Experimental report

29/01/2024

Proposal:	**CRG-2758**	**Council:**	4/2020

Title: High-field neutron diffraction investigation on the field induced spin super-fluid and super-solid states of $MnCr_2S_4$.

Research area: Physics

This proposal is a new proposal

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Samples: $MnCr_2S_4$

Instrument	Requested days	Allocated days	From	To
IN22	9	9	15/06/2021	24/06/2021
IN3	1	1	10/06/2021	11/06/2021

Abstract:

$MnCr_2S_4$ is a bond-frustrated spinel compound with magnetic Mn^{3+} and Cr^{3+} ions. It has attracted immense interest in recent years for showing extremely rich magnetic phase diagram. $MnCr_2S_4$ undergoes two magnetic transitions:(i) at $T_c = 65$ K to a ferrimagnetic state and (ii) at $T_{YK} = 5$ K to a canted-spin state known as Yafet-Kittel (YK)type phase. Recent single crystal magnetization and ultrasound experiments by Tsurkan et al. (one of the co-proposers) up to 60 T between 1.5 K and 28 K revealed that under the influence of an external magnetic field, $MnCr_2S_4$ exhibits a manifold of competing spin states which can be described as spin-superfluid and spin-supersolid phases. To extract direct information about the field induced magnetic structure, we have successfully performed single crystal neutron diffraction measurements up to 26 T on $MnCr_2S_4$. We now aim to perform diffraction up to 40 T using pulsed-field to get information about the spin-structure in the higher field regime.

Experimental report for CRG-1302: High-field neutron diffraction investigation of the field induced spin superfluid and supersolid states of $MnCr_2S_4$

IN22 spectrometer, 15-23 June 2021

Experimental team: Frédéric Bourdarot, Sumanta Chattopadhyay, Fabienne Duc, Virginie Simonet and Manila Songvilay

Scientific background:

$MnCr_2S_4$ is a bond-frustrated spinel compound with magnetic Mn^{2+} and Cr^{3+} ions, which undergoes two magnetic transitions: (i) at T_C = 65 K to a ferrimagnetic order and (ii) at T_{YK} = 5 K to a triangular type canted spin structure known as the Yafet-Kittel (YK) state [1]. Recent single crystal magnetization and ultrasound experiments up to 60 T and ultra-high magnetization up to 110 T revealed an extremely rich magnetic phase diagram exhibiting five field induced phase transitions at $\mu_0 H_1 \approx 11$ T, $\mu_0 H_2 \approx 25$ T $\mu_0 H_3 \approx 50$ T, $\mu_0 H_4 \approx 75$ T and $\mu_0 H_5 \approx 85$ T [2-3] (see *H-T* phase diagram in figure 1).

To clarify the magnetic structures of these competing spins states, we have undertaken a systematic study by means of single crystal neutron diffraction experiments in zero field and in fields up to 6 T on the D23 diffractometer (exp. 5-41-1018, Feb. 2020) and up to 26 T on the HFM/EXED diffractometer at the HZB (Nov. 2019). In both experiments, the magnetic field was applied along the [111] direction. Thanks to the large number of reflections collected on D23 at 1.6, 12 and 50 K, we were able to determine the magnetic structures in zero field and in fields up to 6 T and to demonstrate the existence of the YK state even in zero field. At 1.6 K, a spins arrangement consisting of 12 magnetic domains has been revealed in zero field, the Cr spins being aligned along the <111> directions and the Mn spins corresponding to the YK canting (with Mn1 spins parallel to [110] and Mn2 spins parallel to [-1-10]). In addition, the data measured at low field revealed a selection of magnetic domains with the [111] axis along the field. These results were taken as a starting point to investigate the magnetic structures in the high field regime.

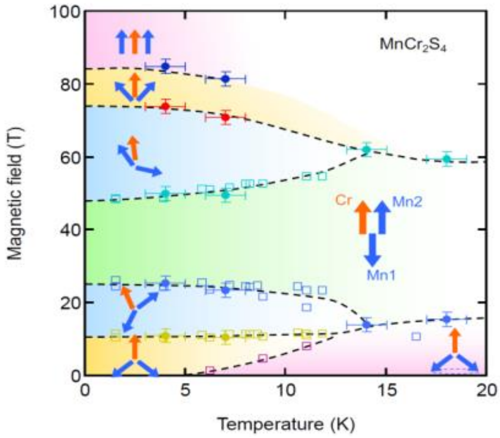

Figure 1: H-T phase diagram of $MnCr_2S_4$ obtained from high-field magnetization, ultrasound and magnetostriction experiments [2-3]. Theoretically obtained magnetic structures revealed by Monte Carlo calculations are illustrated in each phase with Cr (orange arrows) and Mn spins (two blue arrows).

On the HFM/EXED diffractometer, it was not possible to collect a full set of data because of the constraint of the 26-T magnet: 3 selected magnetic reflections, viz. (0 -2 0), (1 -3 1) and (2 -2 0), have been measured at low temperatures (2 and 12 K) and fields up to 26 T. Using these data and the results obtained at 1.6 K and 6 T, combined with high field magnetization data, we were able to determine the magnetic structure in the phase between H_1 and H_2, the application of a high field along [111] leading to the selection of 6 domains out of the 12 detected at low field.

Aim of the proposal:

As an extension to this ongoing project, we proposed to perform pulsed field based single crystal neutron diffraction measurements up to 40 T at several temperatures between 2 and 80 K. This experiment aimed to determine the magnetic structure of the robust magnetization plateau observed between $\mu_0 H_2 \approx 25$ T and $\mu_0 H_3 \approx 50$ T to gather additional information on the spin-structure evolution well beyond the H_2 phase-boundary.

