Experimental report

28/03/2017

Proposal: **5-41-877** **Council:** 4/2016

Title: Magnetic structure of UnRhIn3n+2 (n = 1, 2) compounds

Research area: Physics

This proposal is a continuation of 5-41-808

Main proposer: **Attila BARTHA**

Experimental team: Milan KLICPERA

Local contacts: Bachir OULADDIAF

Samples: U2RhIn8
URhIn5

Instrument	Requested days	Allocated days	From	To
D10	8	4	16/11/2016	21/11/2016

Abstract:

The recently discovered compound U2RhIn8 belongs to the family with chemical composition RnTmIn3n+2m, where R is a lanthanide or actinide and T a transition element. The materials grow with a tetragonal unit cell that can be viewed as n-layers of RIn3 stacked sequentially along the c-axis with m-layers of TIn2. This layered structure allows investigating the influence of stacking on the magnetic and superconducting properties within a certain family. With U2RhIn8 and URhIn5 we can study the effect of exchanging the localized 4f-electrons from the Ce-analogous with the more delocalized 5f-electrons. The cubic compound UIn3 orders antiferromagnetically at TN= 88K. U2RhIn8 and URhIn5 order at high temperatures, TN= 117K and 98K, respectively. The propagation vector of UIn3 equals (1/2,1/2,1/2). The same wave vector is found for URhIn5, while our recent experiment on D10 (proposal 5-41-808) revealed for the U2RhIn8 compound a propagation vector of (1/2,1/2,0). This proposal is a continuation of proposal 5-41-808. We like to determine the magnetic moment of the U in U2RhIn8 as well as the temperature evolution of AFM state in U2RhIn8 and URhIn5 to study the influence of the RhIn2-layer.

ILL Report: Magnetic structure of U_nRhIn_{3n+2} (n = 1, 2) compounds

Scientific background: Remarkably, many intriguing phenomena discovered in condensed matter physics during the past decades were found in intermetallic lanthanide (4*f*) or actinide (5*f*) compounds exhibiting tetragonal crystal structure. Examples are heavy fermion superconductivity in $CeCu_2Si_2$ [1], hidden order in URu_2Si_2 and quantum criticality beyond the Landau-Ginzburg-Wilson paradigm in for instance $YbRh_2Si_2$, $CeRhIn_5$ and Ce_2RhIn_8 [2-4]. The lattice-type naturally inhabits a certain degree of frustration of the 4*f* or 5*f* - moments and the electronic dimensionality is often ascribed being less than 3-dimensional akin to layered structures. Both, frustration and dimensionality, play a crucial role in the description of earlier mentioned phenomena. In that sense, it is interesting to explore the effect of *f*- *s,p,d* electron hybridization on similar type of compounds which can be achieved by replacing the 4*f*-electron element (Ce, Yb) by a 5*f* element. (U). In the series, $CeIn_3$, $CeRhIn_5$ and Ce_2RhIn_8 the Ce 4*f*-electron is strongly localized. The compounds order antiferromagnetically with T_N of 10.2K, 3.8K and 2.8K respectively, and become superconducting under hydrostatic pressure. UIn_3 has been exhaustively investigated. The compound orders antiferromagnetically (type-II AFM) at T_N= 88K with propagation vector (1/2,1/2,1/2) and moments aligned along <110>-axis. Interestingly, NQR experiments reveal that the U-moments have localized character. Until recently solely U_nTGa_{3n+2} (*T*: transition element) compounds exist including only two so-called 2-1-8 compounds, U_2FeGa_8 and U_2RhGa_8 both being paramagnetic. $URhIn_5$ was the first with In as *p*-element to be synthesized. In addition Bartha *et al.* [5] succeeded in preparing U_2RhIn_8, another 5*f* - analogue in the Ce-series. Both compounds order AFM with Néel temperatures T_N = 117K (98K) for U_2RhIn_8 ($URhIn_5$). A recent NMR study on $URhIn_5$ suggested that AFM here is driven by itinerant 5*f* electrons and has propagation vector $\boldsymbol{k}$ = (1/2.1/2.0) [6]. This proposed wave vector was confirmed by a recent neutron experiment on PANDA-FRM II [7]. The size of the magnetic moment yields μ = 1.65 μ_B/U. However, little is known about the magnetic properties of U_2RhIn_8.
The current experiment focuses on the magnetic structure determination of U_2RhIn_8, to find out the magnetic moment on the Uranium and to follow the evolution of the AFM state with temperature in order to clarify the character of the magnetic ordering (2D or 3D). The experiment was a follow up experiment of ILL proposal 5-41-908 where already we were able to determine the propagation vector being (1/2,1/2,0).

