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The neutron response in $U_{0.975}Ce_{0.025}Ru_2Si_2$

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Abstract. We have carried out a neutron scattering study of the pseudo-ternary system $U_{0.975}Ce_{0.025}Ru_2Si_2$. Elastic scattering measurements have indicated antiferromagnetic order at wave vector $Q_1 = (1, 0, 0)$ ($\mu = 0.006 \pm 0.003 \mu_B$ at $T = 1.6$ K). Our inelastic scattering investigation has been focused on the low-temperature excitations showing a dispersive energy gap. Like for URu_2Si_2 , two spin-gap minima occur at wave vectors $Q_1 = (1, 0, 0)$ and $Q_2 = (0.6, 0, 0)$. The substitution of Ce for U in URu_2Si_2 shifts the characteristic spin gap energy from 1.8 meV to 2.4 meV at a wave vector Q_1 , and from 4.5 meV to 4.0 meV at a wave vector Q_2 .

1. Introduction

URu_2Si_2 is one of the few heavy-fermion systems displaying an unusual coexistence of superconductivity and magnetism. It exhibits a superconducting transition at $T_c = 1.2$ K and an antiferromagnetic transition at around $T_N = 17.5$ K [1–3]. The magnetic transition is associated with a weak antiferromagnetic ordering which displays an extremely small magnetic moment ($\mu \sim 0.04 \mu_B$ per U atom) [2] and with the development of a sharp gap [2] in the magnetic excitation spectrum. Polarized neutron analysis revealed that only the spin dipole orders at the transition [4]. The microscopic mechanism of the transition at T_N is still an open question since no model can explain how the appearance of such a small magnetic order parameter can lead to a large jump in the specific heat [1] and to an anomalous behaviour of many properties at T_N . The anomalies at T_N have been mostly interpreted in terms of an energy gap in the relevant excitation spectrum related to charge- or spin-density waves [5], to quadrupolar ordering [6] or to a combination of quadrupolar and dipolar ordering [7, 8]. The description of the magnetic properties of URu_2Si_2 is certainly at the borderline between an itinerant and localized picture of the 5f electrons. An itinerant description is supported by the strong sensitivity of the magnetic ordering to the substitution.

In our previous papers we have studied the effect of the substitution of Ce for U in URu_2Si_2 . We have discovered rapid suppression of the superconductivity and a three-step metamagnetic transition, observed in URu_2Si_2 [9]. Our measurements, taken on single crystals of $U_{1-x}Ce_xRu_2Si_2$ ($0 \leq x \leq 0.075$) showed that Ce substitution strongly affects anomalies in the resistivity, susceptibility and heat capacity which are related to the magnetic phase transition in URu_2Si_2 [9–12]. The anomalies shift to lower temperatures, but their shape remains similar to that for URu_2Si_2 , at least for concentrations $x \leq 0.05$. In this work, we investigate the neutron response in $U_{0.975}Ce_{0.025}Ru_2Si_2$ single crystal. The main

goal is to look for magnetic ordering, magnetic excitations, and correlations between the magnetic ordering and the anomalies which are usually connected with magnetic transition. Our results are compared with known results for the heavy-fermion compound URu₂Si₂.

2. Experimental techniques

The ternary system URu₂Si₂ crystallizes in the body-centred tetragonal ThCr₂Si₂-type structure with space group $I4/mmm$. The single crystal of U_{0.975}Ce_{0.025}Ru₂Si₂, 24 mm long and with a diameter of 5 mm, was grown by the Czochralski method in a 'tri-arc' furnace [13]. Our crystal was grown along the a -axis. Electron-probe microanalysis performed along and perpendicular to the growth direction revealed a homogeneous sample without any trace of parasitic phases. The low-temperature lattice parameters at 1.6 K were $a = 4.136 \text{ \AA}$ and $c = 9.595 \text{ \AA}$.

Table 1. The experimental set-up.

