Polar Oxide Thin Films from Vacancy and Chemical Order

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Abstract

Here I describe a crystal-chemistry design approach for realizing digital oxides without inversion symmetry by combining two centrosymmetric compounds, utilizing periodic anion-vacancy order to generate multiple polyhedra that together with cation order produce a polar structure. The strategy is applied to two brownmillerite (perovskite-derived) oxides known to display centrosymmetric crystal structures in their bulk, $Ca_2Fe_2O_5$ and $Sr_2Fe_2O_5$, with ordered arrangements of oxygen vacancies dependent on a variety of competing crystal-chemistry factors. A microscopic understanding of the interactions among these structural descriptors, including ionic size, distortions of nominally regular oxygen octahedral, and in-plane and out-of-plane separation of tetrahedral chains, enables the design and experimental realization of epitaxial $(SrFeO_{2.5})_1/(CaFeO_{2.5})_1$ thin film superlattices. The ordered superlattices possess both anion-vacancy order and Sr and Ca chemical order at the subnanometer scale, confirmed through synchrotron-based diffraction and aberration corrected electron microscopy. Our resultsdemonstrate how control of anion and cation order at the nanoscale can be utilized to produce acentric structures markedly different than their constituents and open a path toward novel structure-based property design.

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Short Biography

Rondinelli is the Morris E. Fine Junior Professor in Materials and Manufacturing at Northwestern University (NU). His interests are in electronic structure theory and first-principles design of functional inorganic materials using picoscale structure-property relationships. He is a 2018 Kavli Frontiers of Science Fellow and the 2017 Materials Research Society (MRS) Outstanding Young Investigator. 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 also received the 2014 Ross Coffin Purdy Award from the American Ceramic Society. Rondinelli has (co)-authored more than 120 peer-reviewed publications and holds 1 patent. He is a member of the APS, MRS, ACS, TMS, ACerS, and ASEE. He is a Member-at-Large for the APS Division of Materials Physics and Chair of the Argonne Center for Nanoscale Materials (CNM) Users' Executive Committee (2016-19). He holds a Ph.D. in Materials from the University of

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