

Electron doping in $\text{Ba}_2\text{Na}_x\text{Ca}_{1-x}\text{OsO}_6$ spin – orbit quantum oxides: a local structural study

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Spin-orbit oxides belong to the novel class of *quantum materials*, the physical properties of which are greatly influenced by the quantum mechanical nature of interactions between constituent electrons. In this context we study the local atomic and electronic structure of Osmium-based double perovskites (DPs) which exhibit a combination of strong spin-orbit coupling (SOC) and anisotropic exchange interactions. In this system SOC is of similar magnitude to crystal field and electron correlation effects (Hubbard U), making the prediction of properties more complex than for DP containing more well known 3d cations. We have recently studied the evolution of the electronic and structural properties of $\text{Ba}_2\text{Na}_x\text{Ca}_{1-x}\text{OsO}_6$ by substituting monovalent Na with divalent Ca cations. Muon spin spectroscopy and magnetization measurements indicate that the magnetic ground state changes from FM-canted for $x=1$ to AFM for $x=0$ with a monotonic increase of the magnetic transition temperature from 5 to 40 K.

Here we present a study of the changes of the material's local atomic and electronic structure in $\text{Ba}_2\text{Na}_x\text{Ca}_{1-x}\text{OsO}_6$, using a combination of XAFS and *ab-initio* simulations. XAFS is the method of choice to determine local atomic and electronic structure simultaneously; its combination with *ab-initio* simulations is particularly powerful. XAFS measurements at the Ca, Na, Os and Ba edges have been recorded and analyzed. The *ab-initio* calculations, performed with the Vienna Ab initio Simulation Package (VASP) within the non-collinear DFT+U+SOC scheme, provide support to the experimental evidences by delivering independent structural and electronic properties and allowing for a detailed microscopic interpretation of the results.

The extended range (EXAFS) data indicates that the local structure is the expected DP one, with a random distribution of Ca and Na and no anti-site defects; interatomic distances vary linearly with concentration. The Os L_3 near edge spectra (XANES) exhibit no shift with x , indicating that the net local charge on this cation does not change, despite the substitution of a monovalent cation with a divalent one. This indicates a delocalized distribution of the excess charge associated with an increased hybridization between Os-d and O-p states. The intensity of transitions to t_{2g} orbitals increases with x with respect to that relative to transitions to e_g ones. These experimental results are compared to the local electronic structure derived from *ab-initio* electronic structure simulations.