution centered at ~60 K, associated with crystal-field effects. Specific-heat [8] and neutron-scattering experiments [17] have shown that the $J = \frac{5}{2}$ multiplet is split into three doublets with the approximate level scheme 0–64–128 K. The Grüneisen parameter for the lowest crystal field splitting (Δ) amounts to 13, implying an increase of Δ, with pressure at a rate of 1 K/kbar. The strong admixture of the heavy-fermion and crystal-field terms urges for a model that incorporates both effects. On alloying $CeCu_6$ with small amounts of Al, the low-temperature anomaly broadens and shifts towards higher temperatures [2], leading to an increase of T_K, as can be understood from the positive chemical pressure.

3. $CeRu_2Si_2$

The compound $CeRu_2Si_2$ crystallizes in the tetragonal structure. Again no superconductivity or magnetic order is observed down to the mK region. The thermal expansions of single-crystalline $CeRu_2Si_2$ and its non-magnetic analog $LaRu_2Si_2$ have been measured in the temperature range 1.5–250 K [3, 18]. Both $\alpha_{\parallel}$ (along the tetragonal axis) and $\alpha_{\perp}$ (in the plane) are positive, thus the unit cell expands with raising temperature, faster along the tetragonal axis than in the plane. After subtracting the matrix $LaRu_2Si_2$, two pronounced anomalies are observed: (i) a huge low-temperature peak centered at 9 K (see fig. 1), that is attributed to the onset of antiferromagnetic correlations between the f-electrons, and (ii) a negative crystal-field contribution with a minimum at 120 K. According to the specific heat analysis the $J = \frac{5}{2}$ multiplet is split into 3 doublets with a level scheme: 0–220–1000 K [10]. An analysis of the thermal expansion in the crystal-field model leads to a slightly higher first excited level: $\Delta_1 \simeq 288$ K [18].

The effective Grüneisen parameter reaches the huge value of 150 at 1 K, but drops rapidly with temperature and levels off to a value of −4 above 70 K. Employing the extreme values $\Gamma_{hf} = 150$ and $\Gamma_{cf} = -4$ one can nicely separate the heavy-fermion and crystal-field contributions [3]. An analysis of the two contributions will be presented elsewhere [18]. $\Gamma_{cf} = -4$ implies that the first excited crystal-field level decreases with pressure at a rate of −1.1 K/kbar.

Neutron experiments [19] have proven the existence of strong antiferromagnetic correlations below 70 K, revealing that $CeRu_2Si_2$ is close to an antiferromagnetic instability. Indeed, when alloying $CeRu_2Si_2$ with La, long-range antiferromagnetic order occurs between 8 and 90% La, with a maximum Néel temperature of 6.3 K [20]. The evolution of the large low-temperature anomaly for non-ordered $CeRu_2Si_2$ into a negative peak for the doped ordered compounds is at present under investigation.

At $H^* = 8$ T $CeRu_2Si_2$ shows a metamagnetic-like transition for a field along the tetragonal axis

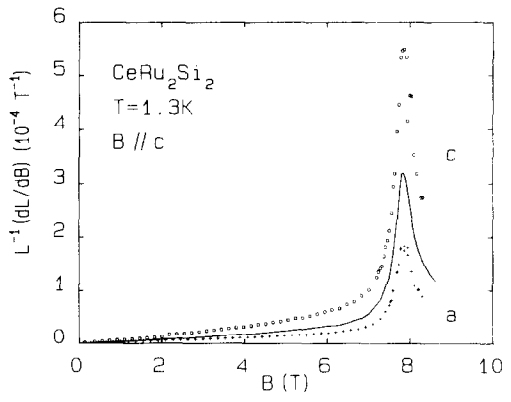

Fig. 4. Magnetostriction of $CeRu_2Si_2$ along the c(∘) and a(+) axis, for a field along the c-axis at $T = 1.3$ K (after ref. [18]). The solid line represents $\lambda_V'/3$.

