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MAGNETOSTRICTION OF MONOCRYSTALLINE URu_2Si_2

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We report magnetostriction measurements on a monocrystalline sample of the heavy-fermion compound URu_2Si_2, below (at 4.2 K) and above (at 20 K) the Néel temperature. For a field direction along the tetragonal axis the magnetostriction is strongly anisotropic; the volume effect amounts to $1.4 \times 10^{-6}\,T^{-1}$ at 8 T and 4.2 K. For a field direction in the tetragonal plane the magnetostriction is nearly two orders of magnitude smaller, and less anisotropic.

1. Introduction

Among the ternary uranium compounds URu_2Si_2 attracts much attention, because it exhibits the unusual coexistence of antiferromagnetism and superconductivity [1–4]. The antiferromagnetic transition (at $T_N = 17.5$ K) is observed as a λ-like anomaly in the specific heat, a bump in the resistivity and a kink in the susceptibility curve. Superconductivity occurs below ~1 K. The bulk nature of the superconductivity has been demonstrated by specific-heat [1–3] and Meissner-effect [1] experiments. Analyses of the upper critical field [1, 3, 5], utilizing the relatively large value of the electronic specific heat just above T_c ($\gamma \cong 50\,\mathrm{mJ/mol\,K^2}$), yield an effective electron mass of about 50 times the free-electron mass. Therefore, URu_2Si_2 is classified as a moderately heavy-fermion system.

Experiments performed on monocrystalline samples have revealed that the magnetic properties are strongly anisotropic. Furthermore, anomalies occur for a field direction along the tetragonal axis (*c*-axis). The susceptibility (measured in a field of 2 T) for $B \| c$ deviates from a Curie–Weiss behaviour below 150 K and develops a broad maximum around 50 K, whereas for $B \| a$ (i.e. B in the tetragonal plane) the susceptibility exhibits hardly any temperature dependence below room temperature [2]. For both field directions T_N is only reflected by a small kink. High-field magnetization [6, 7] and magnetoresistivity [7] experiments, performed for $B \| c$ (at 1.5 K), revealed three sharp transitions above 35 T, suggesting a complex magnetization process. For $B \| a$ the magnetization is linear up to 40 T.

Other physical properties, e.g. electrical resistivity [5], thermal expansion [8] and Hall effect [9], were found to be strongly anisotropic as well.

Recently, it was demonstrated by neutron-scattering experiments [4] that the ordered moment (directed along the *c*-axis) is usually small: $0.03 \pm 0.01\mu_B$. Furthermore, these data suggest that URu_2Si_2 is a singlet ground-state system, with a weak induced moment. This supports the claim of Nieuwenhuys [10] that the system can be described assuming localized $5f^2$ states in the $^3H_4$ configuration, the $J = 4$ multiplet being split into five singlets and two doublets by the tetragonal crystal field. However, as has been pointed out in ref. [4], the shortcomings of this simple model call for a more refined theory.

The purpose of this paper is to report on magnetostriction measurements, that have been performed well below (at 4.2 K), and just above (at 20 K), the Néel temperature. Since the magnetic properties of URu_2Si_2 were found to be strongly anisotropic, the experiments were performed on a monocrystalline sample.

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2. Experimental

URu_2Si_2 crystallizes in the tetragonal $ThCr_2Si_2$-type of structure. The lattice parameters equal $a = b = 4.1239(2)$ Å and $c = 9.5817(8)$ Å at 4.2 K [2]. A large single-crystalline bar was prepared by the Czochralski technique. After a subsequent annealing procedure, a cube ($5 \times 5 \times 5$ mm^3) was cut from the bulk crystal by means of spark erosion. The cubic edges are parallel to the crystallographic directions. Thermal-expansion measurements on this sample were reported in ref. [8].

