

Links between structure, SOC and magnetism in Ir double perovskites

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The iridates are currently a fertile ground for studies of new physics driven by strong spin-orbit coupling (SOC). The comparable energy scales of the SOC, Coulomb (U) and crystalline electric field (CEF) interactions give rise to exotic states as well as a delicately balanced ground state, which is largely sensitive to structural details and drives complex magnetic behaviors. The ability of Ir to exist in different oxidation states and the spatial extent of the 5d orbitals should also result in a rich variety of interactions and in a large sensibility to external stimuli. Moreover, recent works point out that the presence of extended 5d orbitals may also require accounting for longer-range exchange pathways beyond first neighbor exchange in order to understand magnetic phenomena [3, 6-8]. Clearly, in order to tailor the behavior of these materials we need first a better understanding of the local moments and magnetic interactions.

High Pressure XAS and XMCD measurements have been performed on $\text{Sr}_2\text{BiIrO}_6$ powdered samples at low temperature and at the $L_{2,3}$ edges of iridium to probe both, the strength of the spin-orbit coupling and the magnetic state. This information has been completed with high-pressure XRD measurements to determine the crystallographic changes undergone with increasing pressure.

Our results on $\text{Sr}_2\text{NiIrO}_6$, an Ir (IV) double perovskite, indicate that as we apply pressure the SOC ($\langle L \cdot S \rangle$) decreases, from 2.49 down to 2.02 under 18 GPa, and becomes similar to the values obtained on $\text{Sr}_2\text{ZnIrO}_6$ under ambient pressure. Similarly, the XMCD data show that the orbital to spin magnetic moment ratio, m_L/m_S , decreases, from -0.2 to -0.1, and also becomes similar to the values obtained on $\text{Sr}_2\text{ZnIrO}_6$ under ambient pressure. In addition the net magnetic moment is drastically reduced. Our results on Ir(V) perovskites indicate that $\langle L \cdot S \rangle$ presents a 14 % decrease from ambient pressure to 20 GPa.

Therefore, our results suggest that applying pressure causes a tilting of the angles that reduces the Ni-Ir(IV) interaction and increases the antiferromagnetic (AFM) character of the Ir sublattice. Regarding the strength of the SOC, the evolution of the XAS signals under pressure indicates a robust SOC in all the double perovskites, regardless of the Ir oxidation state.

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