Experimental report

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Proposal: 5-31-2548 **Council:** 4/2017

Title: The Crystal and Magnetic Structureof Sr2Mn2CrAs2O2

Research area: Chemistry

This proposal is a new proposal

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Samples: Sr2Mn2CrAs2O2

Instrument	Requested days	Allocated days	From	To
D1B	1	1	03/07/2018	04/07/2018
D2B	2	2	28/03/2018	30/03/2018

Abstract:

Manganese oxypnictides such as LnMnAsO1-xFx (Ln = La, Ce, Pr, Nd) exhibit fascinating electronic and magnetic properties such as colossal magnetoresistance (CMR), spin reorientation transitions and semiconductor insulator transitions. We are now investigating more complex Mn oxypnictides in order to reveal their electronic and magnetic properties. We have recently synthesized the new phase Sr2Mn2CrAs2O2. Sr2Mn2CrAs2O2 has a body centered tetragonal crystal structure that consists of MO2 (M=Cr and/or Mn) oxide layers similar to the CuO2 layers in high temperature superconducting cuprate superconductors, and intermetallic MAs (M=Cr and/or Mn) layers similar to the MnAs layers in the oxypnictides described above. SQUID magnetometry measurements show two magnetic transitions below 150 K and 50 K. These transitions most likely arise from magnetic order within the MAs and MO2 layers respectively. In order to determine the crystal and magnetic structures of Sr2Mn2CrAs2O2 below the two magnetic transitions at 150 K and 50 K we request time on the high resolution diffractometer D2B and the high intensity diffractometer D1B.

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Layered oxypnictides, such as $Sr_2Mn_3As_2O_2$ [1], have been investigated in order to discover their magnetic and electrical behaviour. There are two positions for the Mn ions in this structure, the first M(1) has square planar geometry MO_2^{2-} similar to that seen in high T_C cuprate superconductors. The second position M(2) is also 4-fold coordinated but with arsenic forming $M_2As_2^{2-}$ layers like those in iron arsenide superconductors. $Sr_2Mn_3As_2O_2$ has been identified as a narrow band-gap semiconductor with an activation energy of 133 meV through electrical resistivity measurements. The ground state of this material is a G-type antiferromagnet with ordering temperature of 340 K, the spin alignment occurs on the M(2) site [1]. A chromium analogue $Sr_2Cr_3As_2O_2$ has also been studied which also orders antiferromagnetically below 590 K [2]. We have recently prepared and analysed a mixed manganese and chromium analogue, $Sr_2Mn_2CrAs_2O_2$.

A variable temperature neutron diffraction study of $Sr_2Mn_2CrAs_2O_2$ has been performed between 1.5 and 450 K at the Institut Laue-Langevin on the diffractometers D1B and D2B. Results show $Sr_2Mn_2CrAs_2O_2$ crystallises in the tetragonal space group *I4/mmm* with manganese and chromium splitting occupancy between the M(1) and M(2) positions. The Mn predominantly occupies the M(2) position with a site occupancy of 0.98. The Mn and Cr occupancies on the M(2) site are 0.32 and 0.68 respectively

Below 430 K the M(2) spins align antiferromagnetically parallel to *c* with propagation vector k = (0, 0, 0), the magnetic structure at 300 K can be seen in Figure 1(a). The Rietveld refinement at this temperature (Figure 2 (c)) shows an excellent fit to this model, with calculated *a* and *c* parameters of 4.0935(1) and 19.0646(6) Å respectively (χ^2 = 2.25, R_{WP} = 11.1%). The manganese magnetic moment (Mn_{000}) was calculated as 2.57(2) μ_B and this magnetic phase persists down to 145 K.

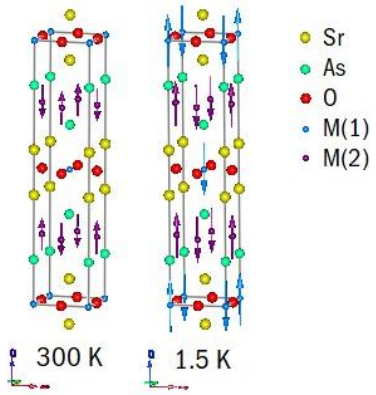

Figure 1: (a) Magnetic structure at 300 K (left), (b) Magnetic structure at 1.5 K (right).

Below 165 K the manganese spins realign on M(2) position with a new k vector (1, 0, 0). The magnetic moments are still parallel to *c*, they are antiferromagnetic in the *ab* plane but ferromagnetic along *c*. At the same time the Cr spins on the M(1) site order antiferromagnetically. The Cr magnetic phase doubles the *a* and *b* magnetic cell parameters from the structural cell parameters as this phase has the propagation vector k = (½, ½, 0). The chromium spins align parallel to *c*, they are ferromagnetically ordered along *c* and

antiferromagnetically ordered along the *a* and *b* plane. The magnetic structure at 1.5 K can be seen in Figure 1(b), Rietveld refinement to the magnetic and structural model at this temperature (Figure 2(a)) shows a good fit to the data and calculated *a* and *c* as 4.0833(1) and 18.9933(3) (χ^2 = 3.12, R_{WP} = 12%). The magnetic moments were calculated for Mn (Mn_{100}) and Cr as 3.38(2) and 3.94(6) μ_B respectively.