In order to measure its magnetostriction, the sample was mounted in a capacitance cell, made of OFHC copper. A sensitive three-terminal method served to determine the length changes. The field was provided by a superconducting magnet ($B_{max} = 8$ T). Data were gathered by raising the field stepwise ($\Delta B = 0.5$ T), at fixed temperatures. Since the coefficients of thermal expansion are rather large in the investigated temperature region ($\alpha_a = 0.6 \times 10^{-6}$ and $\alpha_c = -0.5 \times 10^{-6}$ K^{-1} at 4.2 K; $\alpha_a = 5.3 \times 10^{-6}$ and $\alpha_c = -1.8 \times 10^{-6}$ K^{-1} at 20 K), special care was taken to keep the sample at constant temperature. At 20 K this was accomplished by utilizing a field-insensitive capacitance thermometer. The experimental uncertainty amounts to 5×10^{-8} T^{-1} at the maximum field.

In our experimental set up nine combinations for the field and dilatation directions are possible: for a certain field direction the magnetostriction can be measured parallel and perpendicular (2×) to the field. The tetragonal symmetry reduces these nine combinations to five.

3. Results

For $B \| c$ the magnetostriction has been measured along the field (λ_c) and perpendicular to the field (λ_a and λ_b). For sake of clarity: the a and b-axis correspond to the [100] and [010] direction, respectively. The results obtained at 4.2 K and 20 K, are shown in a plot of $\lambda' = L^{-1}(dL/dB)$ versus B, in fig. 1 and fig. 2, respectively. The volume magnetostriction has

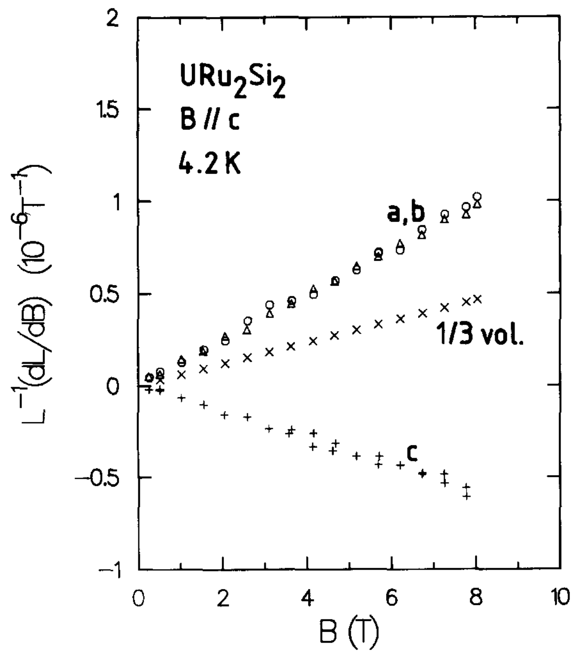

Fig. 1. Magnetostriction ($B \| c$) of URu_2Si_2 at 4.2 K, along the a- (○), b- (△) and c-axis (+), and $\lambda'_v/3$ (×).

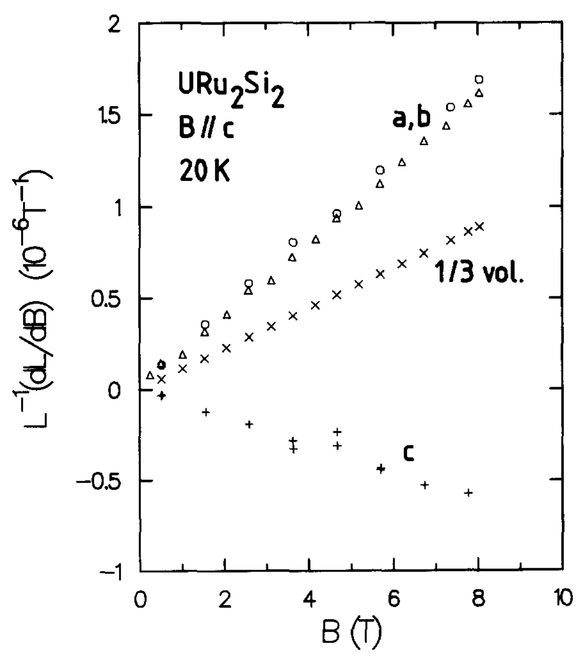

Fig. 2. Magnetostriction ($B \| c$) of URu_2Si_2 at 20 K, along the a- (○), b- (△) and c-axis (+), and $\lambda'_v/3$ (×).