(at liquid He temperatures) [21], as a result of the quenching of the antiferromagnetic correlations. Magnetostriction measurements up to 15 T were performed by Puech et al. [22], revealing large volume effects at the transition (see also fig. 4). The magnetostriction data could be related to the pressure dependence of the magnetization, using a simple scaling law: $M(H, P) = f(H/H^*(P))$. This scaling law has been investigated extensively by Mignot et al. [23]. A close relation between H^* and the characteristic Fermi-liquid energy $k_B T^*$ was concluded.

4. URu_2Si_2

Tetragonal URu_2Si_2 is a particularly interesting compound, because it undergoes an antiferromagnetic transition at $T_N = 17.5$ K, followed by a superconducting one at $T_S = 1$ K [12]. The specific-heat anomaly at T_N is large, but the ordered moment small (0.02 μ_B/U-atom) [24]. It has been suggested that the antiferromagnetic transition is accompanied by a strong rearrangement of the Fermi surface. The thermal expansion of single-crystalline URu_2Si_2 has been studied in the temperature range 1.4–100 K [4], where α is strongly anisotropic: $\alpha_\parallel$ is negative below 30 K, $\alpha_\perp$ is large and positive. The volume expansion is shown in fig. 1. Three anomalies are observed: (i) a broad low-temperature contribution near 5 K, leading to the heavy-fermion Grüneisen parameter of 25 (table I), (ii) a sharp peak at T_N, and (iii) a crystal-field contribution centered at 25 K. The latter anomaly is tentatively ascribed to a singlet-singlet splitting of 40 K. Employing the Ehrenfest relation $\partial \ln T_N/\partial P = \Delta\alpha_V V_m/\Delta c$ one can relate the pressure dependence of T_N to the jumps in α and c. With the values $\Delta\alpha_V = 9 \times 10^{-6}$ K^{-1} and $\Delta c = 5.5$ J/Kmol we calculate $\partial \ln T_N/\partial P = 8$ Mbar^{-1}, in good agreement with the value of 6.8 Mbar^{-1} that followed from a high-pressure resistivity study [25].

The magnetostriction of single-crystalline URu_2Si_2 was studied in fields up to 8 T below (at 4.2 K) and above (at 20 K) T_N [26]. For all field and dilatation directions $\lambda = \Delta L/L \propto B^2$ as expected from the linear magnetization curves (non-linear behaviour occurs above 15 T; three transitions are observed between 35 and 40 T [27]). For a field along the tetragonal axis the magnetostriction is anisotropic: positive in the plane, negative along the tetragonal axis. The volume effect is positive, $\Delta V/V = aB^2$ with $a = 8.75 \times 10^{-8}$ T^{-2} at 4.2 K. For a field direction in the plane the magnetostriction is at least two orders of magnitude smaller. Employing Maxwell relations, $\partial \ln \chi/\partial \ln V$ has been evaluated from the magnetostriction data. The comparison with high-pressure susceptibility measurements [28] is made in table II. Interestingly, the magnetostriction is only weakly temperature dependent. This means that the magnetostriction does not probe the antiferromagnetism.

5. UBe_{13}

The superconductor UBe_{13} ($T_S = 0.9$ K) has a cubic structure. Its thermal expansion coefficient measured on a single crystal along the [1 0 0] direction is shown in fig. 1 for $1.4\text{ K} < T < 80\text{ K}$ [5]. At 10 K, α passes through a minimum and then rises rapidly with decreasing temperature as the onset of the heavy-fermion behaviour takes place. Interestingly, no low-temperature maximum is achieved before the superconducting transition sets in [29], implying a very low T_K. Combining the data of fig. 1 with specific-heat

data on a polycrystalline sample [13] one finds that $\Gamma_{\rm eff}$ rises rapidly at low temperatures. At 1.5 K $\Gamma_{\rm eff}$ attains a value of 34, but when extrapolated to 0 K, $\Gamma_{\rm eff}$ is nearly twice as large. In accordance with specific heat measurements [30] a broad hump is observed near 40 K, which is partly ascribed to a phonon anomaly. Thermal expansion measurements on $ThBe_{13}$ which could elucidate this anomaly, are not available yet.

