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Journal of Magnetism and Magnetic Materials 272–276 (2004) e15–e16
Journal of magnetism and magnetic materials
www.elsevier.com/locate/jmmm

Thermal expansion measurements on a ferromagnetically ordered single-crystal URhGe

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Abstract

We have carried out thermal expansion measurements on single-crystalline URhGe in the temperature range from 2 to 200 K. The hydrostatic pressure dependence dT_C/dp at ambient pressure was found to be $+0.12$ K/kbar. The low-temperature electronic Grüneisen parameter $\Gamma_{sf} = 14$ indicates an enhanced volume dependence of the ferromagnetic spin fluctuations at low temperatures.

PACS: 65.40.De; 75.30.Kz; 75.50.Cc

Keywords: Thermal expansion; URhGe; Ferromagnetism

Recently, the intermetallic compound URhGe has attracted much attention because superconductivity ($T_c = 0.25$ K) was found to coexist with ferromagnetism (Curie temperature $T_C = 9.5$ K) [1]. URhGe crystallizes in the orthorhombic TiNiSi-type structure (space group P_{nma}). The unit cell, with dimensions $a = 6.87$ Å, $b = 4.33$ Å, and $c = 7.51$ Å, contains 4 formula units. Neutron-diffraction experiments on single-crystalline URhGe [2] revealed a collinear ferromagnetic order below $T_C = 9.6$ K with ordered U moments of $0.3\,\mu_B$ confined to the $b-c$ plane. No component of the ordered moment was observed along the a-axis, which acts as the hard magnetization direction.

In this paper, we report thermal expansion measurements on single-crystalline URhGe in the temperature range from $T = 2$ to 200 K. Our principal aim was to determine the pressure dependence of the ferromagnetic transition temperature T_C. For a second-order phase transition, the pressure dependence of T_C (at ambient pressure) can be determined with the Ehrenfest relation from the anomalies in the linear coefficient of thermal expansion and the specific heat. The initial pressure dependence may give an estimate of the critical pressure needed to suppress the ferromagnetic order and reach the quantum critical point at $T_C = 0$. In addition, we have determined the electronic Grüneisen parameter, which characterizes the volume dependence of the ferromagnetic spin fluctuations at low temperatures.

The linear thermal expansion coefficient $\alpha(T) = (1/L)(dL/dT)$ was measured with a parallel-plate capacitance dilatometer along the orthorhombic a-, b-, and c-axis of a single crystalline sample of dimensions $a \times b \times c = 2.4 \times 5.0 \times 2.4\,\text{mm}^3$. From these measurements the volume expansion $\alpha_v = \alpha_a + \alpha_b + \alpha_c$ was determined.

In Fig. 1 the linear thermal expansion coefficient α along the a-, b-, and c-axis is shown as a function of temperature for $T = 2$–200 K. The temperature dependence of the volume expansion $\alpha_v = \alpha_a + \alpha_b + \alpha_c$ is shown for comparison. At high temperatures the thermal expansion is governed by the phonon contribution, closely resembling a Debye curve with estimated Debye temperature of $\theta_D \approx 200$ K. Around a temperature of 25 K a remarkable crossing of the three curves and a clear deviation of the expected T^3 behavior of low-temperature phonons is observed. This is a clear sign for the development of an additional contribution from ferromagnetic spin fluctuations at low

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doi:10.1016/j.jmmm.2003.12.470

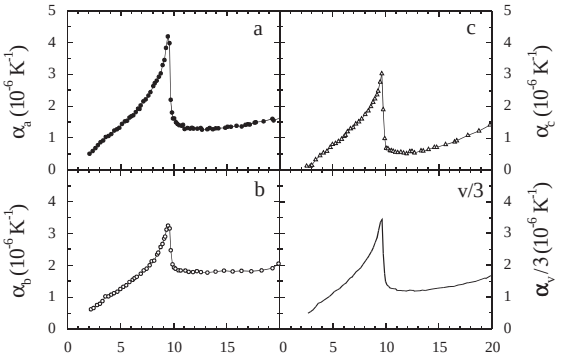

Fig. 1. The coefficients of linear thermal expansion α as a function of temperature T along the orthorhombic a-, b-, and c-axis, and the volume expansion divided by three ($\alpha_v/3$).