Experimental: Single crystals of U_2RhIn_8 have been grown from In-flux. The samples are plate-like with *c*-axis perpendicular to the plate. Typical dimensions are 1×1×0.5 mm^3 (mass ~ 1 mg). Prior to the neutron experiment, the samples were subjected to thermodynamic and transport experiments [5]. One sample was selected for further study on which we performed a Laue neutron single crystal diffraction experiment utilizing CYCLOPS instrument at ILL, Grenoble first. Scans were taken at T = 2 K (AFM state) and 130 K (paramagnetic state). The presence of additional magnetic Bragg reflections in the 2 K-scan indicated that the ground state of U_2RhIn_8 is AFM. The respective Laue patterns were further analyzed using Esmeralda software. By this we were able to confirm that the propagation vector in U_2RhIn_8 indeed is $\boldsymbol{k}$ = (1/2,1/2.0). Subsequent neutron experiment was conducted on D10 in order to resolve the magnetic structure. The neutron wavelength equals λ = 2.36 Å and measurements were done down to T = 1.8 K.

Results: We measured the intensity of two magnetic reflections (-0.5,1.5,-1) and (0.5,2.5,1) at various temperatures between 1.8 K and 135 K. Figure 1 exemplarily presents the temperature dependence of the measured intensity (-0.5,1.5,-1). Clearly, intensity which relates to the ordered moment on the U-ions sets in below T_N (117 K) and increases upon lowering temperature. In order to follow the evolution of the ordered moment of the U-ion the integrated intensity has been calculated and plotted versus temperature in Fig. 2.

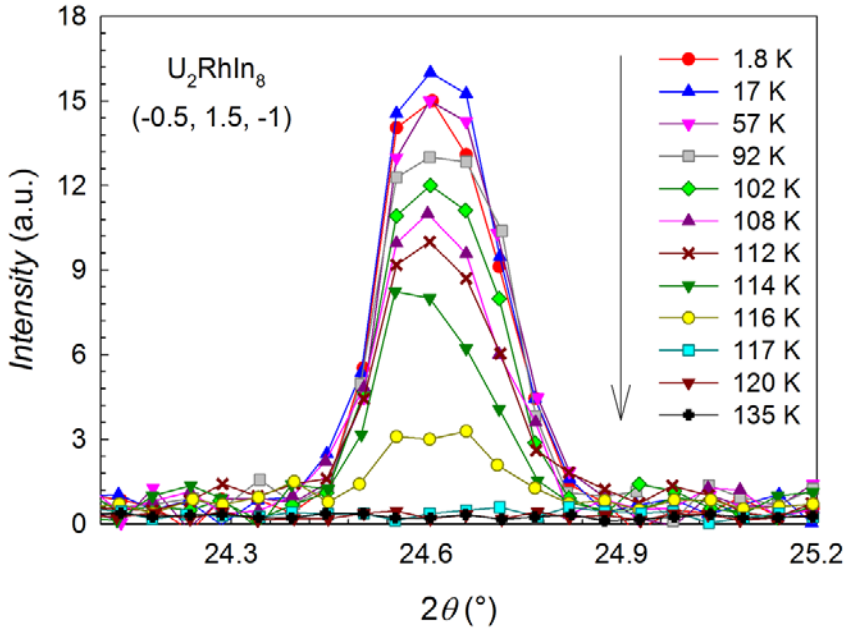

Fig.1: *Temperature dependence of the intensity on the magnetic reflection (-0.5,1.5,-1). Error bars are within symbol size*

Below approximately 80 K, the intensity shows no further increase. Figure 2 includes the nuclear reflection at reciprocal space position (-2,0,0). The integrated intensity of the nuclear reflection is almost constant over the entire measured temperature range demonstrating the absence of a $\boldsymbol{k_0}$ = 0 ferromagnetic component to the magnetic structure in accordance with results on CYCLOPS. Near the transition temperature, the integrated intensity of the magnetic diffraction peaks may be fit to a simple power law, $I \propto (T_N - T)^{2\beta}$ with T_N the Néel temperature and β is the order parameter critical exponent. The fits were performed in a small interval of 108 K $< T <$ 120 K yielding T_N = 117 K in agreement with macroscopic measurements [5]. For the critical exponent β we obtained for both datasets a value of β = 0.325. The value points to a 3D-Ising ordering of the U-moments [8]. In the following we employed MAXMAGN software using the refined structure parameters of U_2RhIn_8 and propagation vector ***k*** as input. We obtained 10 1D irreducible representations associated with the ***k***, but only 6 of them are part of the global reducible magnetic representation of the 2*g* Wyckoff site occupied by U-ions. All 6 subgroups assume unit-cell doubling in *a*-direction. The magnetic space groups P_C4/mbm (127) and P_C4/nbm (125) have the magnetic moment aligned *μ* along [001]. The C_amma (67) and C_ammm (65) require alignment along [100] and the remaining two subgroups P_Bmna (53) and P_Bmma (51) have μ parallel to [110]. To further identify the magnetic structure we measured the intensities of magnetic satellites described by ***k*** at T = 1.8K. From the 174 magnetic reflections only 34 were independent. For refinement we used FullProf and we revealed best agreement with our data considering a commensurate magnetic structure with moments arranged along [001], i.e., tetragonal *c*-axis. The agreement factors yield RF = 13.4% and RF^2 = 12.1%. The magnetic structure is depicted in Fig. 3. It corresponds to the magnetic space group P_C4/nbm – maximal magnetic subgroup of the paramagnetic space group P4/*mmm*.

