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Physica B 206 & 207 (1995) 225–227

PHYSICA B

Thermal expansion and magnetostriction of heavy-fermion $CePd_2Al_3$

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Abstract

We present low-temperature thermal expansion, magnetostriction and magnetoresistivity measurements on a single crystal of $CePd_2Al_3$. The strong anisotropy of $\alpha(T)$ confirms our earlier proposed anisotropic hybridization model for $CePd_2Al_3$. The effective Grüneisen parameter reaches 20 at 2 K. Upon application of magnetic fields along the *a*-axis, the thermal expansion rapidly becomes negative due to the quenching of the Kondo effect.

The intermetallic compound $CePd_2Al_3$ is a antiferromagnetic (AF) heavy-fermion material [1,2], of which annealed polycrystals order AF at $T_N = 2.7$ K, while single crystals remain paramagnetic down to 0.4 K. The hexagonal basal plane is the easy direction, and AF correlations build up due to coupling of the magnetic Ce–Pd layers via the isolated Al layer in between those layers. Recent ^{27}Al-NMR and NQR studies revealed that, although the Al-atom occupies one site in the ideal $PrNi_2Al_3$-type crystal structure, multiple NQR frequencies exist in quenched polycrystals and single crystals [3]. The occurrence of multiple frequencies and the intensity of a satellite peak of the $|\pm 3/2\rangle \leftrightarrow |\pm 5/2\rangle$ transition correlate with the type of ordering, going from long-range AF order in annealed polycrystals via short-range correlations in quenched polycrystals to absence of order in single crystals.

The aim of this paper is to test our anisotropic-hybridization model for $CePd_2Al_3$ [3] by thermal expansion and magnetostriction. This model incorporates two exchange parameters: in the basal plane, the Ce–Pd interaction J_{f-d} describes the f–d hybridization, giving rise to a Kondo effect, while along the *c*-axis, the Ce–Al interaction J_z is of RKKY-type. This leads to magnetic planes of reduced cerium moments, weakly coupled along the *c*-axis.

The thermal expansion, $\alpha(T)$, is measured on a cube ($3.8 \times 3.9 \times 5$ mm^3) of a single crystal, using a capacitance method, parallel to the *a*-axis down to 0.3 K and parallel to *c* to 1.5 K (see Fig. 1). At high temperatures, T, α is dominated by phonon contributions, which above 100 K may be described by Debye functions with $\Theta_D = 230$ K along *a*, and 460 K along *c*. These largely different values of Θ_D cast some doubts on the validity of application of the Debye model. Therefore the low-T electronic part of α, obtained after subtracting these Debye functions and shown in Fig. 2, is only a first approximation. The volume expansion, $\alpha_V = 2\alpha_a + \alpha_c$, exhibits two maxima. The anomaly around 30 K is associated with the depopulation of the first excited crystal-field level at 32 K [2,4]. The anisotropy of $\alpha(T)$, with $\alpha_a > \alpha_c$, indicates the larger in-plane Ce–Pd 4f-4d hybridization compared to 4f–3p hybridization between Ce- and Al-electron bands. The large positive α_a measures the delocaliza-

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SSDI 0921-4526(94)00415-3

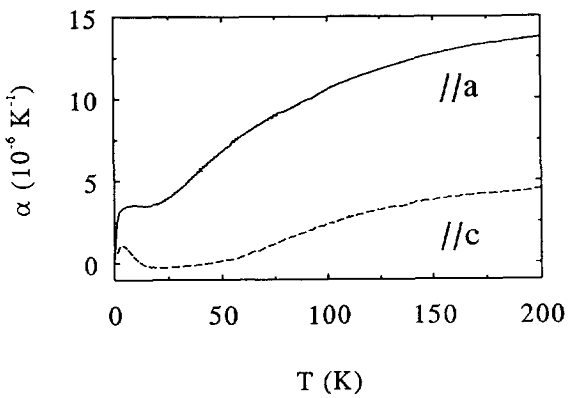

Fig. 1. Thermal expansion of single crystalline $CePd_2Al_3$ in zero field along the a (———) and c (- - -) axes.

tion of the f electrons. This reduces their screening of the nuclear charges, thereby decreasing the ionic radius and unit-cell volume. The maximum in α_V reflects the Kondo-lattice temperature, $T_{coh} \simeq 5$ K, in accord with the maximum in the resistivity [2]. The length change $\Delta L/L$ of the a-axis, obtained after integration of α, is one order of magnitude larger than along c, indicating a different type of coupling in the two directions.

