Local distortions and long range antiferromagnetic order in Mn based antiperovskites

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Magnetic spins arranged on a triangular lattice with antiferromagnetic interactions of equal strength between them is a recipe for frustration. Antiperovskites have a generic formula X_3AB, where X is a transition metal like Mn, A are main group elements like Ga, Ge, Sn, etc. and B are nonmetals like C, N, etc. Here even though the Mn atoms occupy the face centered sites of a cube, collinear antiferromagnetic order is stabilized via a cubic-cubic volume expanding first order magnetic transition. Using extended x-ray absorption fine structure (EXAFS) at the metal K edges in some Mn antiperovskites like Mn_3GaC, Mn_3SnC, Mn_3InC, Mn_3GeC, Mn_3ZnC, we attempt to understand the mechanism of the magneto-structural transition that leads to stabilization of collinear antiferromagnetic order.

The antiperovskites were prepared by solid state reaction technique by vacuum sealing the elemental powders and subjecting them to a heat treatment at about 800^{o}C for several days. The compounds were characterized by x-ray diffraction for their structure and phase purity. Magnetic properties as a function of temperature and magnetic field were measured using SQUID magnetometer while their magnetic structures were determined using neutron diffraction. EXAFS was recorded at several temperatures in 50K – 300K interval at the Mn K as well as at the K edges of other metal atoms like Ga, Sn, In etc. using the P65 beamline at Petra III synchrotron, DESY, Hamburg. The data were analyzed using Athena and Artemis in the Demeter suite.

All compounds studied here crystallize in cubic perovskite structure (Space Group Pm-3m). A volume discontinuous cubic-cubic phase transition is noted in Mn_3GaC and Mn_3SnC at 170K and 280K respectively. Magnetically, Mn_3GaC and Mn_3ZnC have a collinear antiferromagnetic order while Mn_3SnC displays a complex magnetic ground state with presence of both ferromagnetic and antiferromagnetic order below 280K. Mn_3InC and Mn_3GeC tend to exhibit only a ferromagnetic order down to 5K while Mn_3ZnC is antiferromagnetic but does not undergo a volume discontinuous transition.

EXAFS results show that in all the above antiperovskites, the Mn sub-lattice is locally distorted resulting in short and long Mn-Mn bond distances. However, the EXAFS of A site atom (Ga, Sn, In, etc.) can be fitted with correlations belonging to cubic lattice symmetry. The magnitude of distortion in the Mn sub-lattice depends on the size of A site atom. If the A site atom is smaller, like Zn or Ga, the difference between long and short Mn-Mn bond distances is larger and vice versa. In case of Mn_3GaC, the short and long Mn-Mn distances are 2.69 Å and 3.1 Å respectively. Furthermore, it is also seen that the compounds with Mn-Mn bond lengths less than 2.75 Å have antiferromagnetic ground state. While those with Mn–Mn bonds longer than 2.75 Å have dominant ferromagnetic interactions. Based on detailed analysis of EXAFS data, various aspects like field induced ferromagnetism in Mn_3GaC, complex magnetic order in Mn_3SnC can also be understood.

In conclusion, EXAFS studies conclusively show that the long range antiferromagnetic order in some of Mn based antiperovskites is due to distortions in Mn sub-lattice and these distortions depend on the size of A site atom.