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Magnetovolume effects in Ce_{0.985}Y_{0.015}Ru₂Si₂

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We report on the thermal expansion $\alpha(T)$ and magnetostriction $\lambda(B)$ of monocrystalline $Ce_{0.985}Y_{0.015}Ru_2Si_2$ for 0.1 K < T < 1 K and B < 11 T. The one parameter scaling law previously observed for $CeRu_2Si_2$ and $Ce_{0.95}La_{0.05}Ru_2Si_2$ is also valid for the present alloy. The variation of γ versus B is determined from $\alpha(T)$ in the field. The width of the pseudometamagnetic transition is discussed.

1. Introduction

The observation of metamagnetic-like behaviour in the non-ordered heavy-fermion (HF) compound CeRu₂Si₂ has launched a number of elucidating experiments. Among them, studies of volume effects are very powerful. Let us first recall some of the most important features of this compound. CeRu₂Si₂ exhibits a moderate linear term in the specific heat: $\gamma = 0.35 \text{ J mol}^{-1} \text{ K}^{-2}$ [1,2]. Well below its characteristic temperature $T_m = 9 \text{ K}$, it shows a fermi-liquid ground state [3]. At T_m , a broad maximum is observed in the magnetic susceptibility χ [2,3], in the linear thermal expansion coefficient α [4,5] and in the electronic part of the specific heat [1,2]. Applying a magnetic field along the tetragonal c-axis induces a sharp peak in the differential susceptibility $\chi_{\rm H}$ [=dM/ dB [2,3] and in the coefficient of magnetostriction λ' $[\equiv L^{-1} dL/dB]$ [4,5] for a critical field $B^* = 7.7$ T. A softening of the lattice [6] and a pronounced maximum in γ [2,7] are also observed at B^* . T_m decreases down to about 0.3 K on approaching B^* [4,5]. For fields above B^* , α becomes negative and shows a minimum at a temperature that now increases with the field.

 B^* and T_m constitute the characteristic parameters of a pressure-invariant scaling law [4,8]: $S = S(T/T_m(P), B/B^*(P))$, where in the present case, the magnetic and thermal Grüneisen parameters are equal: $-d \ln B^*/d \ln V = -d \ln T_m/d \ln V = 170$.

* Laboratoire associé à l'Université Joseph Fourier, Grenoble, France Short-range antiferromagnetic correlations, which are generally supposed to play an important role for the formation of HF-quasiparticles can be observed by neutron scattering [9] in CeRu₂Si₂ up to 70 K. At low temperature, these correlations are suppressed in the vicinity of B^* [9]. Nevertheless, the high-field specific heat gives still evidence of a considerable γ -value of 80 mJ mol⁻¹ K⁻² at 20 T [7].

 B^* and T_m increase on hydrostatic [8] or chemical [1] pressure, which is positive when replacing cerium by yttrium but negative in the case of lanthanumdoping. Here, we report on the magnetovolume properties of a single crystal of Ce_{0.985}Y_{0.015}Ru₂Si₂. The results are compared with previous results [4,5] for CeRu₂Si₂ and Ce_{0.95}La_{0.05}Ru₂Si₂.

2. Experiments

A single crystalline sample of $Ce_{0.985}Y_{0.015}Ru_2Si_2$ has been grown by the Czochralski technique. By means of spark erosion, it has been cut into a parallelepiped with edges perpendicular to the *c*- and *a*axes. The measurements of the magnetostriction and the thermal expansion were performed using a sensitive three terminal capacitance-dilatometer which was able to detect length variations in the order of 10^{-2} Å. In a ⁴He cryostat, we obtained temperatures between 1.3 and 40 K and fields up to 8 T and in a dilution cryostat, temperatures between 0.1 and 1 K and fields up to 11 T.

The cell was placed in the cryostat with the elongation direction parallel or perpendicular to the magnetic field. From continuous field sweeps, we obtained

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the magnetostriction coefficient $\lambda = dL/L$ and its field derivative λ' and from step-like temperature variations, the coefficient of linear thermal expansion $\alpha = 1/L(dL/dT)_B$. The corresponding volume coefficients are obtained by $\lambda_v = \lambda_{\parallel} + 2\lambda_{\perp}$ ($\alpha_v = \alpha_{\parallel} + 2\alpha_{\perp}$) where the subscripts \parallel and \perp indicate the direction of elongation with respect to the magnetic field directed along the tetragonal *c*-axis. A magnetic field applied along the *a*-axis did not produce any noticeable magnetovolume effect. As in CeRu₂Si₂ itself, we found for the magnetostriction and thermal expansion coefficients a constant ratio $\alpha_{\parallel}/\alpha_{\perp} = \lambda_{\parallel}/\lambda_{\perp} = 3$.