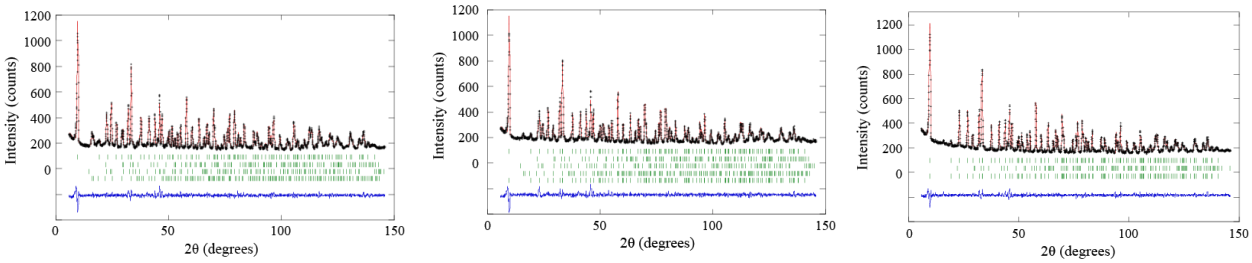

Figure 2: (a) Rietveld refinement fit at 1.5 K (left), (b) 160 K (middle), (c) 300 K (right).

Between 145 and 165 K three magnetic phases are all present within the sample, Rietveld refinement at 160 K (Figure 2(b)) shows an excellent fit to the three magnetic models so that there is magnetic phase segregation.

The calculated magnetic moment at 160 K for the three phases were 1.86(1), 1.97(3) and 2.31(13) μ_B for Mn_{000}, Mn_{100} and Cr respectively. A cyclic refinement was performed between 10 K and 400 K with data recorded on the D1B instrument. The variation of the magnetic moments versus temperature are show in Figure 4, again evidencing magnetic phase segregation between 145 K and 165 K. The *a* and *c* values were also calculated for the full temperature range and are plotted in Figure 5(a) and (b).

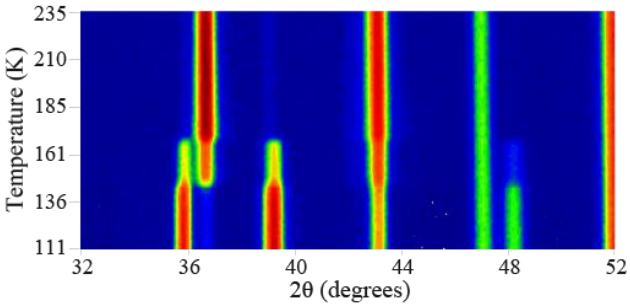

Figure 3: The temperature variation of the peak intensities recorded on D1B beam line. The region of magnetic phase segregation is visible from around 165 to 140 K.

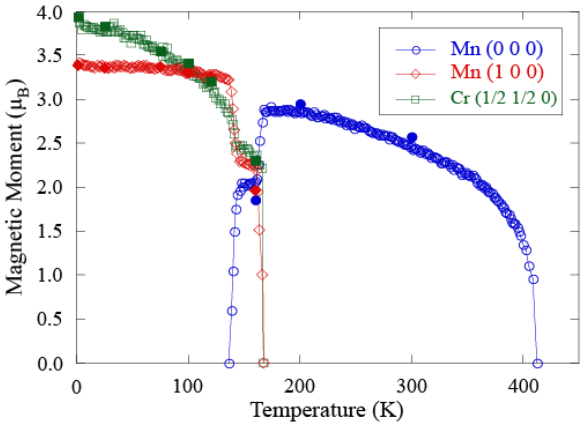

Figure 4: Magnetic moments calculated from cyclic refinements of D1B data. The calculated moments from D2B are also plotted as filled in points.

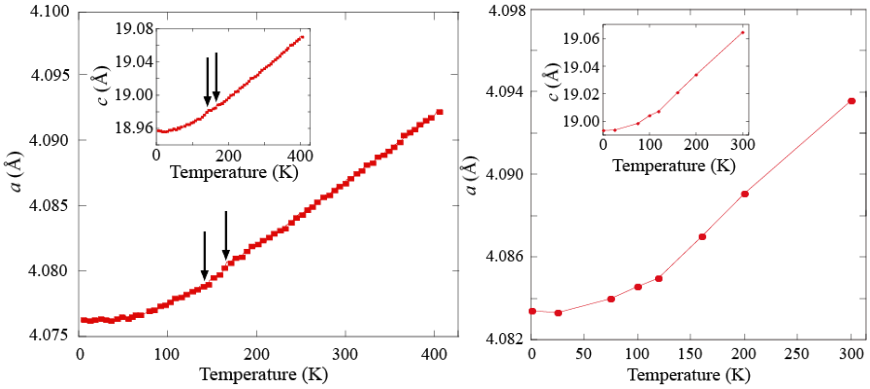

Figure 5: (a) Plot of the calculated cell parameter *a* from D1B data with *c* inset (left), (b) Plot of the calculated cell parameter *a* from D2B data with *c* inset (right). A subtle anomaly is observed in the cell parameters as the Cr moments magnetically order.

References

[1] Nath, R., Garlea, V., Goldman, A. and Johnston, D., Phys. Rev. B, **81**, 224513 (2010).

[2] Jiang, H., Bao, J., Zhai, H., Tang, Z., Sun, Y., Liu, Y., Wang, Z., Bai, H., Xu, Z. and Cao, G., Phys. Rev. B, **92**, 205107 (2015).