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SEARCH FOR NOVEL ORDER IN URu_2Si_2 BY NEUTRON SCATTERING*

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We have made extensive reciprocal space maps in the heavy-fermion superconductor URu_2Si_2 using high-resolution time-of-flight single-crystal neutron diffraction to search for signs of a hidden order parameter related to the 17.5 K phase transition. Within the present sensitivity of the experiment (0.007 μ_B/U-ion for sharp peaks), no additional features such as incommensurate structures or short-range order have been found in the $(h0l)$ or (hhl) scattering planes. The only additional low-temperature scattering observed was the well-known tiny antiferromagnetic moment of 0.03 μ_B/U-ion.

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1. Introduction

The nature of the primary order parameter responsible for the entropy change at the $T_0 = 17.5$ K phase transition in the heavy-fermion superconductor URu_2Si_2 continues to be elusive. The inability of the small 0.03 μ_B/U-ion moment arising from long-range antiferromagnetic static dipolar order to account for the entropy change has led to many theoretical proposals for the nature of a primary hidden order parameter, ranging from

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unconventional spin-density waves through to incommensurate orbital antiferromagnetism arising from charge currents circulating between the uranium ions [1]. To test the validity of some of these suggestions, we have used time-of-flight single-crystal neutron diffraction to search for additional features that may be present below T_0.

2. Experimental

Time-of-flight single-crystal neutron diffraction is an efficient technique that enables data to be simultaneously collected over a wide Q-range, with each detector scanning a radial trajectory across the scattering plane [2]. For these experiments, we have used the PRISMA instrument at the UK ISIS neutron spallation source in its diffraction mode [3] with $30'$ collimation before the detectors, and the detector bank centred at a scattering angle $2\theta = 41°$ giving an accessible Q-range from 0.75 to 5 Å^{-1}. Reciprocal space maps are constructed by rotating the sample about the normal to the scattering plane in 8° steps corresponding to the angular width of the detector array.

The sample used is a large, annealed 0.328 g single crystal with dimensions $25 \times 5 \times 3$ mm^3 with the a direction along the longest axis. EPMA measurements have determined the sample to be single phase and of the required composition with no impurity inclusions. Resistance measurements confirm the presence of the 17.5 K phase transition, and the sample has a residual resistivity of 2.3 $\mu\Omega$m. The sample was oriented with either the $(h0l)$ or (hhl) planes in the scattering plane, and was cooled using a helium flow cryostat. The temperature dependence of the integrated intensity of the $\boldsymbol{Q} = (100)$ magnetic Bragg peak is well described by $I(T) = I(0)[1-(T/T_0)^{\alpha}]$ with $\alpha = 2.92$, in agreement with other high-quality samples [4].

3. Results

Reciprocal space maps are produced from the raw time-of-flight data by normalisation to the incident flux and the scattering from a standard vanadium sample, and then transforming into the reciprocal lattice coordinates of the scattering plane. Subtracting high-temperature (25 K) data from low-temperature data (4.5 K) for each scattering plane leaves only those features arising from the low-temperature ordered phase. In the $(h0l)$ plane, we observe magnetic peaks at (100) and (102), whilst in the (hhl) plane we observe peaks at (111) and (113), all arising from the well-known $\boldsymbol{k} = (001)$ periodic structure associated with the long-range antiferromagnetic static dipolar order of the uranium ions. The subtracted reciprocal space map for the (hhl) plane is shown in Fig 1. A cut along the $(hh0)$ direction through the (111) magnetic Bragg peak is shown in Fig. 2. The clear observation of the tiny ordered moment illustrates the excellent signal-to-noise ratio of the PRISMA instrument in diffraction mode.