3. Results and analysis

The variations of λ' at several temperatures between 4.2 and 0.1 K for Ce_{0.985}Y_{0.015}Ru₂Si₂ are shown in fig. 1. From the location of the maximum, we obtain $B^* = 9.36$ T for temperature $T \rightarrow 0$. At B = 0, α shows a maximum at $T_m = 10.6$ K. The variations of α at different fields up to 11 T between 0.2 and 1 K are shown in fig. 2. At B^* , α changes sign. A maximum is expected slightly above 1 K at 9.25 T. At 9.5 T, a clear minimum can be seen at 0.65 K. This mimics the B^*-T_m diagram previously reported for the pure $(B^* = 7.7 \text{ T}, T_m = 9 \text{ K})$ and 5% La $(B^* = 5.3 \text{ T}, T_m = 6 \text{ K})$ compounds.

The scaling law [4,8] $\lambda' = B\kappa\Gamma(dM/dH)_{P,T}$ has been verified by comparing the magnetostriction data of fig. 1 with differential susceptibility data derived from magnetization measurements of the same crystal.



Fig. 1. Coefficient of linear magnetostriction of $Ce_{0.985}Y_{0.015}Ru_2Si_2$ along the *c*-axis around the pseudometamagnetic field B^* ($||c\rangle$) at different temperatures: (a) 0.1 K, (b) 0.3 K, (c) 0.5 K, (d) 0.7 K, (e) 0.9 K, (f) 1.3 K and (g) 4.2 K.



Fig. 2. Linear thermal expansion coefficient of $Ce_{0.985}Y_{0.015}Ru_2Si_2$ along the *c*-axis between 0.2 and 1 K for different applied magnetic fields (||c|).

Assuming $\gamma_0 B^* = \text{constant}$ for all non-ordered $\text{Ce}_x(\text{La}, \text{Y})_{1-x} \text{Ru}_2 \text{Si}_2$ alloys, gives an estimate of the zero-field linear term of the specific heat γ_0 to $0.3 \text{ J} \text{ mol}^{-1} \text{ K}^{-2}$ for $\text{Ce}_{0.985} \text{Y}_{0.015} \text{Ru}_2 \text{Si}_2$. The Grüneisen parameter $\Gamma = V_m a / (\kappa \gamma)$ amounts to 150 where the molar volume $V_m = 5.17 \times 10^{-5} \text{ m}^3 \text{ mol}^{-1}$ and the isothermal compressibility $\kappa = 0.95 \times 10^{-11} \text{ m}^2/\text{N}$ are set equal to the data of the pure compound. The parameter $a = \alpha / T = 8.5 \times 10^{-6} \text{ K}^{-2}$ is the slope of the coefficient of the linear regime of α below T_m at B = 0.

From the different values of a in an applied field (fig. 2), we can deduce the field dependence of γ using the differential equation [4]

$$a(B) = \Gamma \kappa V_{\rm m}^{-1}(\gamma + B({\rm d}\gamma/{\rm d}B)) . \tag{1}$$

The numerical solution of eq. (1) is displayed in fig. 3. At B^* , γ has a maximum of 0.48 J mol⁻¹ K⁻², i.e. an increase of 60% with respect to its zero field value. In pure CeRu₂Si₂ and the 5% La-alloy increases close to 60% and 30%, respectively, were derived from low temperature magnetization data [5] as reproduced in fig. 3. There is good agreement between these two latter calculated variations and the experimental values [2,7]. Measurements of the specific heat should also be performed on Ce_{0.985}Y_{0.015}Ru₂Si₂ in order to confirm the variation of γ derived from eq. (1).

From our magnetostriction data (fig. 1), we determined the width dB of the pseudo-metamagnetic transition (PMT). In order to obtain the full width without passing beyond the experimental high-field limit, we measured dB at 75% of the maximum value. The results are shown in fig. 4 on a renormalized scale $(\log(dB/B^*)$ versus $T/T_m)$ and are compared with the



Fig. 3. Field dependence of the coefficient γ of the specific heat, calculated from the present thermal expansion data using eq. (1) for Ce_{0.985}Y_{0.015}Ru₂Si₂, and derived from magnetization data [5] for CeRu₂Si₂ and Ce_{0.95}La_{0.05}Ru₂Si₂.

data for CeRu₂Si₂ and Ce_{0.95}La_{0.05}Ru₂Si₂. For the three compounds, the extrapolated values of dB/B^* at $T \rightarrow 0$ depends almost linearly on the impurity concentration (inset of fig. 4). The pure compound shows a transition width for $T \rightarrow 0$ which is about one order of magnitude smaller than in the alloys.