Experiment type at TAS7	λ (Å)	Collimation	Monochromator	Analyser	Filter(s) (cooled Be)
Elastic	4.068	28'-open-60'-65'	PG(002)	PG(002)	2
Inelastic	4.58	32'-open-60'-65'	PG(002)	PG(002)	1

Data were collected using the triple-axis spectrometer TAS7 situated at the cold neutron source of the DR3 reactor at Risø National Laboratory. A vertically focusing pyrolytic graphite PG(002) monochromator was used in the incident neutron beam to select neutrons of available energy. The scattered beam energy was analysed using a flat pyrolytic graphite PG(002) single crystal. The crystal was mounted, with the a - and c -axes in the horizontal scattering plane, in a standard ⁴He orange cryostat. We performed elastic and inelastic neutron scattering experiments (see table 1). The higher-order contamination for the elastic experiment was reduced by using two cooled beryllium filters (Be) of total thickness 24 cm. The collimations were chosen to be 28'-open-60'-65'. Scans were measured along the a^* - and c^* -directions in the low-temperature ordered state as well as in the high-temperature paramagnetic state. Our inelastic experiments have been focused to various wave vectors Q and temperatures T in the energy window from 0–6 meV.

3. Results and discussion

The magnitude of the ordered magnetic moment was measured by neutron elastic diffraction ($\Delta E = 0 \text{ meV}$). Scans performed on the forbidden Bragg reflections (1, 0, 0), (1, 0, 2) and (0, 0, 1) were consistent with the antiferromagnetic structure already found by Broholm *et al* [14] for URu₂Si₂. The magnetic form factor for U decreases with increasing $\sin(\theta)/\lambda$ in URu₂Si₂ [14]; that is why we concentrated on the forbidden Bragg peak at the (1, 0, 0) position. Figures 1 and 2 show the magnetic intensity at the forbidden Bragg point along the l -direction and h -direction, respectively. The experimental resolution was calculated by the Cooper–Nathan method, as well as being estimated by performing identical scans with an unfiltered beam containing a significant contribution of $\lambda/2$ from higher nuclear reflections. Scans are not resolution limited and the resolution widths are indicated by the horizontal line. The size of the magnetic moment was obtained by normalization of the magnetically scattered integrated neutron intensity to the integrated intensity associated

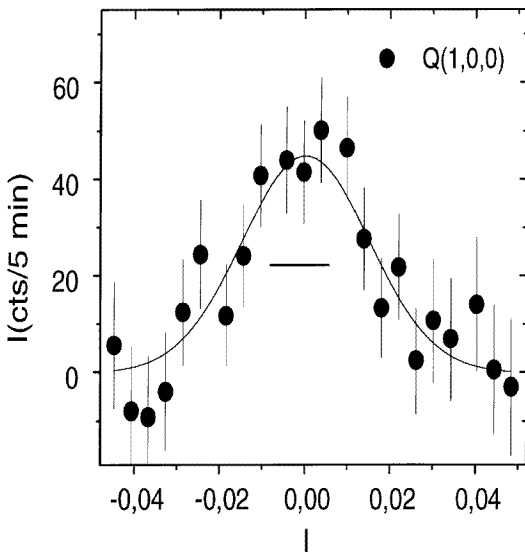


Figure 1. The scattered magnetic intensity along an l -direction at $Q = (1, 0, 0)$ and 1.6 K.

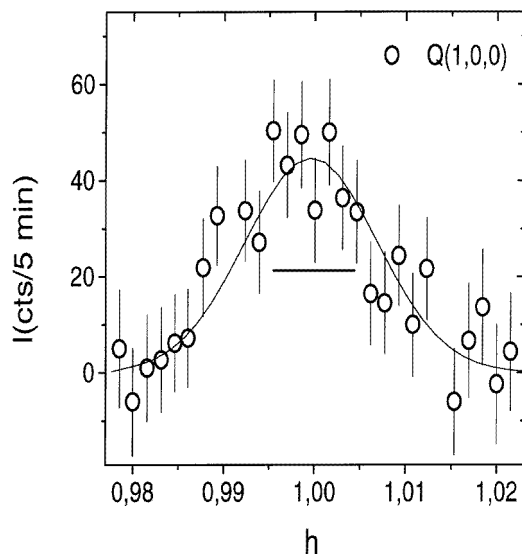


Figure 2. The scattered magnetic intensity along an h -direction at $Q = (1, 0, 0)$ and 1.6 K.

with the known cross section of the nuclear Bragg peak $(1, 0, 1)$. The magnetic moment of $U_{0.975}Ce_{0.025}Ru_2Si_2$ is polarized along the c -axis and determined to be $0.006 \pm 0.003 \mu_B$ per U atom at 1.6 K. The value of the magnetic moment is only 1/5 of the magnetic moment of URu_2Si_2 .

