Experimental report

Proposal: CRG-2775 Council: 4/2020

Title: Look for loop currents in URu2Si2

Research area:

This proposal is a new proposal

Main proposer: Philippe BOURGES

Experimental team:

Local contacts: Frederic BOURDAROT

Samples: URu2Si2

Instrument	Requested days	Allocated days	From	To
22	7	7	15/09/2020	22/09/2020

Abstract:

Proposal numbers: CRG 2755/CRG 2811

Title: Look for loop currents in URu2Si2

Experimental team: Ph. Bourges, D. Bounoua, Y. Sidis, F. Bourdarot, M-A. Méasson

Abstract: Since the 1985 discovery of the phase transition of an hidden order at T_{HO} ~17.5 K in the heavy-fermion metal URu2Si2, neither symmetry change in the crystal structure nor large magnetic moment that can account for the entropy change has been observed, which makes this hidden order enigmatic. We here propose polarized neutron diffraction to test various theoretical models which are considering loop currents phase (which does not necessarily break the translation of the lattice) as the origin of the hidden order. We ask for 7 days on IN22 with polarized neutron and polarization analysis.

Report: We performed the first experiment CRG 2755 from 15/09/2020 to 22/09/2020. We have studied two different scattering planes where (H,H,L) and (H,O,L) Bragg peaks were accessible. The crystal structure of URu2Si2 is a body-centered tetragonal structure, meaning that Bragg peaks occur when H+K+L=2n. An antiferromagnetic ordering of type A with propagation vector Q_0 =(001) has been discussed in the past [1,2,3], but observed with a too small ordered spin moment, of about 0.02 μ_B , incompatible with the observed large specific heat jump. To clarify, such usually assumed type A antiferromagnetic spin structure gives magnetic contributions at H+K+L=2n+1 at forbidden Bragg positions. The tiny magnetic contribution is reported at Q=(1,0,0) [1,2,3] and is often attributed to disorder [2].

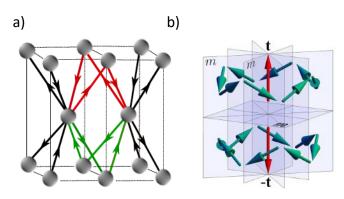


Figure 1: a) Chiral spin liquid phase (corresponding to the space greoup N° 128) proposed in [4] to account for the A2g Raman response (from [5]). That corresponds to loop currents circulating between the 4 U atoms produce magnetic moments respecting the symmetry of the lattice. b) Anti-toroidal vortex phase proposed in [6]. The green arrows represent the magnetic moments (the red arrows the toroidal moments). A ferro vortex is also proposed in ref [6].

To address this issue, two magnetic models with loop currents or toroidal moments have been proposed [4,6]. For the loop-currents model (chiral spin liquid) shown in figure 1a, one would expect a magnetic signal at Bragg positions (1,1,L) with L even. We actually calculate the magnetic structure factor of Fig. 1a. One can model such loop currents with two magnetic components. One is respecting the lattice symmetry (H+K+L=2n) with a moment

within the (a,b) plane, m_{ab} and the other one is not respecting the lattice symmetry (H+K+L=2n+1) with a moment along c, m_c , (actually with a symmetry similar to the type A antiferromagnetic ordering). The structure factor calculation gives for m_c an intensity at (1,0,0) but not at (0,0,1) as the moment is parallel to \mathbf{Q} (similarly to the type-A AF structure). Such a component is indeed present at (100) in the published data [1,2,3] but is very weak. For the in-plane component, m_{ab} , the signal should be non-zero if H=K=1 and zero for H=0 or K=0. We then look for positions such as (1,1,L) for all L.

In the model shown in Fig 2b involving with ferro-vortex and antiferro vortex moments, the calculations of the structure factors at different Bragg positions have been computed in reference [6] for these different electronic phases, either respecting or not the symmetry of the crystal structure. Typically, no intensity is expected for (H,O,L) or (H,H,L) for L=0.

