

Experimental report

16/12/2021

Proposal: 5-31-2667

Council: 4/2019

Title: Magnetic structure determination of ternary intermetallic compounds in the U-Mn-Ge system.

Research area: Materials

This proposal is a new proposal

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Samples: UMnGe
U8Mn3Ge7
UMn1-xGe2
U34Mn4-xGe33
U3Mn4Ge4
U2Mn3Ge
UMn6Ge6
15U-58Mn-27Ge

Instrument	Requested days	Allocated days	From	To
D1B	4	3	08/10/2019	11/10/2019

Abstract:

The association of U with Ge in intermetallics gives birth to some of the most intriguing solid state phenomena: coexistence of superconductivity with ferromagnetic order, magnetic ordering of U-sublattice despite very short U-U distances. Consequently, ternary U-Mn-Ge compounds, where Mn may also carry a magnetic moment and undergo magnetic ordering, must exhibit fascinating physical properties and magnetic structures. Up to now, this system was poorly investigated with only 3 compounds reported, among which the magnetic structure was resolved for UMn2Ge2 only. Our study highlights the existence of 6 further ternary compounds in this system. Magnetic ordering of the Mn- and/or U-based sublattices in these germanides is either revealed by preliminary measurements or expected. In order to understand how these magnetic sublattices cooperate in these compounds, accurate magnetic structure determinations by neutron powder diffraction are necessary. Magnetization, specific heat and electrical resistivity measurements are in progress and will complete this study.

Experimental report on the proposal 5-31-2667: **Magnetic structure determination of ternary intermetallic compounds in the U-Mn-Ge system.**

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1. Introduction

The ternary intermetallic compounds $R_xT_yX_z$ (R = rare earth, Y, La, U; T = d -element; X = p -element) have been relatively well investigated for their structural, magnetic and transport properties. In intermetallics, the magnetic behavior of uranium results from a subtle interplay between the localized and itinerant character of its $5f$ electrons driven by the hybridization of the $5f$ orbitals with neighboring atoms [1]. Moreover, associating this actinide with germanium in binary or ternary phases gives birth to some of the most intriguing solid-state phenomena, *e.g.* coexistence of superconductivity and FM order in UGe_2 [2], $URhGe$ [3] and $UCoGe$ [4] or the FM ordering of U-sublattice in U_2Fe_3Ge despite very short U-U distances [5].

Ternary U-Mn-Ge system was, up to now, poorly investigated with only 3 ternary phases reported: UMn_2Ge_2 ($ThCr_2Si_2$ -type) [6], $UMnGe$ ($TiNiSi$ -type) and U_2Mn_3Ge (Mg_2Cu_3Si -type) [7]. Moreover, despite the expected magnetic properties of U-Mn-Ge compounds, only those of UMn_2Ge_2 were really studied. This compound is characterized by a FM ordering of both Mn and U sublattices around 380 K and 150 K, respectively [8-12], magneto-optic properties [13,14] and evidences a reversible structural phase transition at a pressure of 16.1 GPa [15,16]. Magnetization measurements suggest that $UMnGe$ orders antiferromagnetically below 240 K and exhibits another transition near 65 K [7]. However, its magnetic structure as well as the magnetic properties of U_2Mn_3Ge have never been reported. For these reasons, we started to investigate the U-Mn-Ge system (**Fig. 1**), through crystal structure and magnetic properties. In order to understand how the Mn- and U-based magnetic sublattices cooperate in these ternary germanides, accurate magnetic structure determination by neutron powder diffraction is necessary to complement laboratory investigations.

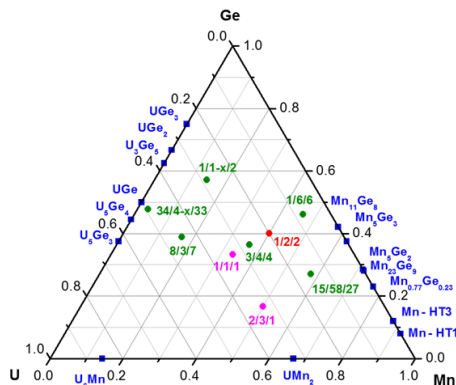


Fig. 1: Gibbs triangle of the U-Mn-Ge ternary system showing binary phases in blue, the well-known UMn_2Ge_2 in red, the reported but poorly characterized phases in magenta and the freshly discovered compounds in green.