The inverse correlation lengths at low temperatures obtained from 1D deconvolution are $\kappa_a \sim 0.016 \text{ \AA}^{-1}$ and $\kappa_c \sim 0.02 \text{ \AA}^{-1}$. These give correlation lengths of $\xi_a \sim 65 \text{ \AA}$ and $\xi_c \sim 50 \text{ \AA}$, which correspond to 15 atomic distances in the basal plane and to 5 planes along the tetragonal c -axis. The correlation lengths are approximately ten times shorter than the correlation lengths in URu_2Si_2 .

Our inelastic scattering investigation has been focused on the low-temperature magnetic

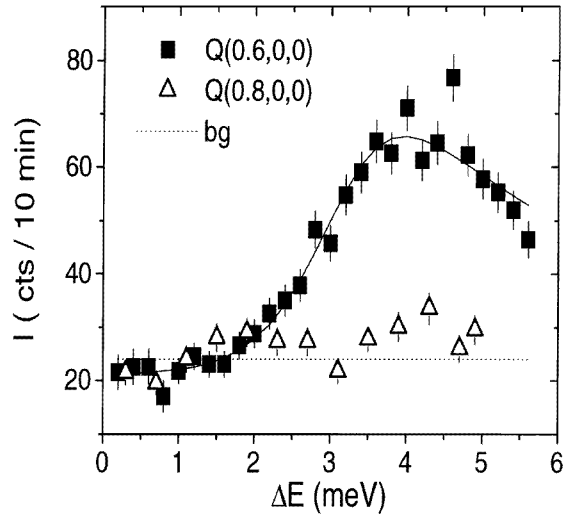


Figure 3. Constant- Q scans at $Q = (0.6, 0, 0)$ and $Q = (0.8, 0, 0)$ at 4.2 K. The solid and dotted curves are guides to the eye.

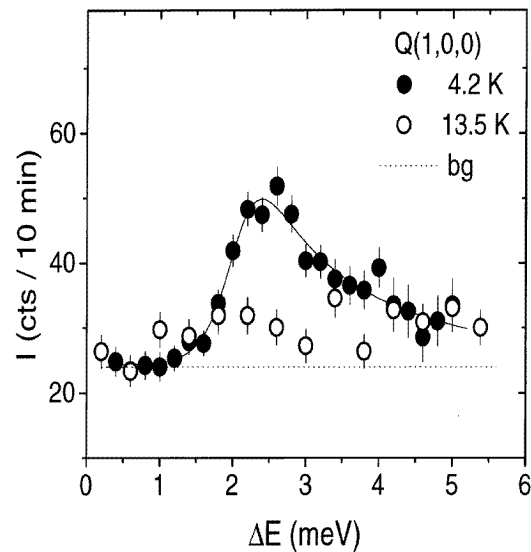


Figure 4. Constant- Q scans at $Q = (1, 0, 0)$ at 4.2 K and 13.5 K. The solid and dotted curves are guides to the eye.

excitations. Sharp dispersive excitations like in URu_2Si_2 were observed at low temperatures. The Q -dependence and the temperature dependence are displayed in figure 3 and figure 4. Like for URu_2Si_2 , two spin-gap minima occur at wave vectors $Q_1 = (1, 0, 0)$ and $Q_2 = (0.6, 0, 0)$. With the substitution of Ce for U in URu_2Si_2 , the energy gap at $Q_1 = (1, 0, 0)$ shifts from a value of 1.8 meV (reference [2]) to a value of 2.4 meV, and the energy gap at $Q_2 = (0.6, 0, 0)$ shifts from a value of 4.5 meV (reference [2]) to 4.0 meV. The difference between the two observed gap minima has decreased.