The magnetostriction of UBe_{13} has been studied at 1.3 and 4.2 K for fields along the [1 0 0] direction up to 8 T [5]. As expected from the linear magnetization curves $\lambda \propto B^2$. The magnetostriction is anisotropic: negative along the field ($\lambda_\parallel$) and positive at right angles to the field ($\lambda_\perp$). The volume magnetostriction: $\lambda_V = \lambda_\parallel + 2\lambda_\perp = aB^2$ is positive, with $a = 6 \times 10^{-8}\,\mathrm{T}^{-2}$ at 4.2 K. Using Maxwell relations we calculate a relatively weak decrease of the susceptibility with pressure: $\partial \ln \chi(4.2\,\mathrm{K})/\partial P = -7.4\,\mathrm{Mbar}^{-1}$. We note that, while for most heavy-fermion compounds the magnetic and thermal Grüneisen parameters are almost equal, this appears not to be the case in UBe_{13} (table II).

6. UPt_3

The hexagonal compound UPt_3 has a γ-value of 430 mJ/mol K^2. Its low-temperature properties are governed by pronounced antiferromagnetic spinfluctuations, that might provide the driving mechanism for superconductivity that occurs at 0.5 K. The thermal expansion of UPt_3 has been measured between 1.4 and 300 K [6, 31]. The coefficient of expansion is positive in the basal plane ($\alpha_\perp$), where it attains a maximum at 12.5 K, and negative until ~40 K, with a minimum at 15 K, for the hexagonal axis. The volume expansion displays an anomaly at 10 K (fig. 1). The thermal properties of UPt_3 have been analyzed in a spin-fluctuation model [32], using a $T^3 \ln(T/T^*)$-term, but this analysis is hampered since an analog non-f electron system, that may serve to subtract the phonon contribution, does not exist. However, the phonon contribution to the specific heat could be determined from measurements of the phonon dispersion curves, obtained by inelastic neutron scattering experiments [33]. Recently, a Grüneisen parameter analysis [7] has been used to separate the various contributions to the thermal properties. At low temperatures two contributions were found to be present. One contribution is related with the heavy-fermion Grüneisen parameter of 71. It peaks at ~10 K and shows some resemblance to a Kondo $S = \frac{1}{2}$ anomaly with $T_K = 15.5$ K. The second contribution, of unknown origin, peaks at ~23 K. The suggestion [30] that this anomaly is of the Schottky-type are not confirmed by the Grüneisen-parameter analysis. It is emphasized that the low-temperature contribution in the specific heat and thermal expansion can still be fitted with a $T^3 \ln(T/T^*)$-term. However, compared to an earlier analysis [32], the fit interval ($T < 5$ K) and correspondingly the spin-fluctuation temperature (12 K) are reduced.

On alloying UPt_3 with Pd long-range antiferromagnetic order occurs for Pd contents between 2 and 10%, with a maximum T_N of 5.8 K [34]. Thermal-expansion measurements [31, 32] on the ordered compounds have shown that the large anomaly in α_V, present at 10 K in pure UPt_3, disappears. The transition temperature is observed as a sharp drop of the expansion coefficient.

Magnetostriction measurements on UPt_3 have been performed on a single-crystalline sample for fields up to 8 T (at 4.3 K) [35] and up to 24 T (at 1.8, 3.0 and 4.2 K) [36]. The high-field measurements aimed to study the metamagnetic-like transition that occurs for field directions in the basal plane and at temperatures below the maximum in the susceptibility [37]. The experimental results for a field along the a and b-axis ($b \perp a$) are shown in fig. 5. A large anisotropy between the basal plane and the c-axis appears, but also the magnetostriction in the basal plane is anisotropic, such that the magnetostriction at right angles to the field is larger than the one along the field (for $B \| a$ $\lambda_b > \lambda_a$ and for $B \| b$ $\lambda_a > \lambda_b$). Apparently, an orthorhombic distortion occurs for field directions in the basal plane. The distortion is largest for $B \| b$. The observed anisotropy can be understood qualitatively in a simple spin fluctuation picture, in which the low-field anti-

