Experimental report

15/02/2021

Proposal: 5-11-439 **Council:** 10/2019

Title: The Nuclear Structure of MnSb2O6 in single crystals:

Research area: Physics

This proposal is a new proposal

Main proposer: Chris STOCK

Experimental team: Navid QURESHI

Local contacts: Oscar Ramon FABELO ROSA

Samples: MnSb2O6

Instrument	Requested days	Allocated days	From	To
D9	3	4	28/01/2021	02/02/2021

Abstract:

We have previously applied polarized neutrons to investigate the magnetic and structural chiralities in single crystals of MnSb2O6. We have found a dominant magnetic chiral domain in our crystals, but no corresponding structurally chiral domain through the application of Schwinger scattering. However, the Schwinger scattering results are highly sensitive to the nuclear structure factors. We therefore request 3 days of time on D9 to measure the nuclear structure factors in our single crystals. Neutrons are preferred over x-rays so that an average can be done over the entire crystal therefore repeating the conditions used for polarized neutrons.

Proposal 5-11-439, 5 days on D9
The Nuclear Structure of $MnSb_2O_6$ in single crystals

Previous results

Powder diffraction data on $MnSb_2O_6$ was refined in P321 space-group. A refinement in C2 (subgroup of P321) was attempted to see eventual magneto-structural effects below T_N=12.5K.

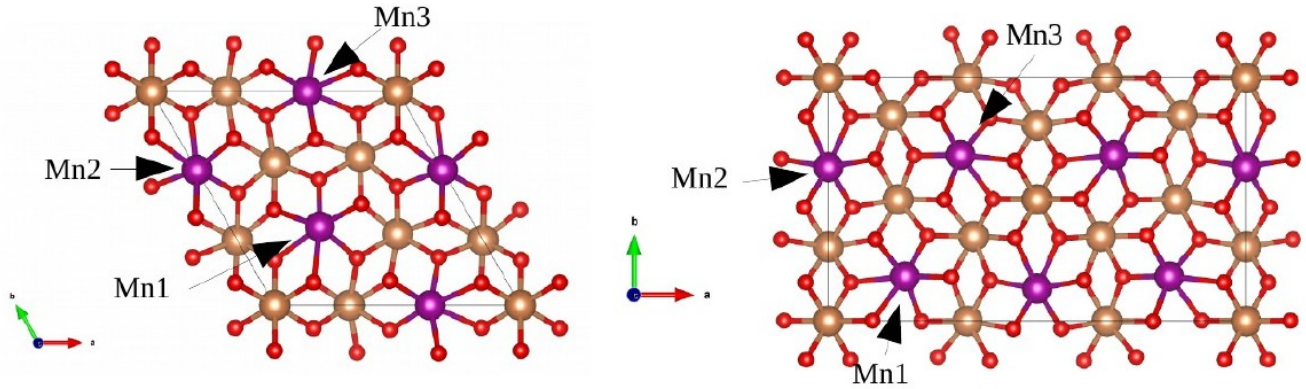

Fig. 1: Nuclear structure of $MnSb_2O_6$. Left: in P321 space-group where Mn atoms are related by three-fold symmetry. Right: C2 space-group where only 2 Mn atoms are independent.

From BT1 data, it seems that the triangle of Mn atoms is effectively distorted at low temperature, but D2B data did not confirm this behaviour. The aim of this experiment on D9 is to determine which results are the most relevant.

Nuclear refinement

Nuclear Bragg peaks were measured both in the paramagnetic phase at 50K and in the magnetic phase at 2K. The data was refined in P321 space-group as reported previously by Johnson et al. [1].

