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Predicting polarization and nonlinear dielectric response of arbitrary perovskite superlattice sequences

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A complete theory of epitaxial perovskite superlattices requires an understanding both of epitaxial strain effects and of electrostatic boundary conditions. Here, focusing on the latter issue, we[1] have carried out first-principles calculations of the nonlinear dielectric properties of short-period "bicolor" and "tricolor" $CaTiO_3$/$SrTiO_3$/$BaTiO_3$ superlattices having the in-plane lattice constant of $SrTiO_3$. In particular, we have calculated the layer polarizations p_j as defined using the Wannier-based method of Wu, Diéguez, Rabe and Vanderbilt[2] for each neutral BaO, SrO, CaO, or TiO_2 layer. We use a cluster expansion (CE) technique to model the layer polarizations p_j of a selected set of bicolor superlattices as a function of the displacement field D (which is uniform throughout the insulating superlattice), the chemical identity of the layer itself, and the chemical identity of its neighboring layers. We find that p_j is a strongly localized function of its chemical environments at fixed D field, i.e., the dependence on the identity of the neighboring layers decays rapidly with distance. This localized property enables us to arrive at a truncated and simplified CE model which can accurately predict $p_j(D)$ in arbitrary layer sequences, both bicolor and tricolor. A similar approach is used to model the dependence of the c lattice constant. With all this information in hand, we can predict the polarization, piezoelectric and nonlinear dielectric response of arbitrary superlattice sequences. The power of the approach is demonstrated by showing that a model fitted only to calculations on inversion-symmetric bi-color superlattices can successfully predict the inversion symmetry breaking in tricolor superlattices such as $2SrTiO_3$/$1BaTiO_3$/$1CaTiO_3$.

[1]In collaboration with Massimiliano Stengel, Karin M. Rabe and David Vanderbilt.
[2]X. Wu, O. Diéguez, K. Rabe and D. Vanderbilt, Phys. Rev. Lett. **97**, 107602 (2006).