Special Quasirandom Structure Approach to Studying Perovskite Solid Solutions

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Perovskite compounds, ABO_3, that have novel, multiferroic properties have increasingly complex compositions, with elemental mixing on both the A and B cation sites. Although it is desirable to use theoretical methods to predict the structures and properties, these complex compounds pose a unique challenge to computational studies. Here we demonstrate using special quasirandom structures (SQS) to predict the propertied of perovskite solid-solutions.

Using a 40-atom SQS the local structure and bonding within $K_{0.5}Na_{0.5}NbO_3$ is studied. To validate the method, a $K_{0.5}Na_{0.5}NbO_3$ specimen is synthesized and its pair distribution function (PDF) is determined by neutron diffraction. A simulated neutron diffraction PDF is determined and compared to the experimental results. Also a 40-atom rock-salt supercell (RSS), which is commonly used for theoretical calculations, is examined. The SQS predicted and experimentally determined PDFs agree out to 12 Å. The RSS, which is commonly found in the literature, deviates from the experimental results at distances as small as 4 Å. This demonstrates that the SQS is significantly more accurate than the existing calculations based on RSS.

The SQS approach is further validated by comparing SQS computed results to those computed using large (8×8×8) supercells to approximate a true random system. Well-characterized core shell atomic potentials are used to represent $Ba_xSr_{1-x}TiO_3$. To demonstrate the impact of system selection on the dynamical properties of perovskite oxides the phonon density of states is predicted for SQSs of varying quality, short-ranged ordered systems, e.g., the RSS, and the true random system. Three compositions are selected for testing.