Pb_2NiOsO_6: antiferromagnetic order breaks inversion symmetry in high pressure perovskite

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The compositional flexibility of double perovskites $A_2BB'O_6$ gives this family of materials a huge range of properties,[1] and interest in accommodating 5d cations such as Os^{6+} on a *B* sites stems from the stronger spinobit coupling for these heavier cations (compared with 3d transition metal cations). This gives interesting electronic and magnetic properties of these phases and the A_2NiOsO_6 (*A* = Ca, Sr, Ba) series spans insulating to metallic phases, with ferromagnetic to antiferromagnetic order,[2-4] and with half-metallicity proposed for Sr_2NiOsO_6.[5] These properties are very sensitive to Ni – O – Os bond lengths and angles and therefore to A^{2+} cation size.[2]

The lower symmetry environments favoured by $6s^2$ "inert pair" cations such as Pb^{2+} give structures and properties that can be quite different from those observed for the group 2 *A* cation analogues. [6, 7] However, high pressure synthetic routes are often required to access these lead analogues.[1]

This presentation describes work on the structural characterisation and properties of Pb_2NiOsO_6 synthesised at high pressure (6 GPa, 1575 K).[8] The rocksalt ordering of NiO_6 and OsO_6 octahedra combined with octahedral tilts gives a crystal structure of $P2_1/n$ symmetry (similar to Ca_2NiOsO_6). Strong coupling between Ni^{2+} and Os^{6+} moments give long-range magnetic order below 58 K, with the collinear magnetic structure described by magnetic propagation vector k = (½ 0 ½) (similar to Pb_2CoOsO_6[6]). This magnetic order, imposed on the (*B*-site ordered) crystal structure, breaks inversion symmetry.[8]

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