

*Neutron studies on high pressure A-site manganites.*

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The discovery of new materials with appealing functional properties is challenging and such properties are often observed in complex oxides containing transition metals in intermediate or unusual oxidation states which must be stabilised under special synthetic conditions.

A<sup>2+</sup> cations in perovskites synthesized at ambient pressures are typically large, nonmagnetic atoms, such as A = Ca, Sr, Ba, Pb. However, materials with the smaller high-spin Mn<sup>2+</sup> ion at A sites have been synthesized under high-pressure and high-temperature conditions. This may introduce additional functionality as found in MnVO<sub>3</sub> perovskite which is metallic but also has coexisting helimagnetic order of localized S = 5/2 Mn<sup>2+</sup> spins [1].

Several Mn<sub>2</sub>BB'O<sub>6</sub> double perovskites have also been synthesized at high pressures. Mn<sub>2</sub>FeReO<sub>6</sub> has a high Curie temperature of 520 K and similar ferrimagnetic and spin polarized conducting properties to the much-studied magnetoresistive material Sr<sub>2</sub>FeMoO<sub>6</sub>, but also shows a novel switch from negative to large positive magnetoresistances at low temperatures driven by Mn<sup>2+</sup> spin ordering [2]. In contrast, Mn<sub>2</sub>MnReO<sub>6</sub> (Mn<sub>3</sub>ReO<sub>6</sub>) shows successive antiferromagnetic ordering transitions for Re and Mn spins at 99 and 109 K, respectively. Our subsequent investigation of possible rare earth (R) double perovskites Mn<sub>2</sub>RSbO<sub>6</sub>, has led to the discovery of a new double double perovskite type for MnRMnSbO<sub>6</sub> with large R cations, as well as more conventional (MnR)MnSbO<sub>6</sub> double perovskites formed for smaller rare earth metal ions.

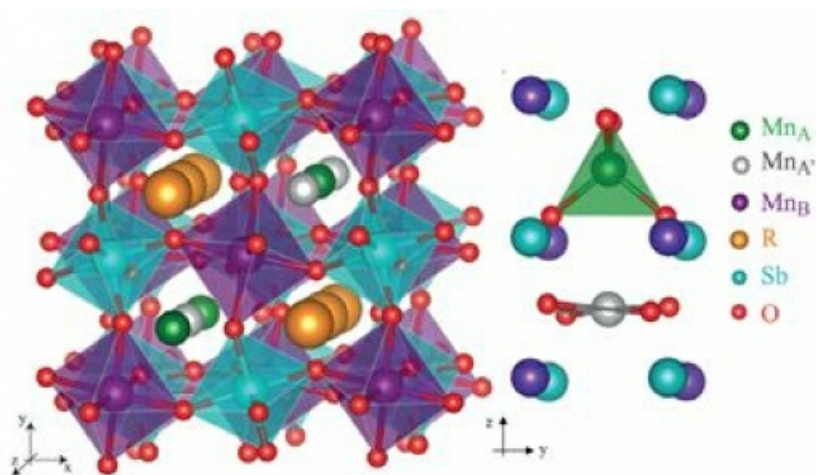
The new type of double double perovskite structure in the MnRMnSbO<sub>6</sub> (R= La, Pr, Nd, and Sm) family has a columnar order of A-site Mn<sup>2+</sup> and R<sup>3+</sup> cations whereas Mn<sup>2+</sup> and Sb<sup>5+</sup> have rock salt order on the B sites. Mn<sup>2+</sup> cations are in tetrahedral and square-planar A-site environments and octahedral B sites, see Figure 1 [3].

In this work, we present our endeavour on new Mn<sup>2+</sup> oxides obtained under high pressure along with structural and physical properties studies.

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[3] E. Solana-Madruga et al. (2016) Angew. Chem. Int. Ed.. 55,9340-9344.



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