



Dilatometry study of the heavy-fermion superconductor URu_2Si_2

N.H. van Dijk*, A. de Visser, J.J.M. Franse, A.A. Menovsky

Van der Waals–Zeeman Laboratory, University of Amsterdam, Valckenierstraat 65, 1018 XE Amsterdam, The Netherlands

Abstract

Dilatation measurements have been performed on a single-crystalline sample of the heavy-fermion superconductor URu_2Si_2 ($T_c = 1.2$ K). Thermal expansion measurements indicate a thermal electronic Grüneisen parameter $\Gamma_T = 27$. A comparison with the magnetic electronic Grüneisen parameter derived from the magnetostriction, $\Gamma_B = 26$, points to a single energy scale. Magnetostriction measurements in the superconducting state reveal hysteresis with a peculiar change of sign close to the upper critical field, B_{c2} .

The heavy-fermion superconductor URu_2Si_2 attracts considerable attention because of the unusual co-existence of superconductivity and antiferromagnetism [1,2]. Long-range antiferromagnetic order is found below the Néel temperature $T_N = 17.5$ K. Neutron-diffraction experiments [3] revealed that URu_2Si_2 is an antiferromagnet with an extremely small ordered moment of $0.03\mu_B$ (on the uranium atom) oriented along the tetragonal axis (c -axis). Superconductivity is found below $T_c = 1.2$ K [1]. The upper critical field of the superconducting state, B_{c2} , is strongly anisotropic and unusually large: $B_{c2}(T \rightarrow 0)$ amounts to 14 T for $B \perp c$ and to 3 T for $B \parallel c$ [4]. Recent point-contact spectroscopy data [5] revealed that URu_2Si_2 has a line of nodes in the superconducting gap. An anisotropic gap structure (with nodes in the gap) is in general attributed to unconventional superconductivity ($L \neq 0$).

Dilatation measurements were performed on a single-crystalline sample of dimensions $4 \times 3 \times 5 \text{ mm}^3$ ($a \times a \times c$) using a sensitive three-terminal parallel-plate capacitance technique. The coefficient of linear thermal expansion, $a_i = L^{-1}dL/dT$, where i refers to the a - or the c -axis of the tetragonal crystal, was

measured stepwise ($\Delta T \approx 40$ mK). The temperature variation of α_a and α_c of URu_2Si_2 at low temperature is shown in Fig. 1. The corresponding coefficient of volume expansion $\alpha_v = 2\alpha_a + \alpha_c$ is given by the solid line. For $T > T_c$, α_a , α_c and α_v vary linearly with temperature as does the specific heat ($c = \gamma T$) [1]. The linear coefficients of the thermal expansion amount to

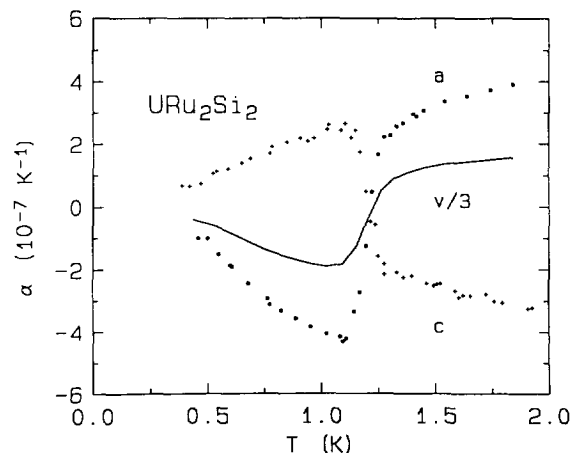


Fig. 1. Linear thermal expansion α_a (●) and α_c (+). The volume effect divided by three is indicated by the solid line.

* Corresponding author.

$a_a = \alpha_a/T = 0.22 \times 10^{-6} \text{ K}^{-2}$ and $a_c = \alpha_c/T = -0.16 \times 10^{-6} \text{ K}^{-2}$. At the superconducting transition ($T_c = 1.18 \text{ K}$) α_a and α_c exhibit sharp jumps and change sign. The results displayed in Fig. 1 are in good agreement with previous measurements on a single-crystalline sample [6], which exhibited a much broader transition.

The linear magnetostriction, $\lambda = [L(B) - L(0)]/L(0)$, was measured for a dilatation (contraction) direction along and perpendicular to the magnetic field ($B \parallel c$) by recording the capacitance change while sweeping the magnetic field at a low rate ($dB/dt \leq 0.1 \text{ T/min}$). The curves of linear magnetostriction measured in the normal state at $T = 1.5 \text{ K}$ are shown in Fig. 2. The volume magnetostriction is given by $\lambda_v = 2\lambda_a + \lambda_c$. The magnetostriction in the normal state behaves like $\lambda = bB^2$, characteristic for paramagnetic systems. Previous magnetostriction data ($B \parallel c$) at 4.2 and 20 K [7] yielded a similar behaviour.

