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Band Gap and Edge Engineering of $SrTiO_3$ and Related Compounds via Ferroic Distortion and Anisotropic Strain

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Due to its electronic band edge energies and its stability in water, the perovskite strontium titanate ($SrTiO_3$) is a promising water splitting photocatalyst. However, its ability to use solar photons to split water into hydrogen and oxygen would be more efficient if its wide optical band gap (3.2 eV) better matched the solar spectrum. Therefore, there is interest in modifying the crystal structure of $SrTiO_3$ (e.g., via chemical doping or epitaxial strain) to tune its electronic and optical properties. We use density functional theory (DFT) and many-body perturbation theory within the GW approximation to calculate the effects of structural and chemical modifications of $SrTiO_3$ on its band gap and edges. Much of our work (Berger, Fennie, and Neaton, Phys. Rev. Lett. 107, 146804 [2011]) focuses on the effects of epitaxial strain and the associated ferroic distortions. Anisotropic strains are shown to reduce the $SrTiO_3$ gap by breaking degeneracies at the band edges. Ferroic distortions are shown to widen the gap by allowing new band edge orbital mixings. To reduce the $SrTiO_3$ gap, one must lower the symmetry from cubic while suppressing ferroic distortions. Our calculations indicate that for engineered orientation of the growth direction along [111], the $SrTiO_3$ gap can be controllably and considerably reduced at room temperature. Chemical doping and substitution can, in combination with strain and distortion, further tune the band gap and edges. Our results and their favorable qualitative agreement with experiment suggest achievable paths toward engineering efficient solar water splitting catalysts, and more generally provide fundamental insight into the relationships between crystal and electronic structure in the property-rich perovskite family.