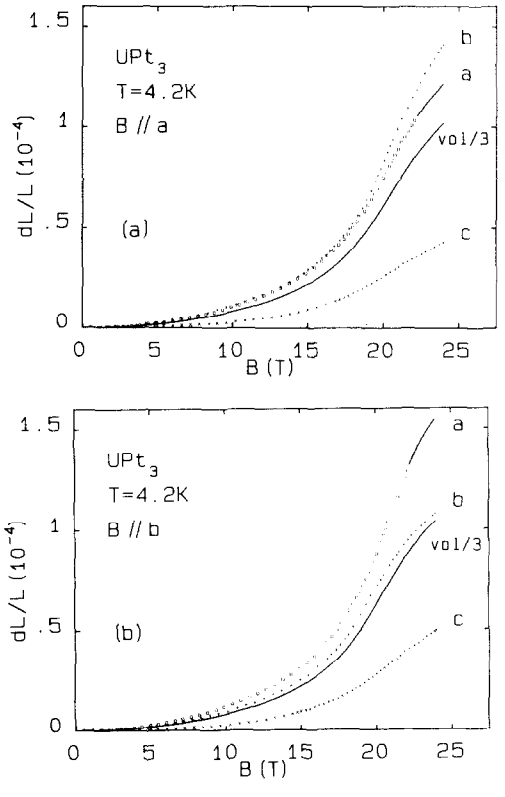

Fig. 5. Magnetostriction of UPt_3 at $T = 4.2$ K, along the a(∘), b(+) and c(×) axis; (a) for a field along the a-axis (after ref. [36]) and (b) for a field along the b-axis. The solid line represents $\lambda_V/3$ (solid line above 22 T for the a-axis is an extrapolation).

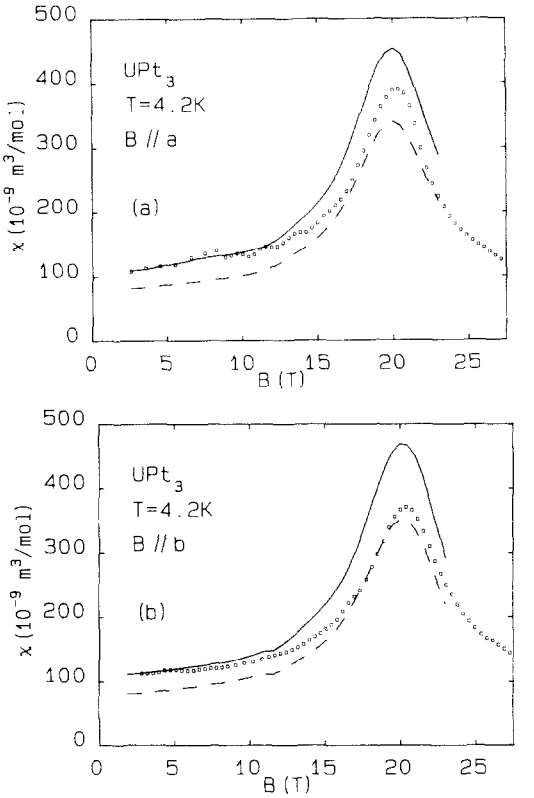

Fig. 6. Differential susceptibility, $\chi = \Delta M/\Delta H$, of UPt_3 at 4.2 K; (a) for a field along the a-axis and (b) for a field along the b-axis. Circles: as measured [32, 37]. Solid and dashed line: as calculated from the magnetostriction using eq. (6), with a value for $\partial \ln \chi/\partial P$ of -21 Mbar^{-1} and -28 Mbar^{-1}, respectively.

ferromagnetic fluctuations are quenched above 20 T. For a field along the c-axis $\lambda \propto B^2$, with $\lambda_{\parallel} < 0$ and $\lambda_{\perp} > 0$. The resulting volume magnetostriction is small.