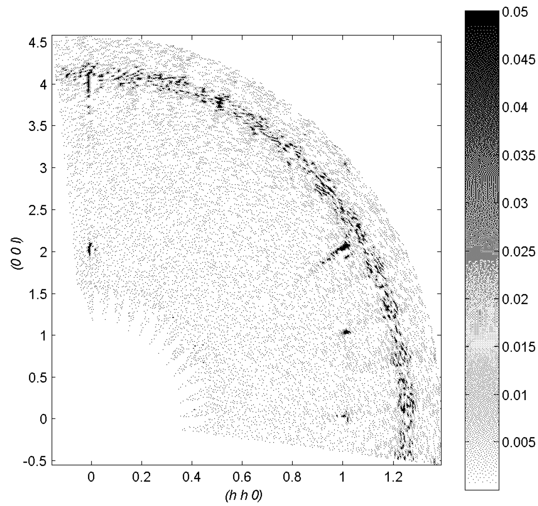

Fig. 1. Subtracted reciprocal space map for the (hhl) scattering plane. Darker points represent intensity above zero after the subtraction, with magnetic Bragg peaks located at (111) and (113). At nuclear Bragg positions, *e.g.* (110), the subtraction is influenced by thermal diffuse scattering and detector saturation, but averages to zero. The band of scattering towards the outer edge arises from the aluminium tails of the cryostat.

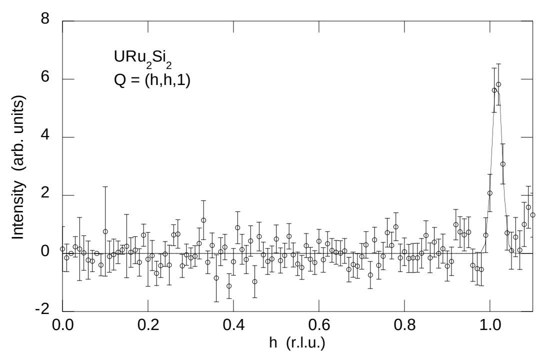

Fig. 2. Cut through the (hhl) subtracted map at $Q = (hh1)$ in a direction $q \parallel (hh0)$ with width $\Delta q_{\perp} = \pm 0.5c^{*}$. The line is a Gaussian fit.

From the measured reciprocal space maps and cuts similar to those shown in Fig. 2, we conclude that within the covered Q-range of the experiment and within the present accuracy, no additional incommensurate structures or any short-range order are observed. Additionally, an upper bound on the intensity of any incommensurate features in the scattering planes investigated can be set. From statistical analysis of the background and peak intensities, the detection limit of our experiment is around 1/20 of the intensity of the (100) magnetic Bragg peak, *i.e.* less than $\sim 0.007\ \mu_{\mathrm{B}}$/U-ion.

4. Discussion

In a recent paper inspired by the results of NMR and high-pressure neutron diffraction experiments, Chandra *et al.* [1] have suggested that the T_0 transition may be dominated by the onset of incommensurate orbital antiferromagnetism. The hidden-order phase arises from orbital currents circulating around square uranium plaquettes in the a-b plane, producing a small net moment perpendicular to each plaquet (*i.e.* along the c axis). The orbital currents give rise to small incommensurate Bragg peaks (with a rapidly decaying Q^{-4} form factor) principally located around $\boldsymbol{Q} = (qq1)$ with $q \approx 0.22$. Furthermore, the intensity of these incommensurate peaks in the first Brillouin zone is estimated to be $\sim 1/50$ of the antiferromagnetic dipolar Bragg peak at *e.g.* $\boldsymbol{Q} = (100)$.

In the present experiment we have not detected any of these signatures. In particular, Fig. 2 shows no signs of any peaks at $(hh1)$ with $h = 0.22$ or $h = 0.78$. While the experimental sensitivity is similar to the predicted intensity, the rapidly decreasing form factor could play a role at the relatively large Q values we have investigated. In fact, kinematic constraints mean that the (001) position could not be accessed in the present set-up. Also, if the orbital moment couples to the neutron spin in the same way as the dipolar moment, *i.e.* only the component of the spin perpendicular to the scattering wave vector $\boldsymbol{Q}$ is observed, then there would be a further decrease of the intensity, in particular for $\boldsymbol{Q}$'s close to the c axis. Further measurements at smaller Q-values using smaller scattering angles are envisaged.

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