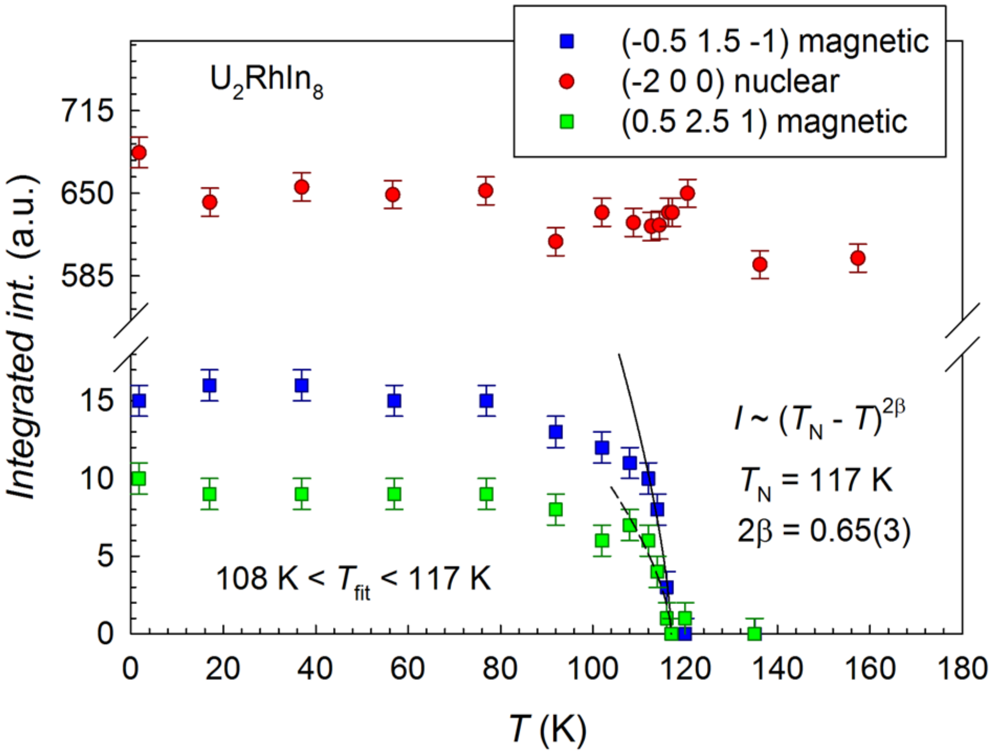

Fig.2: *Temperature dependence of the integrated intensity of the magnetic reflections (-0.5,1.5,-1) and (0.5,2.5,1). For comparison the integrated intensity of the nuclear reflection on (-2,0,0) is added. The solid (dashed) black line is power law fits in order to determine β from (-0.5,1.5,-1) and (0.5,2.5,1, respectively.*

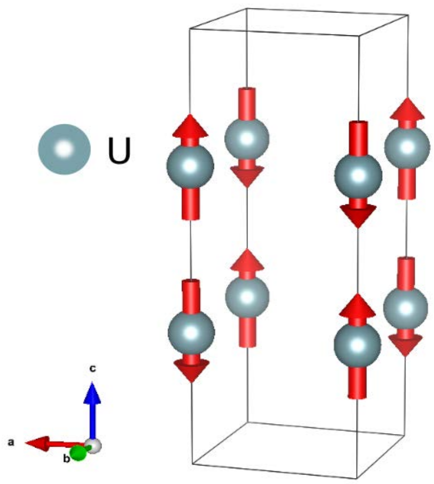

Fig.3: *Magnetic strucuture of U_2RhIn_8. For clarity only the U-ions are displayed.*

References:
[1] O.Stockert *et al*., Nature Physics **7**, 119-124 (2011)
[2] J. Custers *et al*., Phys. Rev. Lett. **104**, 186402 (2010)
[3] H. Hegger *et al.*, Phys. Rev. Lett. **84,** 4986 (2000)
[4] M. Nicklas *et al.*, Phys. Rev. B **67,** 020506(R) (2003)
[5] A. Bartha *et al.*, J. Magn. Magn. Mater. **381,** 310 (2015)
[6] H. Sakai *et al*., Phys. Rev. B **88**, 045123 (2013)
[7] P. Čermák et al., Magnetic structure of recently discovered $URhIn_5$, experimental report for proposal p8818
[8] G. S. Brush, Rev. Mod. Phys **39**, 883 (1967