Finite-Temperature Properties of Ferroelectric Alloys from First-Principles [1]

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A first-principles-derived approach is developed to study finite-temperature properties of perovskite solid solutions. This technique is based on the perturbation of an effective Hamiltonian of a virtual crystal by the alloy configuration. This approach is applied to the homovalent $Pb(Zr_{1-x}Ti_x)O_3$ (PZT) solid solution with x close to 0.5, and to the heterovalent $Pb(Sc_{0.5}Nb_{0.5})O_3$ (PSN) alloy. Structural and piezoelectric predictions are in excellent agreement with experimental data and direct first-principles results for both systems. In particular, the recently discovered low-temperature monoclinic phase is confirmed to exist in PZT, and is demonstrated to act as a bridge between the well-known tetragonal and rhombohedral phases. This first-principles-derived tool also provides a successful explanation to the large d_{33} piezoelectric coefficients found in $Pb(Zr_{0.5}Ti_{0.5})O_3$ ceramics and along the pseudo-cubic direction of rhombohedral heterovalent alloys. Other possibilities offered by this new approach will also be discussed.

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