Table I
Coefficients (in $10^{-7}\,T^{-2}$) of the B^2 term in the magnetostriction of URu_2Si_2 for $B \| c$.

	$T=4.2$ K	$T=20$ K
dilation along:		
basal plane	0.60	1.02
c-axis	−0.36	−0.36
volume effect	0.88	1.68

been calculated according to $\lambda_v = \lambda_a + \lambda_b + \lambda_c$. Apparently, the magnetostriction is strongly anisotropic. Applying a field along the c-axis, results in an expansion of the basal plane and a contraction along the tetragonal axis. In the basal plane λ is isotropic, as is to be expected for the tetragonal symmetry. For all dilatation directions we find that $\lambda \propto B^2$. Coefficients of the B^2 term are listed in table I.

For a field in the basal plane along the a-axis the magnetostriction was measured along the a, b and c-axis (fig. 3 and fig. 4). For this field direction λ' is smaller than $5 \times 10^{-8}\,T^{-1}$, at 8 T, along the three measured dilatation directions.

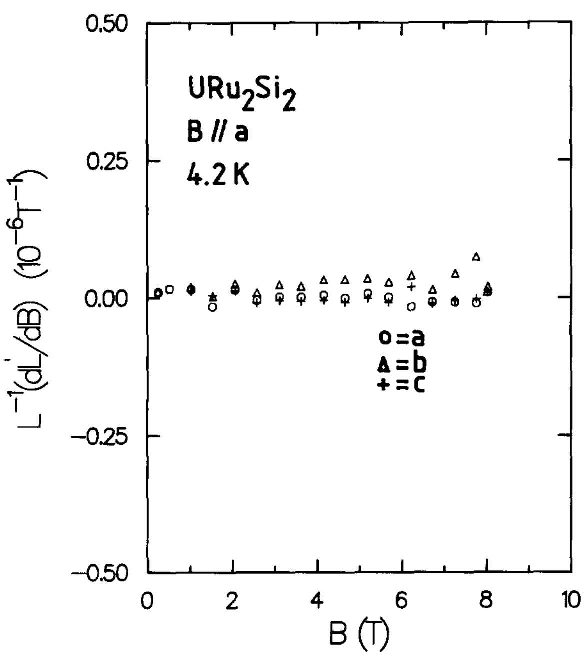

Fig. 3. Magnetostriction ($B \| a$) of URu_2Si_2 at 4.2 K, along the a- (○), b- (△) and c-axis (+).

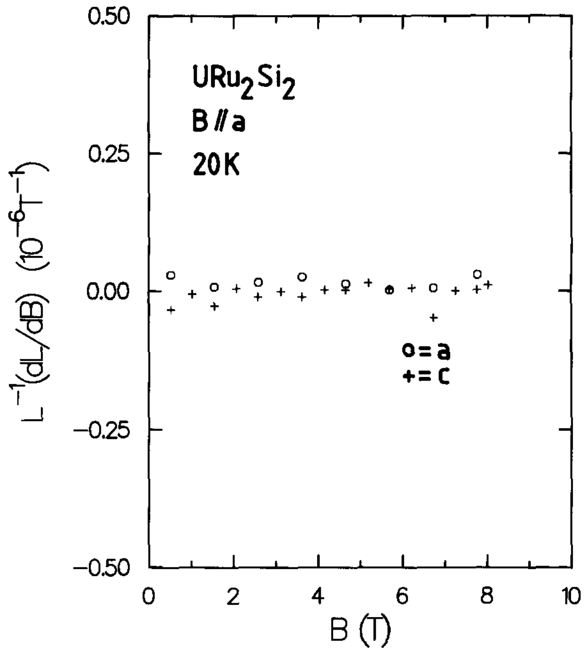

Fig. 4. Magnetostriction ($B \| a$) of URu_2Si_2 at 20 K, along the a- (○) and c-axis (+).

