

# Epitaxial Engineering of Emergent Phenomena in Tantalate Perovskites

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Epitaxial tuning knobs, including epitaxial strain, could serve as a powerful parameter that significantly alter the lattice symmetries, affect phase stability, and reshape the energy landscape. In this presentation I will discuss the epitaxial tuning of tantalate perovskites (KTaO<sub>3</sub> and EuTa<sub>2</sub>O<sub>6</sub>) grown using a sub-oxide molecular beam epitaxy method, which we recently developed for tantalates [1]. Next, I will talk about the effect of epitaxial tuning knobs, such as epitaxial strain, on the lattice and electronic structures. Here, I will discuss that KTaO<sub>3</sub>, a cubic perovskite, can be epitaxially strained into a highly tunable ferroelectric. KTaO<sub>3</sub> films, grown commensurate to SrTiO<sub>3</sub> (001), experience an in-plane compressive strain of -2.1 % that transforms the otherwise cubic structure into a tetragonal polar phase with a transition temperature of 475 K, consistent with our phase-field calculations. The Curie temperature and the spontaneous electric polarization are systematically controlled with epitaxial strain. Scanning transmission electron microscopy reveals cooperative polar displacements of the potassium columns with respect to neighboring tantalum columns at room temperature. Optical second harmonic anisotropic rotation results are described by a tetragonal polar point group ( $4mm$ ), indicating emergence of a global polar ground state.

Finally, I discuss our recent results on epitaxial control of ordering in fractionally occupied double perovskite, EuTa<sub>2</sub>O<sub>6</sub> [2]. The intrinsic crystal anisotropy of EuTa<sub>2</sub>O<sub>6</sub> plays a pivotal role, underscoring how targeted structural modifications can facilitate the emergence of novel quantum states. The crystal and electronic structures of EuTa<sub>2</sub>O<sub>6</sub> are investigated. X-ray diffraction and electron microscopy reveal the layered *A*-site ordering. Angle-resolved photoemission spectroscopy, along with density functional theory calculations, provide direct insight into the electronic structure, unveiling the potential for engineered confined states within bulk materials. These findings highlight EuTa<sub>2</sub>O<sub>6</sub> as a platform for studying 2D-like electronic phenomena in a 3D context, paving the way for novel device architectures.

[1] T. Schwaigert, S. Salmani-Rezaie, M. R Barone, H. Paik, E. Ray, M. D Williams, D. A Muller, D. G Schlom, K. Ahadi, *Journal of Vacuum Science & Technology A* **2**, 41(2023).

[2] T. Schwaigert, A. Barooni, B. Gregory, P. Malinowski, A. Tenneti, S. Hasko, B. Palazzolo, J. W Hodgson, B. Faeth, P. M Woodward, K. M Shen, A. Singer, M. Ghazisaeidi, S. Salmani-Rezaie, D. G Schlom, K. Ahadi, *Advanced Functional Materials* e13656 (2025)

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