Discovering New Tricks in Older Complex Oxides

Transition metal oxides offer an exciting platform for electronics owing to the allure of phenomena they offer, including ferroic functionality, correlated-electron behavior, and coexisting contraindicated properties. Owing to the sensitivity of their properties on (local and crystal) structure and composition, picoscale structure-property relationships are necessary to design function. Here, I briefly provide an overview of our progress in identifying these relationships and finding new phases through quantum-mechanical approaches combined with multiple materials-theory methods. Then, I describe two examples of how external perturbations to picometer scale distortions of bond lengths and angles produce unanticipated phenomena in thin films and bulk oxides of the form $A_{n+1}B_nO_{3n+1}$, originally discovered by Ruddlesden and Popper in the 1950s. First, although large epitaxial strains are believed to induce ferroelectricity, I show that biaxial strain induces an unforeseen polar-to-nonpolar (P-NP) transition in (001) thin films of $\text{Ca}_3\text{Ti}_2\text{O}_7$ ($n=2$) at experimentally accessible biaxial compressive and tensile strains. I explain the origin of the P-NP transition as arising from the interplay of anharmonic lattice interactions, which are directly strain tunable, and then propose a design rule for the appearance of the P-NP transition. Second, I identify a quasi-two dimensional (quasi-2D) phonon mode with a negative Grüneisen parameter in the layered titanate with quadratic dispersion akin to that found in 2D nanomaterials (graphene, monolayer transition metal dichalcogenides, etc.). This phonon mode can be utilized to realize unusual membrane effects, including tunable negative thermal expansion and a rare pressure-independent thermal softening of the bulk modulus. I conclude by emphasizing that older complex oxides, which are now understood to exhibit nontrivial anharmonicities, offer a plentiful playground for realizing new functionalities in thin film and bulk form.

Biography: James Rondinelli is the Morris E. Fine Junior Professor in Materials and Manufacturing at Northwestern University in the Materials Science and Engineering (MSE) Department, where he leads the Materials Theory and Design Group. His interests are in electronic structure theory and first-principles design of functional inorganic materials using atomic scale structure-property relationships. In 2016 he received a Sloan Research Fellowship in Physics, the Presidential Early Career Award for Scientists and Engineers (PECASE), and the 3M Non-Tenured Faculty Award. Additional honors include a NSF-CAREER Award (2015), DARPA Young Faculty Award (2012), and ARO Young Investigator Program (YIP) award (2012). He received the 2014 Ross Coffin Purdy Award from the American Ceramic Society and was named an Emerging Young Investigator by the Royal Society of Chemistry (J. Mater. Chem. C, 2016) and the American Chemical Society (Chem. Mater., 2014). Dr. Rondinelli has (co)-authored more than 95 peer-reviewed publications and holds 1 patent. He is a member of the APS, MRS, ACS, TMS, and AChemS, and has organized multiple symposia for these societies on the physics and chemistry of transition metal compounds. He serves as an editorial board member of npj Computational Materials (2015-) and is member of the MRS Academic Affairs Committee (2015-) and the Argonne Center for Nanoscale Materials (CNM) Users’ Executive Committee (2016-19). He received a B.S. in MSE from Northwestern (2006) and a Ph.D. in Materials from the University of California, Santa Barbara (2010). From 2010-2011, he was the Joseph Katz Named Fellow in the X-Ray Science Division at Argonne National Laboratory. Prior to joining Northwestern, he was an assistant professor at Drexel University (2011-14).