In the superconducting state, the linear magnetostriction shows a pronounced hysteresis as shown in Fig. 3 for λ_c and $B \parallel c$. The hysteresis loop closes at B_{c2} . For $B > B_{c2}$ the usual B^2 behaviour is observed. Remarkably, at the lowest temperatures, in the superconducting state, the hysteresis changes sign at a field close to B_{c2} . With increasing temperature, the inversion point of the hysteresis moves to lower field values, and, finally, vanishes for a temperature close to 1.0 K. Data for λ_a and λ_c obtained by the discrete method, i.e. by stepwise increasing the magnetic field ($\Delta B = 0.2 \text{ T}$) at a temperature of 0.5 K, are shown in Fig. 4. In Fig. 4, a $\lambda = bB^2$ term, representing the

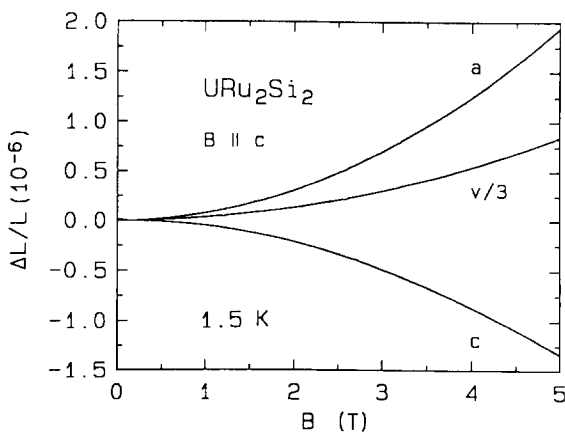


Fig. 2. Normal-state linear magnetostriction of URu_2Si_2 for $B \parallel c$ at 1.5 K. The calculated volume magnetostriction divided by three is indicated by $v/3$.

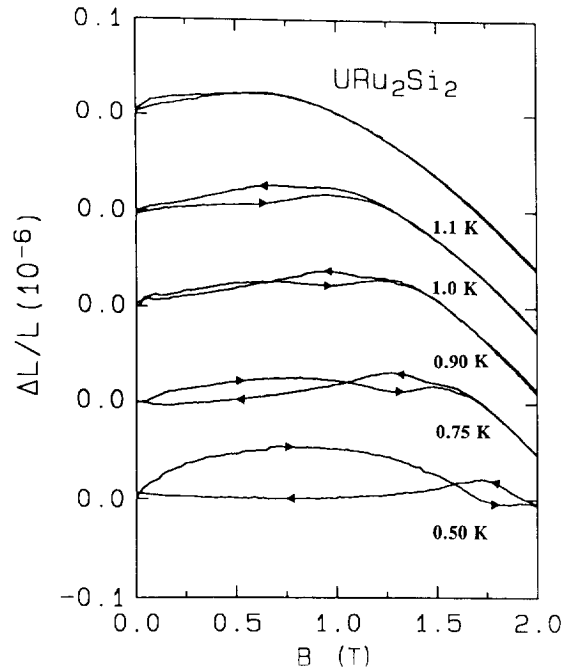


Fig. 3. Sweep measurements of the linear magnetostriction λ_c at $T = 0.50, 0.75, 0.90, 1.0$ and 1.1 K for $B \parallel c$. The magnetostriction curves are shifted along the vertical axis for the sake of clarity.

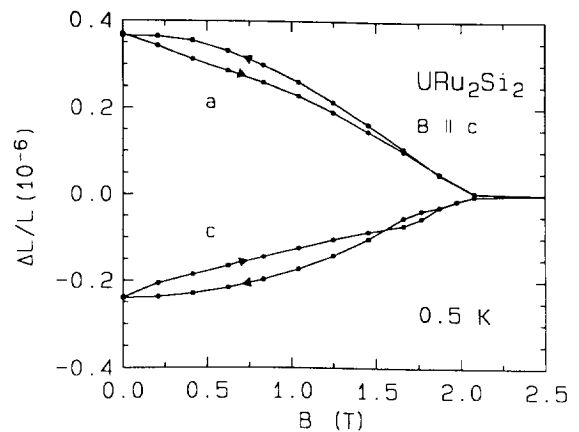


Fig. 4. Discrete measurement of the linear magnetostriction λ_a and λ_c in the superconducting state ($T = 0.5 \text{ K}$) for $B \parallel c$. The quadratic field dependence of the normal state is subtracted. The magnetostrictive hysteresis for λ_c in the superconducting state changes sign close to the upper critical field where the hysteresis loop closes. The solid lines are guides to the eye.

normal-state contribution determined in the field range $B > B_{c2}$ (≈ 2 T), is subtracted. The results of the discrete and sweep methods are in good agreement. Note that in low fields the hysteresis in λ_a has a different sign compared to the hysteresis in λ_c . The afore-mentioned inversion of the hysteresis close to B_{c2} for λ_c is not observed for λ_a . However, the hysteresis in λ_a becomes negligibly small in that field region. The complex behaviour of the magnetostriction is illustrated by a sign reversal of the hysteresis loop in λ_a by raising the temperature to 1.0 K.