4. Discussion

The present experiments show that the sharpest increase of γ in the vicinity of B^* is obtained for the assumed pure lattice of CeRu₂Si₂. The relative width of the pseudometamagnetic anomaly dB/B^* increases



Fig. 4. Log plot of the normalized transition width dB/B^* versus normalized temperature T/T_m , for $CeRu_2Si_2$, $Ce_{0.985}Y_{0.015}Ru_2Si_2$, and $Ce_{0.95}La_{0.05}Ru_2Si_2$, with guidelines to the eye. Inset shows dB/B^* for $T \rightarrow 0$ versus impurity concentrations.

drastically on doping either with La or Y, i.e. whatever is the relative change of the characteristic parameters T_m and B^* (decreasing on substituting by La, increasing on substituting by Y according to the relative volume variation [1]). There is some evidence for a linear dependence of the relative transition width $(T \rightarrow 0)$ on the impurity concentration. This underlines the importance of the lattice invariance. For confirmation, further experiments must be performed.

The broadening of the PMT may result from doping-induced volume inhomogeneities within the sample. However, the relative volume effect of Y-doping is about two times larger than La-doping [1]. A weighting factor of two on the Y concentration yields a linear dependence of the absolute transition width dB on the impurity concentration. A simple model taking into account the volume inhomogeneities [5] could not reproduce the experimental findings sufficiently. A more elaborated model is desirable.

A yet unsolved question is the importance of the electronic mean free path since the measured pure CeRu₂Si₂ lattice has still a rather large residual resistivity ($\rho_0 \sim 2 \,\mu\Omega \,\text{cm}$ [3]) while doping induces an increase of $1 \,\mu\Omega \,\text{cm}/\%$ for La and of $3 \,\mu\Omega \,\text{cm}/\%$ for Y. Thus, in contrast to the transition width, ρ_0 does not depend linearly on the impurity concentration since it is of the same order of magnitude ($\sim 6 \,\mu\Omega \,\text{cm}$) for the two alloys.

In conclusion, the transition width can be discussed in the light of two opposing ideas. In the first, the local volume inhomogeneities are considered which may act on the short range antiferromagnetic correlations. In the second, the itinerant character of the HF system prevails, i.e. the extended Fermi liquid state is perturbated by the impurities. A final explanation is not yet possible. The observation of a nonvanishing transition width for $T \rightarrow 0$ even in the purest crystal of CeRu₂Si₂ seems to show that there is no first order transition at B^* .

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References

- M.J. Besnus, J.P. Kappler, P. Lehmann and A. Meyer, Solid State Commun. 55 (1985) 779.
- [2] R.A. Fisher, C. Marcenat, N.E. Phillips, P. Haen, F. Lapierre, P. Leyay, J. Flouquet and J. Voiron, J. Low Temp. Phys. 84 (1991) 49.
- [3] P. Haen, J. Flouquet, F. Lapierre, P. Lejay and G. Remenyi, J. Low Temp. Phys. 67 (1987) 391.

- [4] A. Lacerda, A. de Visser, L. Puech, P. Lejay, P. Haen, J. Flouquet, J. Voiron and F.J. Okhawa, Phys. Rev. B 40 (1989) 11429.
- [5] C. Paulsen, A. Lacerda, L. Puech, P. Haen, J.L. Tholence, P. Lejay, J. Flouquet and A. de Visser, J. Low Temp. Phys. 81 (1990) 317.
- [6] G. Bruls, D. Weber, B. Lüthi, J. Flouquet and P. Lejay, Phys. Rev. B 42 (1990) 4329.
- [7] H.P. van der Meulen, A. de Visser, J.J.M. Franse,

T.T.J.M. Berendschot, J.A.A.J. Perenboom, H. van Kempen, A. Lacerda, P. Lejay and J. Flouquet, Phys. Rev. B 44 (1991) 814.

- [8] J.M. Mignot, J. Flouquet, P. Haen, F. Lapierre, L. Puech and J. Voiron, J. Magn. Magn. Mater. 76 & 77 (1988) 97.
- [9] J. Rossat-Mignod, L.P. Regnault, J.L. Jacoud, C. Vettier, P. Lejay, J. Flouquet, E. Walker, D. Jaccard and A. Amato, J. Magn. Magn. Mater. 76 & 77 (1988) 376.