Table 1
Step anomalies in the coefficients of linear thermal expansion along the orthorhombic a-, b-, and c-axis and the corresponding pressure dependence of T_C

	a-axis	b-axis	c-axis	Volume
$\Delta\alpha$ (10^{-6} K^{-1})	3.4(1)	1.7(1)	2.7(1)	7.8(2)
dT_C/dp (K/kbar)	0.052(3)	0.026(2)	0.041(2)	0.119(6)

temperatures. It is interesting to note that this contribution mainly affects the anisotropy of the thermal expansion in the $b-c$ plane, which acts as the easy plane for the magnetization. At $T_C = 9.7$ K the ferromagnetic order sets in and a peak is observed for all three directions. The values of the steps are listed in Table 1.

In order to determine the uniaxial and hydrostatic pressure dependence of the ferromagnetic transition temperature we have applied the Ehrenfest relation: $dT_C/dp_i = V_m\Delta\alpha_i/\Delta(c/T)$, where the index i refers to the orthorhombic axis, $V_m = 3.36 \times 10^{-5}$ m^3/mol is the molar volume and $\Delta(c/T) = 0.22(1)$ J/mol K^2 is the anomaly in the specific heat divided by temperature [2]. These calculated values are listed in Table 1. The different pressure dependencies of T_C are all positive, which strongly suggests that the ferromagnetic order cannot be suppressed by moderate mechanical hydrostatic or uniaxial pressures, but instead a *negative* uniaxial pressure is needed to suppress T_C for all crystallographic directions or a hydrostatic pressure of $p_{cr} \approx -80$ kbar. It is important to note that this value should be regarded as an upper bound for the negative critical pressure as the pressure dependence of T_C is expected to show significant non-linear corrections to the initial pressure dependence at ambient pressure. A negative critical pressure of the order of $p_{cr} \approx -80$ kbar might be achieved by suitable chemical substitutions.

In the ferromagnetically ordered state three different contributions to the thermal expansion can be identified, namely contributions due to phonons, ferromagnetic spin waves, and ferromagnetic spin fluctuations. The phonon contribution shows a T^3 power-law behavior at low temperatures. The ferromagnetic spin-wave contribution is expected to obey a $T^{3/2}$ power-law behavior, while the ferromagnetic spin fluctuations lead to an enhanced linear electronic term at low temperatures. Both the phonon and spin-wave contributions to the volume expansion divided by temperature α_v/T vanish at low temperatures and, as a consequence, the extrapolated value of $\alpha_v/T = 5.8(2) \times 10^{-7} K^{-2}$ at $T = 0$ is solely due to the contribution of the ferromagnetic spin fluctuations.

In order to characterize the volume dependence of the electron correlations we have calculated the effective Grüneisen parameter by $\Gamma_{eff}(T) = V_m\alpha_v(T)/\kappa c(T)$ where $\kappa = -(1/V)(dV/dp)$ is the isothermal compressibility. As the compressibility of URhGe is unknown we will use an estimated value $\kappa = 0.8$ Mbar^{-1} because κ for other UTX compounds vary from $\kappa = 0.6$ to 1.0 Mbar^{-1} [3]. At high temperatures Γ_{eff} shows a small constant value of $\Gamma_{ph} = 2$, describing the volume dependence of the characteristic energy scale for phonons. Below 30 K Γ_{eff} rapidly increases and reaches a value of $\Gamma_{eff} \approx 14$ just above T_C. In the low temperature limit Γ_{eff} corresponds to the enhanced electronic Grüneisen parameter $\Gamma_{sf} = d\ln\gamma/d\ln V \approx 14$ of the ferromagnetic spin fluctuations.

Comparing the volume dependence of the energy scales for the ferromagnetic order (T_C) and the ferromagnetic spin fluctuations (T_{sf}), it turns out that $\Gamma_F = -d\ln T_C/d\ln V \approx 16$ is of the same order of magnitude and has the same sign as $\Gamma_{sf} = -d\ln T_{sf}/d\ln V \approx 14$. This situation is in strong contrast to pressure-induced antiferromagnetic superconductors like $CePd_2Si_2$ [4] where the antiferromagnetic order competes with the spin fluctuations with an opposite scaling behavior with volume. It can, therefore, be expected that the spin-mediated superconductivity of URhGe exists over a wide pressure range, as observed for the ferromagnetic superconductor $ZrZn_2$ [5].

We thank B. Fåk and P.C.M. Gubbens for stimulating discussions.

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