Experimental report

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Title: The nuclear and magnetic structureof the superconducting phase of CrAs at high pressure and low temperature

Research area: Materials

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Samples: CrAs

Instrument	Requested days	Allocated days	From	To
D10	10	0		
D23	10	0		
D9	0	7	13/09/2021	20/09/2021
ORIENTEXPRESS	0	7		

Abstract:

Chromium arsenide (CrAs) is an unconventional superconductor, exhibiting superconductivity induced by pressure. The aim of this investigation is the precise determination of the nuclear and magnetic structure at high pressure and low temperature within the superconducting phase region in vicinity of the helical antiferromagnetic phase. The knowledge of this structure will provide a better understanding of the ground state of this material.

The nuclear and magnetic structure of the superconducting phase of CrAs at high pressure and low temperature

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Chromium arsenide exhibits superconductivity at high pressures and low temperatures in the vicinity of a helically ordered antiferromagnetic phase. The precise knowledge of the magnetic structure near and in the superconducting phase region is needed in order to understand the interplay of magnetism and superconductivity in the multiparameter field. We performed single-crystal diffractometry experiments on D9 with λ = 0.84 Å at two pressure points at 2 K.

Chromium arsenide (CrAs) exhibits pressure-induced superconductivity in the vicinity of a helically ordered antiferromagnetic phase, forming a dome-like shaped region with maximum T_c = 2.2 K at about 1 GPa [1]. In the literature, the magnetic structure is described as double helix propagating along the *c*-axis (CrAs: MnP-type *Pnma*) with the incommensurable propagation vector $\boldsymbol{k} = (0\ 0\ k_c)$, $k_c \approx 0.356$. In order to understand the interplay of the superconductivity and the helimagnetism, the magnetic structure has to be determined in the *p*-*T* multiparameter region in the superconducting phase region.

Single-crystal neutron diffraction experiments were performed on the hot neutron diffractometer D9. To full data sets were measured, one at 2 K and a nominal pressure of 1.7 kbar and one at 2 K and a nominal pressure of 12 kbar. The data point at 2 K / 12 kbar is in or near the superconducting phase region, the data point at 2 K / 1.7 kbar serves as reference point in the purely magnetic phase region. To perform these experiments, a TiZr monobloc clamp cell provided by the ILL high-pressure laboratory was used. An as-grown CrAs single-crystal (only cut to a suitable length) was oriented using the in-house neutron Laue camera OrientExpress and loaded into the clamp cell. The nominal pressure was applied at room temperature using a hydraulic press. After the clamping, the cell was inserted into the cryostat of D9 and cooled to the base temperature of 2 K. The pressure was assumed to stay constant, and the inner part of the cell (at the sample position) was assumed to be in thermal equilibrium with the cryostat.

After the respective determination of the sample's orientation matrix, both nuclear and magnetic reflections were measured, amounting to a total of approximately 580 nuclear and 100 magnetic reflections at each of the two temperature/pressure points.

In addition to the two full data sets, the temperature-dependence of a strong magnetic reflection (0 0 1.65) was measured upon heating the cell after the measurement at high pressure. Following the intensity of this reflection allows to experimentally determine the pressure to which the sample was subjected on the basis of the published phase diagrams of CrAs.

Measurements of the pressure-dependence of the (0 0 1.65) reflection as originally proposed could not be performed due to the available beam time and the long time that would have been needed for cooling/heating and force loading at each point.

On the basis of the measurement at a nominal pressure of 0.17 kbar, the magnetic structure of the antiferromagnetic phase was analyzed. This analysis shows conclusively that the magnetic structure reported for CrAs in the literature is not the correct one, as the measured neutron single-crystal data do not fit the double-helical model. This is clearly reflected in unsatisfactory agreement factors. While our data do not allow the unambiguous determination of the actual magnetic model, we obtain several new models for the magnetic structure of CrAs that fit the data equally well. These are described in the magnetic superspace groups $P2_1.1'$(a0g)0s, $Pa.1'$(00g)0s, $P\bar{1}.1'$(abg)0s, and $P1.1'$(abg)0s, in each case with the restriction of constant and equal magnetic moments for all Cr sites. Following this restriction, all models are spiral-like (rotating constant magnetic moment), with no spin-density wave character that would cause a non-constant magnetic moment. The common feature of a value of about 3.2(2) μ_B for the magnetic moment of the Cr atoms indicates that also the previously assumed value ~1.7 μ_B is not correct. Since the electronic model used for CrAs is based on this value, our result has far-reaching consequences also for properties of CrAs not directly coupled to the magnetic moment itself.

The basic reason for the discrepancy in the magnetic model between the literature and our measurement lies in the used experimental method: the model was reported and validated only on the basis of neutron powder data, which are not suitable to allow a distinction of the double-helical model and the new models mentioned above. This is also confirmed by additional neutron powder measurements which we performed.

To determine the actual magnetic structure unambiguously, polarized neutron single-crystal diffraction experiments could be of great help. Assuming that the magnetic structure of CrAs does not change within the antiferromagnetic phase region – for which there are no indications so far – these experiments do not have to be conducted at high pressure, which would be very challenging for polarized neutron radiation. Hence, purely temperature-dependent single-crystal experiments below T_N = 267 K using polarized neutrons might be sufficient for an unambiguous identification of the correct model.

[1] W. Wu, J. Cheng, K. Matsubayashi, P. Kong, F. Lin, C. Jin, N. Wang, Y. Uwatoko and J. Luo, *Nat. Commun.* **5**, 5508 (2014).