In order to study the volume dependence of the thermal and magnetic energy scales of the heavy-fermion state a Grüneisen parameter analysis may be performed [8]. One can introduce an experimental effective thermal Grüneisen parameter $\Gamma_{\text{eff}}(T)$:

$$\Gamma_{\text{eff}}(T) = V_m \alpha_v(T) / \kappa c(T) \quad (1)$$

where V_m is the molar volume, $\kappa = -V^{-1}dV/dp$ is the isothermal compressibility and c is the molar specific heat at constant volume. In the low-temperature limit Γ_{eff} attains a large constant value for heavy-fermion systems which equals the thermal Grüneisen parameter, Γ_T , as only the linear terms in the thermal expansion ($\alpha_v = a_v T$) and the specific heat ($c = \gamma T$) are retained. In the case of URu₂Si₂, we obtain $\Gamma_T = 27$, where we used $a_v = 0.27 \times 10^{-6} \text{ K}^{-2}$, $\gamma = 67 \text{ mJ/mol K}^2$, $V_m = 4.9 \times 10^{-5} \text{ m}^3/\text{mol}$ and a value for the compressibility estimated at 0.73 Mbar^{-1} [8]. When superconductivity sets in, Γ_{eff} changes sign and takes the value -27 . For a quadratic field dependence of the volume magnetostriction, $\lambda_v = b_v B^2$, the volume dependence of the molar susceptibility, $\chi = \mu_0 M/B$, can be calculated from the relation: $d \ln \chi / d \ln V = 2b_v \mu_0 V_m / \kappa \chi$. The molar susceptibility of URu₂Si₂ at 1.5 K is given by $\chi = 62 \times 10^{-9} \text{ m}^3/\text{mol}$ for $B \parallel c$ [1]. Using the coefficient b_v from the magnetostriction curves of Fig. 2, we obtain a value of $d \ln \chi / d \ln V = 25$. The magnetic Grüneisen parameter, Γ_B , is determined by: $d \ln \chi / d \ln V = 2\Gamma_B - \Gamma_T$ where χ is independent of temperature and magnetic field [9]. The evaluated value of Γ_B at 1.5 K is 26. For $B \parallel c$, we find that $\Gamma_B \approx \Gamma_T$, indicating that the magnetic and thermal properties of the electronic system are strongly coupled in URu₂Si₂.

The magnetostriction in the superconducting state can be described in terms of the spontaneous and

forced magnetostriction. The spontaneous magnetostriction is equal to the zero-field values of λ_a and λ_c in Fig. 4 and reflects the pressure dependence of the condensation energy. It can also be derived by integrating the superconducting contributions to α_a and α_c . The length changes, $(L_s - L_n)/L_n$, derived from the thermal expansion are $\Delta\lambda_a = 0.37 \times 10^{-6}$ and $\Delta\lambda_c = -0.23 \times 10^{-6}$ at 0.5 K, in good agreement with the zero-field values of Fig. 4. The forced magnetostriction is caused by the diamagnetic shielding of the external field and may show hysteresis for increasing and decreasing fields. The most obvious origin of this hysteresis is the effect of flux pinning, which leads to different flux profiles in the sample for increasing and decreasing fields. For a magnetostriction perpendicular to the direction of field, the length of the sample is smaller for increasing fields than for decreasing fields [10]. For a magnetostriction along the field the hysteresis has opposite sign. In the low-field and low-temperature range, the appropriate sign for the discussed flux pinning effects is found, while in the region close to B_{c2} an opposite sign for the hysteresis is observed. An opposite sign for the magnetostrictive hysteresis has also been observed for UBe₁₃ [11]. The additional contribution of the magnetostriction near B_{c2} may be related to an interaction of the vortices and the heavy quasiparticles.

This work was part of the research programme of the ‘Stichting FOM’ (Dutch Foundation for Fundamental Research of Matter).

References

- [1] T.T.M. Palstra et al., Phys. Rev. Lett. 55 (1986) 2727.
- [2] A. de Visser et al., Phys. Rev. B 34 (1986) 8168.
- [3] C. Broholm et al., Phys. Rev. Lett. 58 (1987) 1467.
- [4] J.P. Brison et al., Physica B 199–200 (1994) 70.
- [5] K. Hasselbach et al., Phys. Rev. B 46 (1992) 5826.
- [6] A. de Visser et al., Physica B 165–166 (1990) 375.
- [7] F.E. Kayzel et al., Physica B 147 (1987) 231.
- [8] A. de Visser et al., Physica B 161 (1989) 324.
- [9] P. Thalmeier et al., Europhys. Lett. 1 (1986) 367.
- [10] H. Ikuta et al., Phys. Rev. Lett. 70 (1993) 2166.
- [11] A. de Visser et al., Phys. Rev. B 45 (1992) 2962.