The similarities between UPt_3 and $CeRu_2Si_2$ have raised the question whether the scaling law $M(H, P) = f(H/H^*(P))$ that applies to $CeRu_2Si_2$ [22], also holds in the case of UPt_3. We checked this scaling law by comparing the volume magnetostriction with the differential susceptibility, according to:

$$\left(\frac{\partial V}{\partial H}\right)_{P,T} = H \frac{\partial \ln H^*}{\partial P} \left(\frac{\partial M}{\partial H}\right)_{P,T}. \qquad (6)$$

The results are shown in fig. 6, where we used $\partial \ln H^*/\partial P = -\partial \ln \chi_0/\partial P$, since $H^*(P) = H^*(0)\chi_0(0)/\chi_0(P)$ (χ_0 refers to the Pauli susceptibility). For both field directions in the plane one observes that using the measured value of -28 Mbar^{-1} [32] for $\partial \ln \chi_0/\partial P$ χ_{calc} is too small in the low field region, but agrees quite well at high fields, whereas adjusting χ_{calc} to χ_{m} at low-fields, using $\partial \ln \chi/\partial P = -21$ Mbar^{-1}, leads to $\chi_{\mathrm{calc}} > \chi_{\mathrm{m}}$ at high fields. Apparently we cannot scale the data with a single energy parameter at this temperature. It is, however, of interest to reinvestigate this scaling law at lower temperatures.

7. Concluding remarks

The observation of large positive values for α_V in the heavy-fermion systems, reveals that the

electron–electron correlation and its coupling with the lattice, leads to a smaller ground-state volume (leaving eventual superconducting or magnetic transitions besides consideration) than without narrow f-bands. Quantitatively this can be understood in the accepted picture that the f-electrons are localized at high-temperatures but become itinerant at low temperatures and thus contribute to the binding. A similar picture exists for the large volume changes in intermediate valence compounds, where the increase (decrease) in valence is accompanied by a large decrease (increase) in volume. This was nicely illustrated by thermal expansion measurements on, for instance, YbCuAl [38] where a valence change $2+ \rightarrow 3+$ (with raising temperature) occurs with a large negative α_V, and the system $Ce_{1-x}Sc_xAl_2$ [39] where a valence change $4+ \rightarrow 3+$ leads to a large positive α_V. The anomalies could be quantitatively accounted for by a model of Müller-Hartmann [40]. In a first attempt to model the α of heavy-fermion compounds the resonance level model of Schotte and Schotte [41] can be used. In this model a Kondo resonance with width $\Delta \sim k_B T_K$ is formed near or at the Fermi level E_F. The sign of the thermal expansion anomaly depends in this case on the volume dependence of Δ and the exact position of E_F. The volume dependence of Δ is given by the heavy-fermion Grüneisen parameter $\Gamma_{hf} = -\partial \ln \Delta / \partial \ln V$. With this model we are able to fit the low-temperature data of $CeRu_2Si_2$ surprisingly well [18].

An intriguing question is the occurrence of a minimum at $T = 0.5$ K in the thermal expansion of the well-known heavy-fermion compound $CeAl_3$ [42] 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 α of these coherent (normal-state) heavy-fermion systems at low temperatures. Recently, thermal expansion measurements on $Ce_xLa_{1-x}Al_2$ [32] have been used to demonstrate that diluted (single-ion Kondo) compounds can have a negative α as well. Therefore, a negative α 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 state with pressure (see also ref. [44]).

A negative thermal expansion might also be a consequence of the occurrence of long-range magnetic order as is illustrated by the α of doped UPt_3 [31, 32] and $CeRu_2Si_2$ [18]. An alternative explanation of the negative α of $CeAl_3$ might therefore be found in the recently discovered long-range antiferromagnetic order with small magnetic moments [45] at $T = 0.7$ K. The important role of antiferromagnetic interactions in the heavy-fermion systems might also explain the anisotropy in the expansion coefficients of $CeCu_6$, $CeRu_2Si_2$ and UPt_3, for which we notice a correlation between the direction of confinement of spin fluctuations and the anisotropy in α. Related to this, one observes that α is largest for the easy direction of magnetization. For instance in $CeCu_6$ inelastic neutron-scattering experiments [46] have shown that the antiferromagnetic fluctuations are confined to the c-axis, which is also the easy axis for magnetization. The coupling between adjacent bc-planes was found to be antiferromagnetic. Consequently, the coefficient of expansion is largest along the confinement direction, α_c, and smallest (negative) between the coupled planes (α_a). Any model able to account for the anisotropy in the expansion coefficient of heavy-fermion compounds, should therefore include intersite correlations.

