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Electrostatic Chemical Strain: An Approach to Electronic Structure Engineering in Layered Oxides[1]

JAMES RONDINELLI, Northwestern University

Traditional approaches to create and control functional electronic materials have focused on new phases in previously unknown bulk minerals. More recently, interlayer physics has spawned interest in known materials in unexplored atomic scale geometries, especially in complex transition metal oxides (TMO), where heterostructures and superlattices with abrupt interfaces can be created on demand. The interfaces between TMO overs a handle to direct the electrostatic field exerted on the transition metal centers via the coordinating oxygen ligands, which alter the M cation's *d*-orbital occupancies and spin state, thereby imparting desirable electronic functionality. In this talk, I describe an atomistic engineering approach that makes use of long-range electrostatic interactions between atomic metal-monoxide planes (AO and A'O) in naturally occurring superlattices, e.g., Ruddlesden-Popper (RP), phases, to tune interlayer atomic structure, orbital degeneracies, and magnetic properties. Using first-principles electronic structure calculations, I show how this electrostatic chemical strain (ECS) effect can be used to tune both crystal field energies and the frontier orbital structure in correlated (La,A)$\mathrm{NiO_4}$ RP phases at fixed stoichiometry. I describe how to enhance the Ni e_g orbital polarization, resulting in $\mathrm{NiO_6}$ units that exhibit a single $d(x^2-y^2)$ band at the Fermi level—electronic features similar to the layered superconducting cuprates. This approach is generic in construction, making it applicable to any layered topology supporting heterovalent cation substitutions. I conclude by showing it is a realistic strategy to tailor the electronic properties of known materials, and discover yet-to-be realized novel functional oxides without resorting to complex assembly of multi-component heterostructures.

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