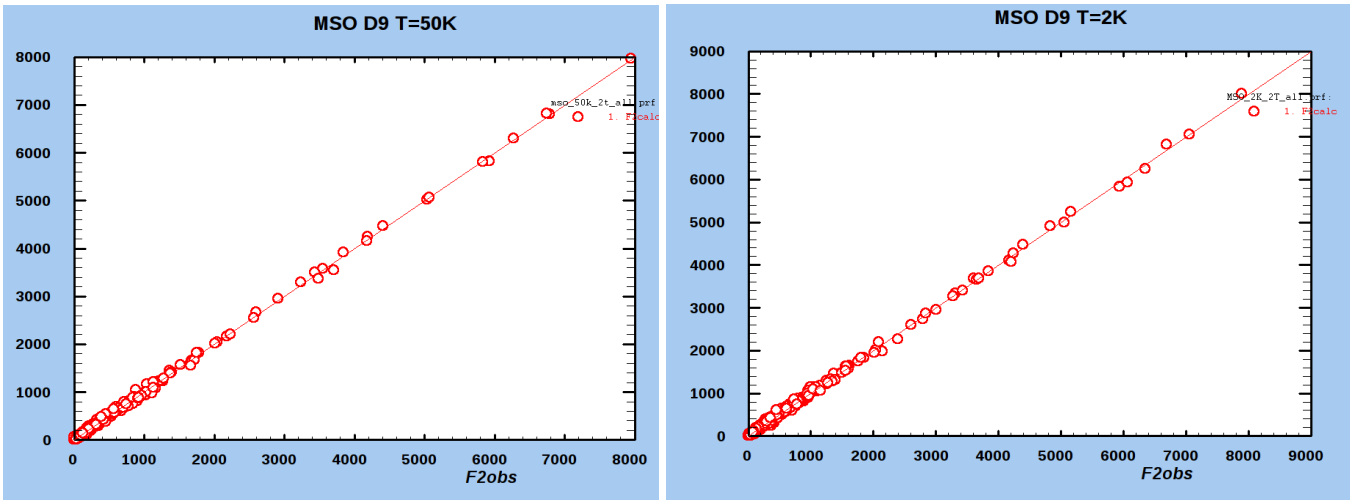

Fig. 2: Refinement of the nuclear structure of $MnSb_2O_6$ on Fullprof, in P321 space-group. Left: at 50K, R_F = 4.874. Right: at 2K, R_F = 5.120.

406 independent nuclear reflections were measured at 50K. 529 independent nuclear reflections were measured at 2K, out of 845 reflections, to check if the reflections are really equivalent in P321 symmetry. The refinement are shown in Fig. 2.

P321 space-group is in agreement with the data at both 50K and 2K. A tentative to refine the data in reduced P1 symmetry was attempted, by only letting free the positions of the Mn atoms. This refinement did not improve ($R_F = 5.7$) the previous analysis, and the eventual distorsion of the Mn atoms triangle is within the error bar. This disproves the structural transition indicated by our previous studies.

Structural twins

Four structural twins can coexist in P321 space-group. We can label them as (hkl), $(\bar{h}\bar{k}l)$, $(\bar{h}\bar{k}\bar{l})$, $(hk\bar{l})$, Actually, in terms of Bragg reflections, (hkl) (resp. $(\bar{h}\bar{k}l)$) and $(\bar{h}\bar{k}\bar{l})$ (resp. $(hk\bar{l})$) give Friedel pairs, thus are indistinguishable by unpolarized neutrons. But two-fold twins do lead to different nuclear structure factors. That is why the nuclear refinement was performed considering two domains.

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=> Domain fraction (  1):          0.008729        0.002921
=> Domain fraction (  2):          0.991271        0.002921
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From the nuclear refinement (at both 50K and 2K), it seems that only two (>99%) out of four structural twins are found present in this $MnSb_2O_6$ single-crystal, namely $(\bar{h}\bar{k}l)$ and $(hk\bar{l})$ (with respect to the indexation in D9). This gives a constraint on our Schwinger scattering [2] experiment performed on D3 on the same single-crystal.

As shown in Fig. 3, the measured flipping ratios are fitted to a linear combination of flipping ratios for twins $(\bar{h}\bar{k}l)$ and $(hk\bar{l})$. Both structural twins are found present in the single-crystal with a proportion ~66%/33%.

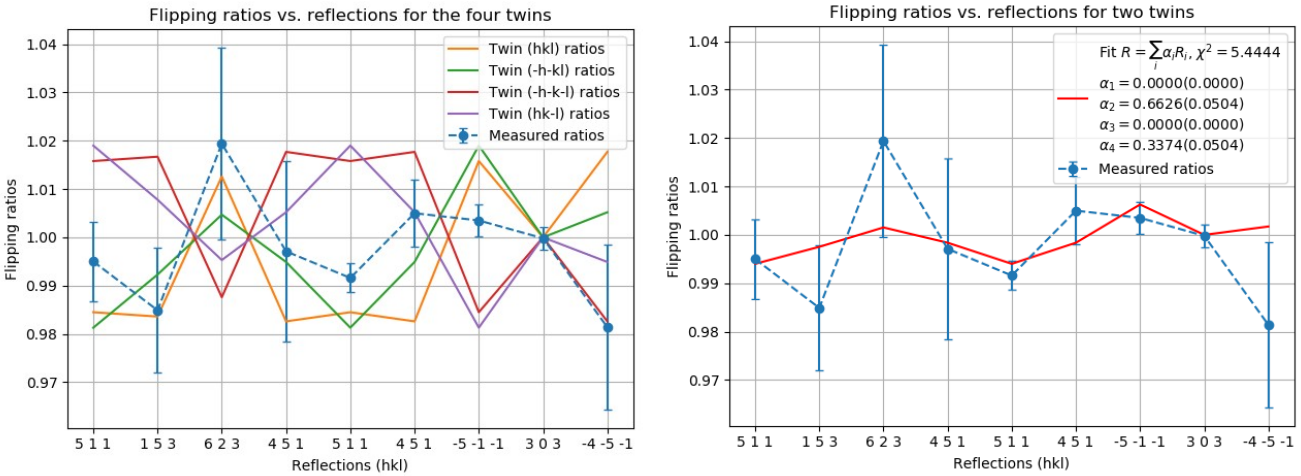

Fig. 3: Left: Calculated flipping ratios for the four structural twins, and measured ones for different nuclear reflections. Right: Population fit with the constraint found on D9.

