Experimental report

22/02/2017

Proposal: **5-41-859**

Council: 4/2016

Title: Identification of unusual magneticstructure of UAu2Si2

Research area: Physics

This proposal is a new proposal

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Samples: UAu2Si2

Instrument	Requested days	Allocated days	From	To
D10	7	12	28/09/2016	06/10/2016
			04/11/2016	08/11/2016

Abstract:

Tetragonal UT2Si2 (T: transition metal) compounds are intensively studied for their interesting properties such as the hidden order and local-moment antiferromagnetic orders with commensurate and incommensurate propagation vectors, and their coexistence with itinerant heavy-fermion states and/or superconductivity. UAu2Si2 has shown puzzling low-temperature properties inconsistent among reported studies. Recently, we succeeded in growing single-crystalline samples of UAu2Si2 for the first time, and obtained a reliable set of experimental data of its thermal, magnetic, and transport properties.
The neutron Laue diffraction on a UAu2Si2 single crystal revealed the propagation vectors of magnetic structure, k = (±⅔, 0, 0) and (0, ±⅔, 0). This result is well in agreement with our 29Si-NMR experiment on powder sample, which suggests the conic magnetic structure with ferromagnetic component along the c-axis and antiferromagnetic component confined within the tetragonal basal plane. In order to confirm this scenario and identify the magnetic structure of UAu2Si2, we intend to perform the single crystal neutron diffraction experiment employing D10 four-circle diffractometer.

1 Scientific background

UAu_2Si_2, which crystallizes in a $ThCr_2Si_2$ type tetragonal structure (space group: I4/mmm, D_{4h}^{17}), shows a second-order phase transition at 19 K. The low-temperature-ordered state had been believed to be a simple ferromagnetic one for about 30 years, receiving much less attention than those of other UT_2Si_2 relatives [1–5]. Recently we succeeded in growing single crystalline samples, and revealed that nature of the phase transition is antiferromagnetic through macroscopic measurements of thermal, magnetic, and transport properties [6]. In addition, ^{29}Si-NMR spectra, which we measured recently, are well explained by a magnetic structure where uranium magnetic moments are aligned parallel to the [001] axis with a propagation vector of $\boldsymbol{k}$ = (2/3, 0, 0). No other uranium 1-2-2 compound orders with this propagation vector in zero-magnetic field, making UAu_2Si_2 a unique system within the UT_2Si_2 family. Further study of magnetic structure and physical properties of this compound will eventually lead to understanding of 5f-electronic properties in UT_2Si_2 systems.

2 Aim of proposal

The aim of the present experiment is to establish a picture of the magnetic structure, which is realised in UAu_2Si_2 below T_N = 19 K. We intend to perform a neutron diffraction experiment employing D10 single crystal diffractometer for a precise magnetic structural refinement.

3 Experimental procedures

UAu_2Si_2 single crystal was grown by the floating zone melting method in an optical furnace at Charles University, Czech Republic. A rectangular piece with dimensions of $\sim$ 1 mm $\times$ 1 mm $\times$ 2 mm was cut out of the grown rod-shaped crystal. The sample was attached to an Al holder with GE varnish.

First, we performed a neutron Laue diffraction experiment using CYCLOPS in order to determine magnetic propagation vectors of UAu_2Si_2. The Laue images acquired at temperatures from 2 K up to 70 K clearly indicate that the magnetic reflections appearing below T_N are described by the propagation vector of $\boldsymbol{k}$= (2/3, 0, 0) and (0, 2/3, 0). Subsequently, the single crystal was investigated using a four-circle diffractometer D10 employing an area detector. The sample [100]-axis was parallel to the omega-axis of the spectrometer. The measurements of nuclear and magnetic reflections described by $\boldsymbol{k}$ were performed using incident neutron wavelength of λ = 2.36 Å. Finally, the measurements of nuclear reflections at 30 K were carried out with λ = 1.26 Å to reach high-Q reflections. The sample was cooled down to 2 K by a ^{4}He gas-flow refrigerator. The half-lambda component was removed by a graphite filter; its intensity was reduced to 10^{-4} of the main component.