4. Discussion

Comparing the data for $B \| c$ in the antiferromagnetic phase with the data above T_N, it is observed that the measured curves are much alike. The increase of the volume effect at the highest temperature is caused by the larger values for λ_a and λ_b; λ_c is temperature independent. The similarity of the data at these temperatures demonstrates that the occurrence of long-range magnetic order hardly influences the magneto-elastic properties. Therefore, it is likely – in view of forementioned properties of URu_2Si_2 – that the magneto-elastic coupling parameters are of the crystal-field type.

From the temperature dependence of λ_v' ($B \| c$) it follows that a relation of the type

$$\omega \equiv \frac{\Delta V}{V} = \rho \kappa C M^2 \tag{1}$$

is obeyed, where ρ is the density, κ is the compressibility, C is the magneto-volume parameter and M is the molar magnetic moment. The field derivative (at constant temperature) of eq. (1) can be written as

$$\left(\frac{\partial \omega}{\partial B}\right)_T = \lambda_v' = 2\rho\kappa C\chi^2 \frac{H}{\mu_0} . \quad (2)$$

Substituting in relation (2) the values for λ_v' ($B\|c$) from table I and the values for χ_c of 70×10^{-9} m^3/mol and 96×10^{-9} m^3/mol [2], at 4.2 K and 20 K, respectively, we compute $\rho\kappa C = 2.8 \times 10^{-5}$ (Am^2/mol)$^{-2}$. Using the value for ρ of 1.01×10^4 kg/m^3 and the estimated value for κ of 0.73×10^{-11} m^2/N [8], the magneto-volume parameter amounts to 3.8×10^2 mol^{2}T^{2}J^{-1}kg^{-1}.

The forced-magnetostriction data can be related to the hydrostatic pressure dependence of the molar magnetic moment by the thermodynamic relation:

$$\left(\frac{\partial M}{\partial p}\right)_{H,T} = -\left(\frac{\partial V}{\partial \mu_0 H}\right)_{p,T} , \quad (3)$$

or in terms of the relative pressure dependence of the molar magnetic susceptibility

$$\frac{H\chi}{V}\left(\frac{\partial \ln \chi}{\partial p}\right)_{H,T} = -\frac{1}{V}\left(\frac{\partial H}{\partial \mu_0 H}\right)_{p,T} . \quad (4)$$

For a field direction along the c-axis, we derive, inserting the value for the susceptibility at zero pressure and the appropriate values from table I in eq. (4), values for $\partial \ln \chi/\partial p$ of -15 ± 3 Mbar^{-1} and -22 ± 3 Mbar^{-1}, whereas the experimental values for $\partial \ln \chi/\partial p$ amount to -29 ± 5 Mbar^{-1} [11] and -21 ± 5 Mbar^{-1} [12], at 4.2 K and 20 K, respectively. The origin of the discrepancy at 4.2 K is not clear. Since the experiments have been performed on different samples, sample dependence might play a role. However, one should bear in mind that the pressure-induced volume changes are larger, by more than a factor 10^2, than the field-induced volume changes. Therefore, a non-linear pressure dependence of χ in the antiferromagnetic phase cannot be excluded.

To summarize: we reported magnetostriction experiments on a monocrystalline sample of URu_2Si_2, just above and well below the Néel temperature. For a field in the tetragonal plane the magnetostriction is small, whereas for a field along the tetragonal axis a large and anisotropic magnetostriction is observed.

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