2. Neutron diffraction results

Long duration neutron diffraction patterns, as well as isotherms, were recorded in the paramagnetic state and in the magnetically ordered state for $\text{UMn}_{0.33}\text{Ge}_2$, UMn_6Ge_6 , $\text{U}_{34}\text{Mn}_3\text{Ge}_{33}$, $\text{U}_2\text{Mn}_3\text{Ge}$, UMnGe , $\text{U}_8\text{Mn}_3\text{Ge}_7$, and $\text{U}_3\text{Mn}_4\text{Ge}_4$ compounds. These neutron data allowed us to confirm their crystal structures determined by powder and single-crystal X-ray diffraction, and their magnetic behavior studied by macroscopic magnetic measurements. For example, these data confirm the structural phase transition of UMnGe at low temperature (Fig. 2) suggested by magnetic susceptibility measurements (Fig. 3).

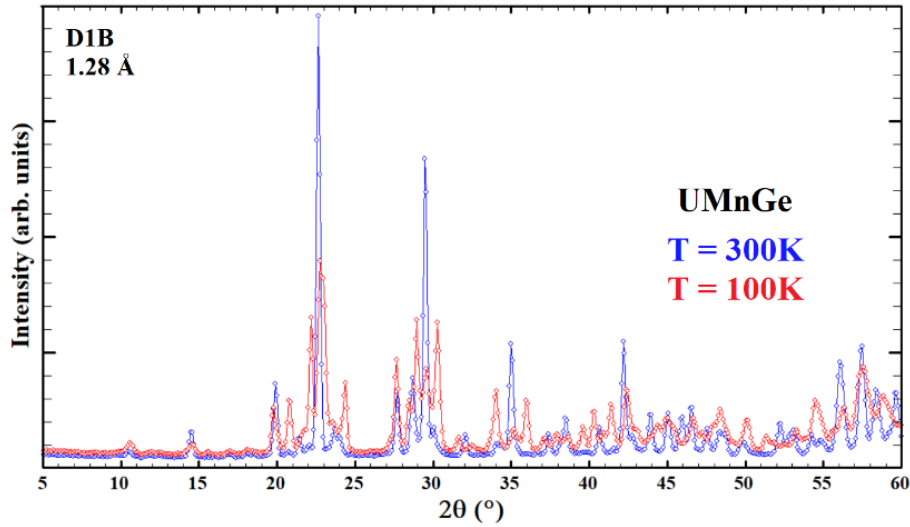


Fig. 2: Comparison of the neutron diffraction patterns of UMnGe recorded at 300 K and 100 K.

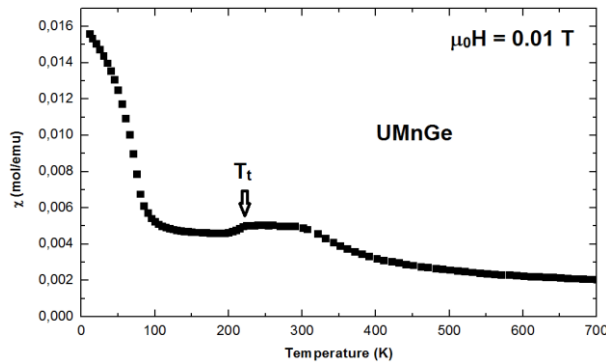


Fig. 3: Magnetic susceptibility curve of UMnGe in the 4 - 700 K temperature range.

The complexity of the ternary U-Mn-Ge system (Fig. 1) leads to the presence of magnetic secondary phases in the samples, as exemplified in Fig. 4 with the presence of UMn_2Ge_2 and UMn_4Ge_2 secondary phases in the $\text{U}_3\text{Mn}_4\text{Ge}_4$ sample. Consequently, magnetic structure determination of the phases of interest requires a knowledge of the magnetic structure of every ternary U-Mn-Ge compounds. This implies to consider almost simultaneously the neutron data of each sample before finalizing the refinement of the magnetic structures. For this reason, the analysis of such data is a hard work, and the interpretations are still in progress. Nevertheless, these neutron data will provide us very important information to better understand the structure

– magnetic properties relationships encountered in these ternary compounds and will be the subject of several upcoming publications.

