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Orbital reconstruction through interlayer cation charge imbalance: insights from wave-function-based quantum chemistry calculations

LIVIU HOZOI, IFW Dresden, Germany

A promising route to engineering the electronic properties of quantum materials and devices rests on the idea of orbital reconstruction in multilayered oxide heterostructures. In this context, we identify and discuss in detail one simple, appealing mechanism for tailoring the sequence of d-electron energy levels: interplanar cation charge imbalance (ICCI) along successive metal-oxygen layers [1,2]. Through interplay with distortions of the ligand cages, it provides a knob for tunning the order of electronic levels in even intrinsically stacked oxides. We analyze in this regard electron spin resonance data for the 214 Sr-iridate oxide compound [1]. While canonical ligand-field theory predicts z-axis g factors smaller than 2 for positive tetragonal distortions as present in Sr214, the experiment indicates values larger than 2. This implies that the iridium d levels are inverted with respect to their normal ordering. State-of-the-art quantum chemistry calculations confirm the level switching in Sr214, whereas we find them to be instead normally ordered in the sister compound Ba214. Given the nonpolar character of the metal-oxygen layers, our findings highlight the tetravalent transition-metal 214 oxides as ideal platforms to explore d-orbital engineering in the context of oxide electronics. The crucial role of internal anisotropic fields related to the environment beyond nearest-neighbor ligands is further highlighted by ab initio quantum chemistry calculations on 3D pyrochlore osmates and iridates [3,4]. [References: [1] N. Bogdanov et al., Nature Commun. 6, 7306 (2015); [2] V. Katukuri et al., Inorg. Chem. 53, 4833 (2014); [3] L. Hozoi et al., Phys. Rev. B 89, 115111 (2014); [4] N. Bogdanov et al., Phys. Rev. Lett. 110, 127206 (2013).]