Using polarized neutron, we have measured the temperature dependence of various Bragg peaks to see if a magnetic signal occurs at these positions following the method we developed and used in cuprates (ref [7]). Within error bars, no magnetic signal is sizeable at the positions (1,1,L) for L=0 and 2. The figure 2 shows the temperature dependence of the flipping ratio of the (1,1,0) Bragg peak. No change is observed across the hidden order temperature (17K), indicating no sizeable component at these positions. That shows that the component respecting the lattice symmetry expected in the chiral spin liquid model is absent or too weak to be measured. For completeness, we measured as well Bragg peaks in the (H,0,L) with L=1,3 plane where no effect was also observed. We would expect a signal at these positions for the ferro-vortex state discussed in ref. [6].

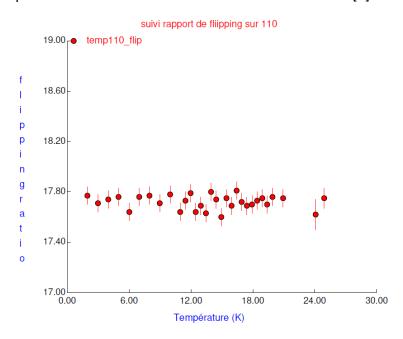


Figure 2: Temperature dependence of the flipping ratio of the (1,1,0) Bragg peak.

We also looked at positions in Q-space that are not respecting the lattice (such as H+K+L=2n+1). Figure 3 shows the temperature dependence at Q=(1,1,1) where a sizeable signal (but weak) is observed in the spin-flip channel. A similar signal is also observed at Q=(1,0,0) in agreement with previous neutron diffraction measurements [1,2,3]. An estimation of a magnetic moment of 0.02 μ_B has been deduced in agreement with previous

results on the same sample [3]. In terms of symmetry, the signal at (1,1,1) might be nevertheless compatible with the anti-toroidal vortex phase [6] of Fig. 1b but with a tiny magnetic moment. However, this model is not expecting a magnetic intensity at (1,0,0) with a similar amplitude as the (1,1,1) peak (even if it is weak as it is reported in [1,2,3] or observed here as well). For the chiral spin liquid model [4], the first estimates suggest that no magnetic signal can be expected at (1,1,1) for the component, m_c , not respecting the lattice symmetry. However, further quantitative calculations of the magnetic structure factors (including realistic form factors) for both models could be instructive to address precisely these questions.

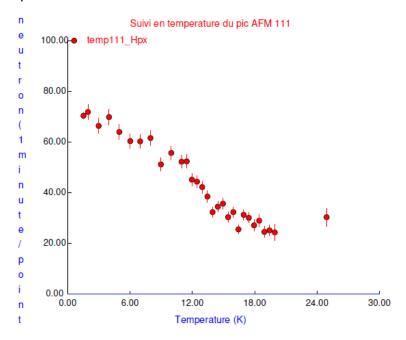


Figure 2: Temperature dependence of the spin flip intensity at the (1,1,1) position.

The second following experiment CRG 2811 to test positions such as (2,1,L) proposed in ref [6] to test the anti-ferro vortex model could not be done as the wave-vector where loop currents were expected was actually to large to be accessible on IN22 (just at the limit of the instrument). Further experiments using a diffractometer installed on a hot source such as D3 could be important to address this issue.

References:

- [1] C. Broholm, et al, Phys. Rev. Lett. 58, 1467 (1987). Phys. Rev. B. 43, 12809 (1990).
- [2] P.G. Nicklowitz et al, Phys. Rev. Lett, **104**, 106406 (2010).
- [3] F. Bourdarot, S. Raymond & L.-P. Regnault, Philosophical Magazine, 94, 3702-3722 (2014)
- [4] Carlene Silva de Farias, Marie-Aude Méasson, Alvaro Ferraz, Sébastien Burdin Phys. Rev. B 101, 205114 (2020).
- [5] J. Buhot, M.-A. Méasson, et al. Phys. Rev. Lett. 113, 266405 (2014).
- [6] Vladimir E. Dmitrienko and Viacheslav A. Chizhikov, Phys. Rev. B 98, 165118 (2018).
- [7] Ph. Bourges and Y. Sidis, C. R. Physique **12**, 461-479 (2011).