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Detailed Structure of Hybrid Improper Ferroelectric $\text{Ca}_3\text{X}_2\text{O}_7$ Systems¹

TREVOR TYSON, New Jersey Institute of Technology

In hybrid improper ferroelectric systems, polarization arises from the onset of successive nonpolar lattice modes. Detailed measurements and modeling were performed to determine the spatial symmetries of the phases involved in the transitions to these modes. Structural and optical measurements reveal that the tilt and rotation distortions of the MnO_6 or TiO_6 polyhedra relative to the high symmetry phases driving ferroelectricity in the hybrid improper $\text{Ca}_3\text{X}_2\text{O}_7$ system (X=Mn and Ti) condense at different temperatures. The tilt angle vanishes abruptly at $T_T \sim 400$ K for $\text{Ca}_3\text{Mn}_2\text{O}_7$ (and continuously for X=Ti) and the rotation mode amplitude is suppressed at much higher temperatures $T_R \sim 1060$ K. Moreover, Raman measurements in $\text{Ca}_3\text{Mn}_2\text{O}_7$ under isotropic pressure reveal that the polyhedral tilts can be suppressed by very low pressures (between 1.4 and 2.3 GPa) indicating their softness. These results indicate that the $\text{Ca}_3\text{Mn}_2\text{O}_7$ system provides a new platform for strain engineering of ferroelectric properties in film-based systems with substrate-induced strain. Collaborators: S. Liu, H. Zhang, S. Ghose, M. Balasubramanian, Zhenxian Liu, S. G. Wang, Y-S. Chen, B. Gao, J. Kim, and S.-W. Cheong

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