Acknowledgement

The work of A. de V. has been made possible by a fellowship of the Royal Netherlands Academy of Arts and Sciences.

References

[1] G.R. Stewart, Rev. Mod. Phys. 56 (1984) 755.
[2] A. de Visser, A. Lacerda, P. Haen, J. Flouquet, F.E. Kayzel and J.J.M. Franse, Phys. Rev. B 39 (1989) 11301.
[3] A. de Visser, P. Haen, P. Lejay and J. Flouquet, Proc. Int. Conf. on Magnetism, D. Givord, ed. (Les Éditions de Physique, Paris, 1989) p. 767.

[4] A. de Visser, F.E. Kayzel, A.A. Menovsky, J.J.M. Franse, J. van den Berg and G.J. Nieuwenhuys, Phys. Rev. B 34 (1986) 8168.
[5] A. de Visser et al., to be published.
[6] A. de Visser, J.J.M. Franse and A. Menovsky, J. Phys. F 15 (1985) L53.
[7] J.J.M. Franse, M. van Sprang, E. Louis, K. Kadowaki and A. de Visser, J. Magn. Magn. Mat. 76 & 77 (1988) 47.
[8] T. Fujita, K. Satoh, Y. Onuki and T. Komatsubara, J. Magn. Magn. Mat. 47 & 48 (1985) 66.
[9] A. Shibata, G. Oomi, Y. Onuki and T. Komatsubara, J. Phys. Soc. Jpn. 55 (1986) 2086.
[10] M.J. Besnus, J.P. Kappler, P. Lehmann and A. Meyer, Solid State Commun. 55 (1985) 779.
[11] J.-M. Laurant and M. Rossignol, private communication.
[12] T.T.M. Palstra, A.A. Menovsky, J. van den Berg, A.J. Dirkmaat, P.H. Kes, G.J. Nieuwenhuys and J.A. Mydosh, Phys. Rev. Lett. 55 (1985) 2727.
[13] H.R. Ott, H. Rudigier, Z. Fisk and J.L. Smith, in: Proc. NATO/CAP Inst. on Moment Formation in Solids, Vancouver Island, 1983, W.J.L. Buyers, ed. (Plenum, New York, 1984) p. 305.
[14] R.A. Robinson, J.D. Axe, A.I. Goldman, Z. Fisk, J.L. Smith and H.R. Ott, Phys. Rev. B 33 (1986) 6488.
[15] N.E. Philips, R.A. Fisher, J. Flouquet, A.L. Giorgi, J.A. Olsen and G.R. Stewart, J. Magn. Magn. Mat. 63 & 64 (1987) 332.
[16] G. Oomi, K. Takahashi, F.J. Ohkawa, Y. Onuki and T. Komatsubara, Physica B 148 (1987) 61.
[17] U. Walter, D. Wohlleben and Z. Fisk, Z. Phys. B 62 (1986) 325.
[18] A. Lacerda, A. de Visser, P. Lejay, P. Haen and J. Flouquet, Phys. Rev. B, in print.
[19] L.P. Regnault, W.A.C. Erkelens, J. Rossat-Mignod, P. Lejay and J. Flouquet, Phys. Rev. B 38 (1988) 4481.
[20] P. Haen, F. Lapierre, J.P. Kappler, P. Lejay, J. Flouquet and A. Meyer, J. Magn. Magn. Mat. 76 & 77 (1988) 143.
[21] P. Haen, J. Flouquet, F. Lapierre, P. Lejay and G. Remenyi, J. Low Temp. Phys 67 (1987) 391.
[22] L. Puech, J.-M. Mignot, P. Lejay, P. Haen, J. Flouquet and J. Voiron, J. Low Temp. Phys. 70 (1988) 237.
