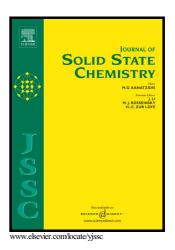
### Author's Accepted Manuscript

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Emily C. Hunter, Peter D. Battle



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#### **ACCEPTED MANUSCRIPT**

# Evolution of the crystal structure and magnetic properties of Sr<sub>2-x</sub>Ca<sub>x</sub>CrSbO<sub>6</sub> with composition

Emily C. Hunter and Peter D. Battle\*

Inorganic Chemistry Laboratory, University of Oxford, South Parks Road, Oxford, OX1 3QR, U. K.

\*author to whom correspondence should be addressed: peter.battle@chem.ox.ac.uk

#### **Abstract**

Polycrystalline samples of compositions in the perovskite solid solution  $Sr_{2-x}Ca_xCrSbO_6$  (x=0, 0.25, 0.5, 0.75, 1.0, 1.25, 1.50, 1.75, 2.0) were synthesized and characterised by synchrotron x-ray diffraction and SQUID magnetometry. Neutron powder diffraction data were collected on the x=0, 0.25, 0.5, 1.0, 1.5 and 2.0 compositions. It was found that  $Sr_2CrSbO_6$  crystallised in the triclinic  $I\overline{1}$  space group. The other compositions were all found to crystallise in the monoclinic space group  $P2_1/n$  although  $Sr_{1.5}Ca_{0.5}CrSbO_6$  was found to be phase separated into a calcium-deficient and calcium-rich phase, both having the space group  $P2_1/n$ .  $Sr_2CrSbO_6$  was found to order antiferromagnetically at 12 K and the ordered  $Cr^{3+}$  moment was refined to be 2.39(1)  $\mu_B$ . The antiferromagnetic order was rapidly quenched on doping with Ca and compositions with x≥0.75 exhibited ferromagnetic order.  $T_C$  increased with x to reach a maximum of 14 K at x=2.0, with an ordered moment of 2.56(3)  $\mu_B$  per  $Cr^{3+}$  cation. The tilt angles of the B-site octahedra about [110] and [001] increase from 5.4(1)° and 4.6(1)° to 13.7(1)° and 13.9(1)°, respectively, across the series; the switch from antiferromagnetism to ferromagnetism occurs when both tilts are at a value of ~8°.

**Graphical Abstract:** 

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