These two twins are related by inversion symmetry and thus, a non-racemic mixture of two chiral structural domains is found in the compound. This is the same conclusion as in the previous study by Johnson et al. [1], but the structural twins were not taken into consideration. Later on, Kinoshita et al. [3] found an unique domain in terms of structure factors (as in our study), but they attributed this to the presence of a single structural chirality in their single-crystal. As shown in our study, this can not be stated without performing Schwinger scattering to unambiguously determine the proportions of structural twins.

Magnetic refinement

At the end of this experiment, we had collected 262 inequivalent magnetic reflections at 2K on D9. The magnetic structure was refined using Mag2Pol, and a co-refinement with our previous spherical neutron polarimetry (SNP) data was attempted.

The refinement favors the tilted cycloid model proposed by Kinoshita et al. [3] over the former straight cycloid model proposed by Johnson et al. [1]. It also indicates the presence of a single negative tilt of the cycloids from the c-axis, which somehow agrees with the dominant tilt (~90%) found from our SNP data. But the statistics of the fit are not good enough ($R_{F,mag}$=25.37) to directly co-refine with the SNP data. Further studies are necessary to deduce the magnetic structure of $MnSb_2O_6$ in single-crystals.

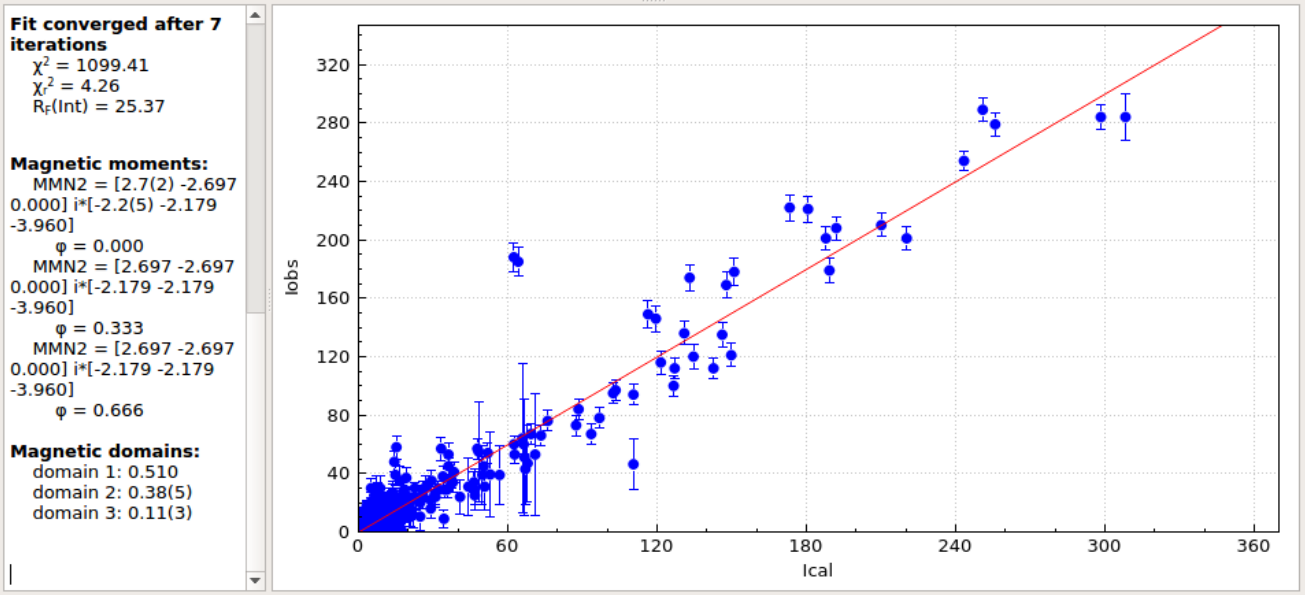

Fig. 4: Refinement of the magnetic structure in Mag2Pol, using the tilted cycloid model proposed by Kinoshita et al. [3]

[1] R. D. Johnson *et al.* Phys. Rev. Lett. **111**, 017202 (2013).
[2] J. Schwinger Phys. Rev. **73**, 407 (1948); C. G. Shull Phys. Rev. Lett. **10** 297 (1963).
[3] M. Kinoshita *et al.* Phys. Rev. Lett. **117**, 047201 (2016).