At 30 K, i.e. above T_N, about 100 independent nuclear reflections were measured for the crystal structure refinement. In the magnetically ordered state, at 2 K, $\sim$ 300 independent magnetic reflections were measured for the magnetic structure refinement. The least-square refinements were carried out using the FullProf package.

4 Experimental results

The I4/mmm crystal structure was confirmed by refining the nuclear reflections data collected above T_N (at 30 K). The reliability factors are R_{F2} = 7.8 %, R_{F2w} = 6.8 %, and R_F = 4.9 %. The only free structural parameter, z coordinate of Si atoms, was refined to be z_{Si} = 0.3893(3). At the base temperature, 2 K, we observed magnetic superlattice reflections with indices of $\boldsymbol{\tau}\pm$(2/3, 0, 0) or $\boldsymbol{\tau}\pm$(0, 2/3, 0) ($\boldsymbol{\tau}$: reciprocal lattice vector). Figure 1 depicts the temperature dependence of one typical reflection (-2, -0.67, 0). The intensity on magnetic reflection below 20 K increases with decreasing temperature (see Fig. 1) and starts to saturate below 5 K (see Fig. 2).

Our previous magnetization data point out two magnetic anomalies in UAu_2Si_2. The first one below 50 K is ferromagnetic (FM), while the second one below 20 K can be ascribed to antiferromagnetic (AFM) order. We note that the former anomaly is connected with only a small magnetization change (i.e. no anomaly in the specific heat), which is related to a small ferromagnetic (FM) component. The neutron diffraction experiment confirms the AFM ordering together with the presence of weak FM component below 20 K. However, the value of magnetic moment of FM component cannot be determined precisely. The agreement between measured data and our models: (i) nuclear intensities on nuclear reflections only and (ii) nuclear plus FM contributions, does not allow to distinguish between the two cases as the FM contribution is much smaller than nuclear one. We decided to fix the magnetic moment of the FM component to 0.3 μ_B /U at 2 K as the calculated magnetic intensities are comparable with the intensity change on the nuclear reflections with decreasing temperature below 20 K (see Fig. 2).

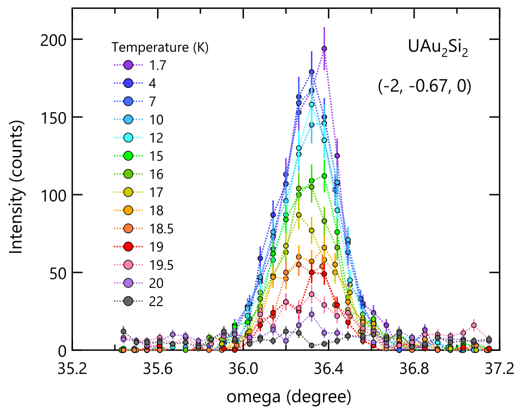
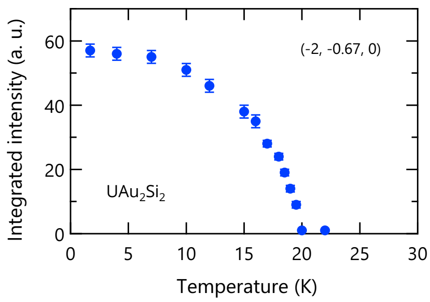


Figure 1: Temperature development of the intensity on a magnetic Bragg reflection (-2, -0.67, 0).



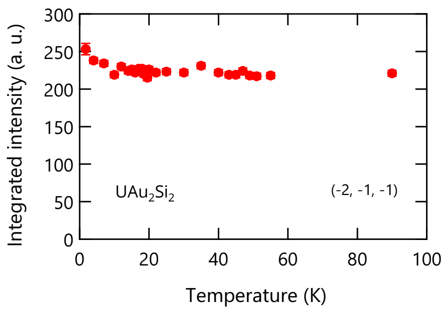


Figure 2: Temperature dependence of integrated intensity on a magnetic Bragg reflection (-2, -0.67, 0) (left) and nuclear (and FM) Bragg reflection (-2, -1, -1).

