Ferroelectric switching in Aurivillius phase oxides from first principles

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Ferroelectricity where the polarization is induced by a trilinear coupling to two non-polar lattice distortions has been discovered in layered perovskite oxides such as the Ruddlesden-Popper phases in recent years. Since non-polar lattice distortions (such as octahedral rotations) often couple to the electronic and magnetic degrees of freedom in a material, these ferroelectrics may offer new pathways for electric field control of electronic/magnetic degrees of freedom. In addition, the coupling between non-polar lattice distortions and polarization has important implications for the ferroelectric switching mechanism in these materials: when the polarization reverses, one (but not both) of the non-polar lattice distortions must reverse. First-principles calculations and experiments have revealed a complex ferroelectric switching mechanism in the Ruddlesden-Popper layered perovskites, where the polarization reverses via multiple two-step switching paths.

Another family of layered perovskite-like oxides with a trilinear coupling between polarization and two non-polar lattice distortions is the Aurivillius family. Aurivillius phase oxides have the general formula $Bi_{2m}A_{n-m}B_nO_{3(n+m)}$, and are well known as ferroelectrics that are resistant to fatigue and have low coercive fields. One of the most studied Aurivillius phase oxides is $SrBi_2Ta_2O_9$; its crystal structure consists of alternately stacked Bi_2O_2 slabs and blocks consisting of two perovskite-like layers. Theoretical and first-principles studies during the past decade have revealed that the coupling between the non-polar lattice distortions and the polarization is key for understanding both the ferroelectric ground state of $SrBi_2Ta_2O_9$ as well as its sequence of structural phase transitions. However, the implications of this coupling on the ferroelectric switching mechanism of $SrBi_2Ta_2O_9$ (and related compounds) has to our knowledge not been investigated.

Here, we use a combination of group theoretic analysis and density functional theory (DFT) calculations to explore and calculate the energy barriers of low-energy ferroelectric switching pathways of $SrBi_2Ta_2O_9$. By tracking the evolution of the non-polar lattice distortions as well as the polarization along each switching path, we uncover how the couplings between multiple lattice distortions facilitate low switching barriers. This work provides new understanding of the role of non-linear couplings between lattice distortions in the ferroelectric switching mechanisms of the structurally complex family of Aurivillius phase oxides.