The electronic parts of $\alpha(T)$ and the specific heat $c(T)$ are related by an effective Grüneisen parameter

$$\Gamma_{eff}(T) = \frac{V_{mol}\,\alpha_V(T)}{\kappa\, c(T)}, \tag{1}$$

where V_{mol} is the molar volume (6.586×10^{-5} m^3/mol for $CePd_2Al_3$) and κ the isothermal compressibility, roughly estimated from the pure elements [5] to be 1.605/Mbar. Using the specific-heat results from Ref. [2], we find $\Gamma_{eff} \simeq 20$ at 2 K, which is larger than for UPd_2Al_3 ($\Gamma = 5.5$ just above $T_N = 14$ K [6]).

The low $T_{coh} = 5$ K suggests large effects of a magnetic field on α. We investigated the magnetostriction up to 8 T and α_a in constant fields up to 5 T. Fig. 3 shows the change of α_a with fields parallel to a. For fields as small as 0.3 T, α_a exhibits a minimum, which is absent in zero field. In larger fields α_a changes sign. Magnetostriction data have been taken from $T = 0.5$ to 10 K (not shown). At low T, a change of slope occurs around 1 T, best illustrated in the field derivatives of the magnetostriction, as displayed in Fig. 4. The maximum in $1/L\ \mathrm{d}L/\mathrm{d}H$, clearly visible at 0.5 and 1.2 K, shifts to higher fields for increasing T. The 10 K curve follows the magnetization at this temperature [2], indicating a regular paramagnetic behavior.

Fig. 5 presents magnetoresistance, $\Delta\rho(H)/\rho(0)$ with $\Delta\rho(H) = \rho(H) - \rho(0)$, experiments on $CePd_2Al_3$ with current and field parallel to a, at temperatures from 1.5 to 40 K in fields up to 8 T. The large negative $\Delta\rho$ stems from the reduction of the Kondo scattering with increasing H. Quantitative fitting of the data to the $S = 1/2$ Coqblin–Schrieffer model [7] failed, especially for low T. Nevertheless, we can assign a characteristic field to each of the curves by the minimum in their field derivative. These fields closely correspond to the maxima in the derivative of the magnetostriction.

The magnetic-ordering temperature, T_{ord}, in a Kondo lattice is controlled by competing Kondo

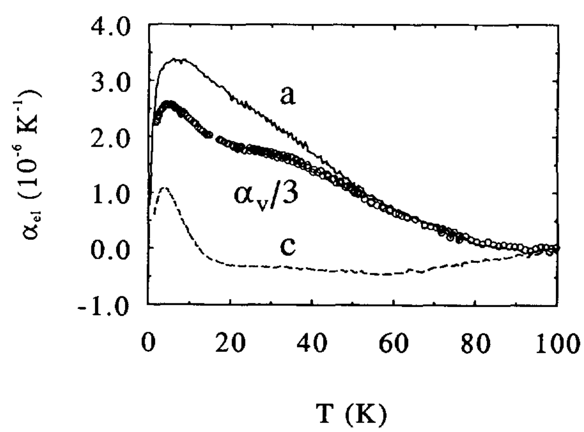

Fig. 2. Electronic part of the thermal expansion of $CePd_2Al_3$. The circles denote the volume expansion, divided by 3.