The AFM component was determined unambiguously and is shown in left panel of Fig. 3: AFM order with an up-up-down configuration of magnetic moments of U ions. The magnetic moments are oriented along the [001], and one (-) moment of the (+, +, -) configuration has a magnitude twice of that of two (+) moments (the result of an amplitude modulated magnetic structure). The final magnetic structure is a sum of the FM and AFM components, as displayed in the right panel of Fig. 3. The magnetic moments at each U sites have the same magnitudes of approximately 1.0 μ_B, and two thirds and one thirds of them point parallel and antiparallel to [001] axis, respectively.

The magnetic structure at 2 K seems to be solved neglecting possibly small tilt of magnetic moments from the c-axis. However, we challenge an unexpected temperature evolution of intensity on at least two nuclear reflections: 200 and 220. The intensity strongly increases below 20 K and at 2 K reaches 40-times the value calculated for the FM component (see Fig. 4). Such a rapid increase cannot be ascribed to magnetic moments on U atoms, only. It might imply a subtle lattice distortion below T_N. It should be further investigated by means of XRD, electron diffraction, thermal expansion measurements, ultrasonic elastic constants measurements, and so on.

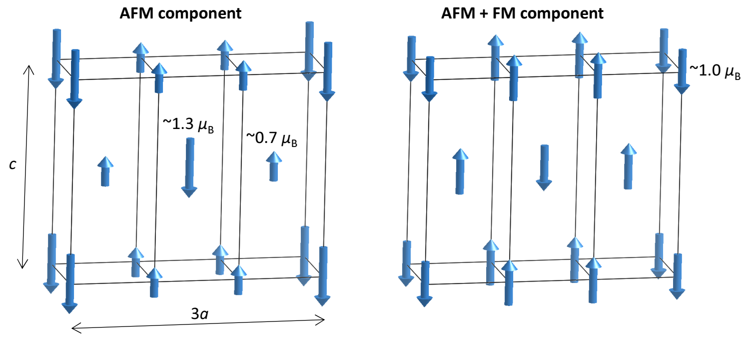


Figure 3: The deduced magnetic structure analyzing only the AFM component (left panel) and the structure which is obtained by adding an FM component of 0.3 Bohr magneton per U ions to the AFM component (right panel).

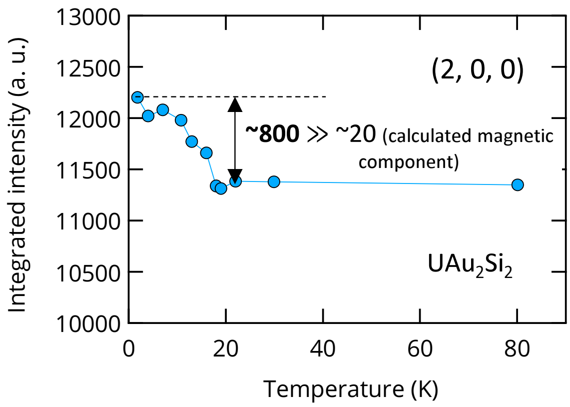


Figure 4: Temperature dependence of integrated intensity on the nuclear Bragg reflection (2, 0, 0).

5 Summary

We performed magnetic structure analyses on single-crystalline UAu_2Si_2, for the first time. The magnetic superlattice reflections with a propagation vectors $\boldsymbol{k}$ = (2/3, 0, 0) and (0, 2/3, 0) were observed below the magnetic transition temperature, T_N = 19 K. The magnetic structure was refined to be a (+, +, -) structure along the [100] or [010] axes, with magnetic moments of $\sim 1\,\mu_B$ parallel to the [001] axis. The large enhancements of 200 and 220 reflections which cannot be accounted for by the magnetic components have been unsolved, implying that careful investigation of crystal structure deformations is needed in future.

References

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