Figure 5 shows the temperature dependence of the magnetic intensity at the wave vector $Q_1 = (1, 0, 0)$ measured with an elastic set-up. The weak antiferromagnetic peaks which were shown in figure 1 and figure 2 appear at temperatures below that of the phase transition at $T_N \sim 10$ K. Like for URu_2Si_2 [2, 16], the intensity of the magnetic peak at

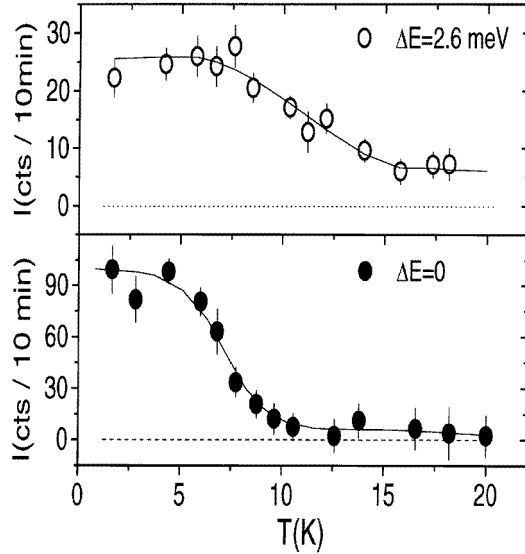


Figure 5. The temperature dependence of the magnetic peak (●) and spin gap (○) at $Q = (1, 0, 0)$ in elastic and inelastic spectrometer configurations (see table 1). The solid curves are guides to the eye.

Q_1 slowly increases with decreasing temperature and saturates at low temperatures. The Néel temperature determined from the neutron experiment differs from the temperature $T_A \sim 15.5$ K at which the kink in susceptibility [11] and the minimum in resistivity [15] appear. Similar differences between T_A and T_N have been found for annealed and as-grown single-crystal URu_2Si_2 [16]. The difference between T_N and T_A can be interpreted in terms of two successive phase transitions in the system [7, 8], the first at T_A being quadrupolar and the second at $T_N < T_A$ being dipolar. Due to the small moment, the macroscopic anomaly at T_N would be too small to observe; on the other hand the neutron scattering is not directly coupled to the quadrupolar moments; that is why no anomaly would be observed at T_A in neutron scattering experiments. Since the dipolar character of the ordering at T_N is established [4], the search for quadrupolar instability in URu_2Si_2 is of great interest. Investigation of elastic constants shows evidence of the quadrupolar moment below 70 K [17].

The temperature dependence of the inelastically scattered intensity ($\Delta E_m = 2.6$ meV) at $Q_1 = (1, 0, 0)$ (figure 5) shows neither a deviation nor an anomaly at the antiferromagnetic transition temperature $T_N \sim 10$ K. The inelastic background was measured with the analyser turned away from the reflection. With increasing temperature the spin energy gap at $Q_1 = (1, 0, 0)$ in the magnetic excitation spectrum softens and vanishes at about 15 K. The magnetic excitations are still present at temperatures well above the transition temperature $T_N \sim 10$ K determined from elastic neutron scattering. This seems to support an idea suggested by Fåk *et al* [16] that magnetic excitations are not directly related to the ordered moment. The anomalous behaviour observed in the resistivity and susceptibility measurements on $U_{0.975}Ce_{0.025}Ru_2Si_2$ appears at a temperature which corresponds to the temperature at which the spin energy gap at $Q_1 = (1, 0, 0)$ vanishes.

4. Conclusion

The main characteristic features of the heavy-fermion superconductor URu_2Si_2 , namely an antiferromagnetic order and a characteristic spin gap, persist in the compound

$U_{0.975}Ce_{0.025}Ru_2Si_2$. However, the size of the magnetic moment, the correlation length and the temperature of the transition to the ordered state are reduced upon substitution of Ce for U in URu_2Si_2 . The small magnetic moment polarized along the c -axis was determined to be $0.006 \pm 0.003 \mu_B$ per U atom at 1.6 K. The correlation lengths at low temperature are $\xi_a \sim 65 \text{ \AA}$ and $\xi_c \sim 50 \text{ \AA}$. Sharp dispersive excitations were observed at low temperatures, and two spin-gap minima occur at $Q_1 = (1, 0, 0)$ and $Q_2 = (0.6, 0, 0)$. The difference between the two observed gap minima, $\Delta_1 = 2.4 \text{ meV}$ and $\Delta_2 = 4.0 \text{ meV}$, has decreased. The weak elastic antiferromagnetic peak occurs below $T_N \sim 10 \text{ K}$. The transition temperature T_N does not correspond directly to the temperature T_A at which the anomalous behaviour of the susceptibility and resistivity has been observed. On the other hand, the temperature at which the spin energy gap at Q_1 appears correlates better with T_A .

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