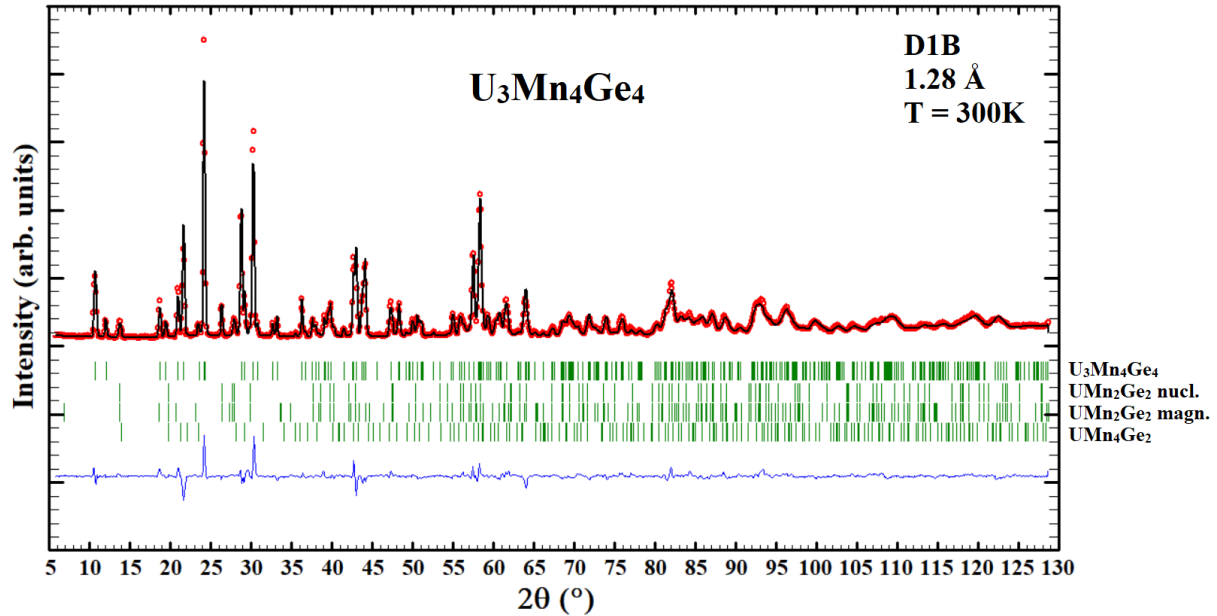


Fig. 4: Rietveld refinement of $\text{U}_3\text{Mn}_4\text{Ge}_4$ neutron diffraction pattern recorded at 300 K.

3. References

- [1] Zwicknagl *et al.*, J. Phys.:Cond. Matter, 15 (2003) S1911.
- [2] Saxena *et al.*, Nature, 406 (2000) 587-592.
- [3] Aoki *et al.*, Nature, 413 (2001) 613-616.
- [4] Huy *et al.*, Phys. Rev. Lett., 99 (2007) 067006.
- [5] Henriques *et al.*, Solid State Commun., 148 (2008) 159-162.
- [6] Marazza *et al.*, J. Less-Common Met. 53 (1977) 193.
- [7] Hoffmann *et al.*, Solid State Sci. 21 (2013) 73.
- [8] Szytula *et al.*, J. Phys. Chem. Solids 49 (1988) 1113.
- [9] Dirkmaat *et al.*, J. Magn. Magn. Mater. 84 (1990) 143.
- [10] Slaski *et al.*, J. Alloys Compd. 178 (1992) 249.
- [11] Parker *et al.*, Phys. Rev. B 91 (2015) 174401.
- [12] Tan *et al.*, Phys. Rev. Mater. 2 (2018) 074402.
- [13] van Engelen *et al.*, IEEE Trans. Magn. 24 (1988) 1728.
- [14] Kirby *et al.*, J. Appl. Phys. 69 (1991) 4574.
- [15] Siruguri *et al.*, Physica B 344 (2004) 255.
- [16] Matar *et al.*, J. Alloys Compd. 436 (2007) 34.