[23] J.-M. Mignot, J. Flouquet, P. Haen, F. Lapierre, L. Peuch and J. Voiron, J. Magn. Magn. Mat. 76 & 77 (1988) 97.
[24] C. Broholm, J.K. Kjems, W.J.L. Buyers, P. Matthews, T.T.M. Palstra, A.A. Menovsky and J.A. Mydosh, Phys. Rev. Lett. 58 (1987) 1467.
[25] F.R. de Boer, J.J.M. Franse, E. Louis, A.A. Menovsky, J.A. Mydosh, T.T.M. Palstra, U. Rauchschwalbe, W. Schlabitz, F. Steglich and A. de Visser, Physica B 138 (1986) 1.
[26] F.E. Kayzel, A. de Visser, A.A. Menovsky and J.J.M. Franse, Physica B 147 (1987) 231.
[27] A. de Visser, F. R. de Boer, A.A. Menovsky and J.J.M. Franse, Solid State Commun. (1987) 527.
[28] E. Louis, A. de Visser, A. Menovsky and J.J.M. Franse, Physica B 144 (1986) 48.
[29] H.R. Ott, Physica B 126 (1984) 100.
[30] R. Felten, Thesis, Technische Hochschule Darmstadt (1987), unpublished.
[31] M. van Sprang, Ph.D. Thesis, Universiteit van Amsterdam (1989), unpublished.
[32] A. de Visser, A. Menovsky and J.J.M. Franse, Physica B 147 (1987) 81.
[33] B. Renker, F. Gompf, J.B. Suck, H. Rietschel and P.H. Frings, Physica B 136 (1986) 376.
[34] A. de Visser, J.C.P. Klaasse, M. van Sprang, J.J.M. Franse, A. Menovsky, T.T.M. Palstra and A.J. Dirkmaat, Phys. Lett. A 113 (1986) 489.
[35] A. de Visser, E. Louis, J.J.M. Franse and A. Menovsky, J. Magn. Magn. Mat. 54–57 (1986) 387.
[36] A. de Visser, L. Puech, W. Joss, A.A. Menovsky and J.J.M. Franse, Jpn. J. Appl. Phys. 26 Suppl. 26–3 (1987) 513.
[37] P.H. Frings, J.J.M. Franse, F.R. de Boer and A. Menovsky, J. Magn. Magn. Mat. 31–34 (1983) 240.
[38] R. Pott, R. Schefzyk, D. Wohlleben and A. Junod, Z. Phys. B 44 (1981) 17.
[39] E. Ruderer, R. Schefzyk, H. Biesenkamp, W. Klämke, S. Horn and F. Steglich, J. Magn. Magn. Mat. 45 (1984) 219.
[40] E. Müller-Hartmann, Solid State Commun. 31 (1979) 113.
[41] K.D. Schotte and U. Schotte, Phys. Lett. A 55 (1975) 38.
[42] M. Ribault, A. Benoit, J. Flouquet and J. Palleau, J. de Phys. 40 (1979) L413.
[43] M. Lang, R. Schefzyk, F. Steglich and N. Grewe, J. Magn. Magn. Mat. 63 & 64 (1987) 79.
[44] E.P. Fetisov and D.I. Khomskii, Solid State Commun. 56 (1985) 403.
[45] S. Barth, H.R. Ott, F.N. Gygax, B. Hitti, E. Lippelt, A. Schenck and C. Baines, Jpn. J. Appl. Phys. 26 Suppl. 26-3 (1987) 519.
[46] L.P. Regnault, W.A.C. Erkelens, J. Rossat-Mignod, J. Flouquet, E. Walker, D. Jaccard, A. Amato and B. Hennion, J. Magn. Magn. Mat. 63 & 64 (1987) 289.