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Symmetry engineering of functionality in ultrathin layered oxides

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The breaking of spatial and temporal symmetries are ubiquitous concepts in condensed-matter science. Such symmetry breaking defines the appearance of technologically relevant effects such as piezoelectricity, photovoltaic effects, nonlinear optical properties and spin-transport phenomena. It also determines fundamental properties such as the electronic topology in quantum materials. Therefore, the creation of materials where symmetry can be set by design stands out as a versatile approach for controlling emergent phenomena. Here we show how engineering spatial inversion symmetry on a sub-unit-cell level in ultrathin layered oxide films leads to the controlled activation and deactivation of optical second harmonic generation as a prototypical inversion-symmetry-sensitive functionality. Combining this nonlinear optical detection with thin-film deposition, we further present a novel route for in-situ tracking of the symmetry state of our thin films directly during synthesis. Symmetry engineering on the sub-unit-cell level in layered oxides thus suggests a new platform for the controlled activation and deactivation of symmetry-governed functionalities in oxide-electronic epitaxial thin films.