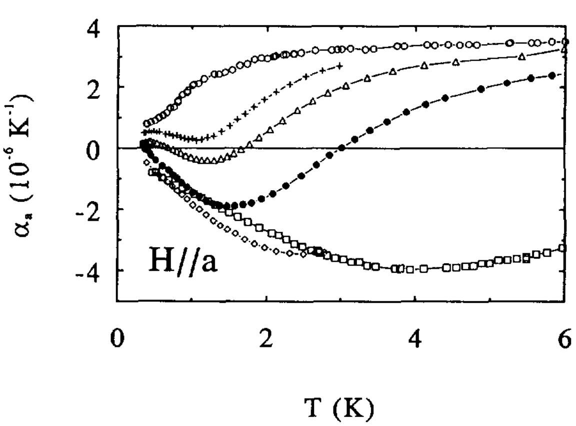

Fig. 3. Thermal expansion of $CePd_2Al_3$ along the a-axis with magnetic fields of 0 (○), 0.3 (+), 0.5 (△), 1 (●), 2.5 (◊) and 4.75 T (□) parallel to the a-axis.

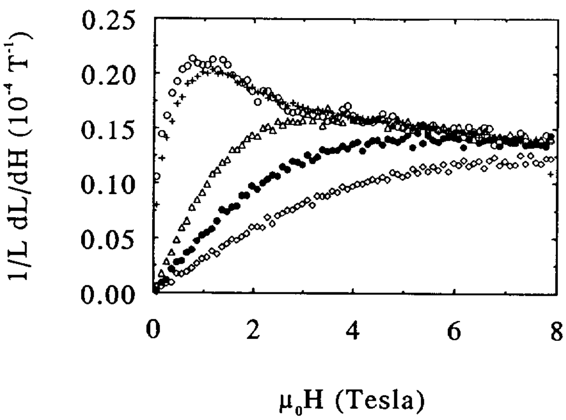

Fig. 4. Field derivative of the length change of the *a*-axis of $CePd_2Al_3$ at various temperatures: (○) 0.5 K, (+) 1.2 K, (△) 4.5 K, (●) 7 K, (◊) 10 K.

screening and RKKY interaction, resulting in the Doniach phase diagram for a one-dimensional array of Kondo ions [8]. For $CePd_2Al_3$, the small Ce–Pd distance leads to large f-d hybridization $J_{f\text{-}d}$. The reduced Ce-moments are confined to the basal plane. Long-range 3D magnetic order is generated by inter-

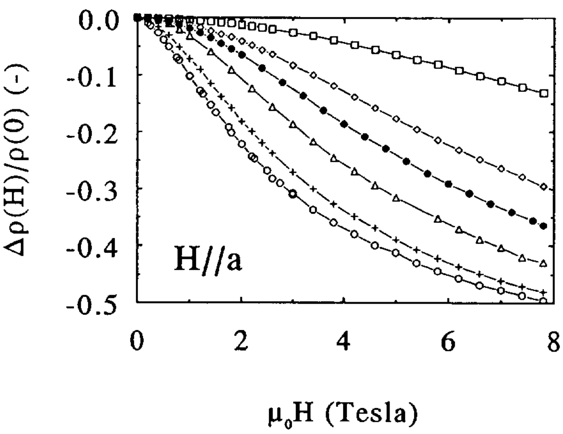

Fig. 5. Typical magnetoresistance curves of $CePd_2Al_3$ with current and field parallel to the *a*-axis at 1.5 K (○), 2.5 K (+), 4.3 K (△), 6 K (●), 8 K (◊) and 15 K (□).

plane magnetic coupling, J_z, via the aluminum layer. This J_z determines the ground state of $CePd_2Al_3$ [3] and involves RKKY-type interactions between the partially hybridized magnetic f-electrons of Ce with the s,p-electrons of Al. The anisotropic exchange implies that the properties of $CePd_2Al_3$ cannot be understood by the Doniach model alone. The large anisotropy allows for a separate treatment of the two interactions: J_{f-d} in the basal plane and J_z between the planes [3]. This analysis is similar to the proposed two nearly separate subsystems of (hybridized) 5f-quasi-particle states in UPd_2Al_3 [9].

The presented thermal expansion experiments have provided new evidence for the crystal-field scheme of $CePd_2Al_3$. The large anisotropy of $\alpha(T)$ has demonstrated the consistency of the anisotropic-hybridization model for $CePd_2Al_3$. The magnetostriction and magnetoresistance results indicate the presence of a characteristic field, associated with the formation of the Kondo lattice.

Acknowledgement

This work was partially supported by the Dutch Foundation FOM.

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