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Interplay of spin-orbit coupling, correlations, and crystal anisotropy in 5d oxides

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We investigate the correlated d -level electronic structure of 5d Ir and Os oxide compounds by fully *ab initio* quantum-chemical many-body calculations on finite embedded clusters. The wave-function quantum-chemical methods provide a promising alternative to density-functional-based approaches to the electronic structure of solids. The computed d - d excitations in square-lattice, honeycomb, pyrochlore, and chain-like iridates compare well with recent RIXS (resonant inelastic x-ray scattering) data. We also perform a detailed analysis of the relativistic spin-orbit wave functions and compute observables such as the $\langle \mathbf{L} \cdot \mathbf{S} \rangle$ ground-state expectation value of the spin-orbit operator. The latter is in principle accessible from x-ray absorption and provides information on the role of t_{2g} - e_g couplings in the ground-state wave function and on the strength of non-cubic fields that lift the degeneracy of the t_{2g} levels. As concerns the departure from cubic symmetry, interesting effects are found in $A_2\text{Ir}_2\text{O}_7$ pyrochlores, where the highly anisotropic, hexagonal configuration of the adjacent A -site ions breaks cubic symmetry even in the absence of O-ligand trigonal distortions and moreover competes with the latter. Our findings open new perspectives in pyrochlore oxides. In 227 iridates, the outcome of this competition is decisive for the actual realization of any type of non-trivial topological ground state. In 227 spin systems with $S > 1/2$, e.g., $\text{Cd}_2\text{Os}_2\text{O}_7$, this interplay decides the sign of the single-ion anisotropy and the degree of magnetic frustration.