Experimental setup:

This experiment was carried out on the CEA-CRG thermal neutron spectrometer IN22 (CEA-CRG), operated in the double-axis mode (without analyser) and equipped with the pulsed field set-up described in Ref. [4], including a 1.15 MJ generator and a 40-T conical pulsed magnet. A wavelength of 1.208 Å provided by the (200) reflection of a PG monochromator was used.

Two single-crystal samples were pre-aligned using IN3 and glued onto 2 different sapphire supports for the measurements of the (002) and the (0-22) reflections, both specimens being of dimensions 2 x 2 x 1-2.5 mm^3.
To reach the (002) reflection, the field was applied along the [1 1 0], the scattering plane being defined by the [1 1 0] and [0 0 1], whereas it was applied along [1 1 1] to measure the (0 -2 2) reflections, the horizontal scattering plane being this time fixed by [1 1 1] and [0 -1 1].

Results:
We started the measurements on the sample with the scattering plane defined by [1 1 0] and [0 0 1], applying the field along [1 1 0]. The nuclear Bragg peak (0 0 4) was first measured at room temperature to check the alignment of the crystal. The temperature was then lowered to the base temperature T = 2 K. After adjusting the alignment of the sample at low temperature, we have accumulated more than 200 magnetic field pulses up to 35 T to acquire the field dependence of the diffracted intensity at the position **Q** = (0 0 2). The result obtained is shown in figure 2. A clear plateau regime is observed for field above 25 T in agreement with the high field magnetization data.

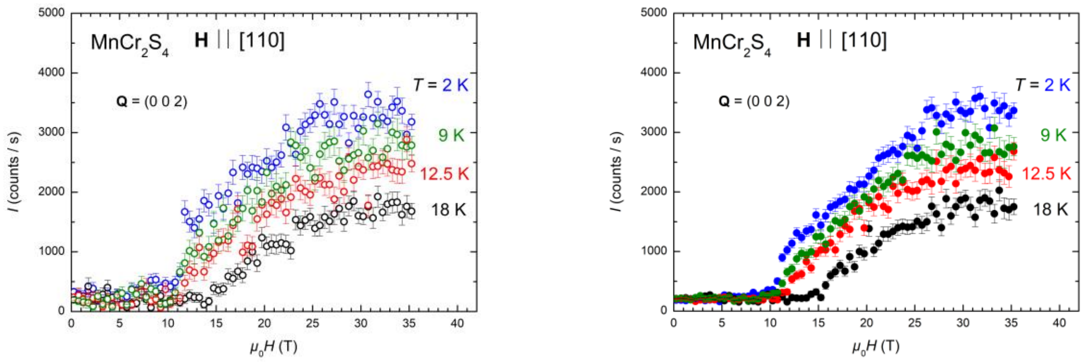

Figure 2: Field dependences of the diffracted intensities at the position **Q** = (0 0 2) with increasing (open symbols, left) and decreasing field (solid symbols, right) as recorded with the incident wavelength λ = 1.208 Å. The higher statistics of the data measured in the decreasing field is entirely related to the asymmetric sweep rate of the field pulse for rising and falling fields [4].

Data were then collected at T = 9, 12.5 and 18 K, accumulating more than 130 magnetic pulses at each temperature.
Next, one day was dedicated to warm the cryostat and to change the sample. The second sample with the scattering plane defined by [1 1 1] and [0 -1 1] was mounted inside the cryomagnet and cooled down to 2 K. Then, based on the results obtained on the D23 and HFM/EXED diffractometers, we looked for magnetic signal at **Q** = (0 -2 2). In this configuration, only this peak was accessible with the magnet environment. However, a series of peaks was detected close to the position of (0 -2 2) revealing the high mosaïcity of the sample.

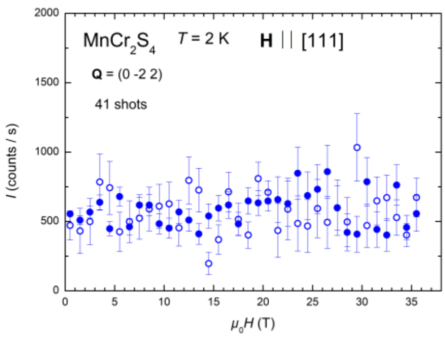

Figure 3: Field dependence of the diffracted intensities at the position **Q** = (0 -2 2) and at T = 2 K, with increasing (open symbols) and decreasing field (solid symbols) as recorded with the incident wavelength λ = 1.208 Å.

Considerable time was spent selecting the peak. A candidate peak was chosen. Pulsed-field

measurements up to 35 T were then started as time was progressing, looking for changes in the (0 -2 2) Bragg peak. No changes were detected (figure 3) in disagreement with what we measured previously on the HFM/EXED diffractometer. Other peaks were then tested but none of them shows a change in field.
In spite of this problem, the data collected on **Q** = (0 0 2) with the field applied along [1 1 0] give us the possibility to propose a model of magnetic structure for field above $\mu_0 H_2 \approx 25$ T.
A paper that will gather the complementary results obtained in the three datasets mentioned above is in progress.

References:
[1] Y. Yafet, and C. Kittel, Phys. Rev. **87**, 290 (1952).
[2] V. Tsurkan et al., Sci. Adv. 3, e1601982 (2017).
[3] A. Miyata et al., Phys. Rev. B 101, 054432 (2020).
[4] F. Duc et al., Rev. Sci. Instrum. 89, 053905 (2018).