# Photoinduced dynamics of ordering in polar vortex supercrystals

***V.A. Stoica*[1,2*], *T.Yang*[1], *S. Das*[3], *Y. Cao*[4], *H. Wang*[1], *H. Padmanabhan*[1], *C. Dai*[1], *Y. Kubota*[5,6], *D. Zhu*[7], *Y. Sato*[8], *A. Mangu*[9], *Q.L. Nguyen*[7], *Z. Zhan*[2], *S. Owada*[5,6], *K. Miyanishi*[5,6], *K. Tamasaku*[5,6], *T. Sato*[7], *J.M. Glownia*[7], *V. Esposito*[7], *S. Nelson*[7], *S. Song*[7], *Y. Sun*[7], *J.J. Turner*[7], *M.C. Hoffman*[7], *R.D. Schaller*[10], *A.M. Lindenberg*[9], *L.W. Martin*[3], *R. Ramesh*[3], *I. Matsuda*[8], *L.Q. Chen*[1], *H. Wen*[2], *V. Gopalan*[1], and *J.W. Freeland*[2]**

[1]*Dept. of Materials Science and Engineering, Pennsylvania State University, University Park, 16802, USA*
[2]*Advanced Photon Source, Argonne National Laboratory, Argonne, 60439, USA*
[3]*Department of Materials Science and Engineering, University of California, Berkeley, 94729, USA*
[4]*Materials Science Division, Argonne National Laboratory, Argonne, 60439, USA*
[5]*Japan Synchrotron Radiation Research Inst., 1-1-1 Kouto, Sayo-cho, Sayo-gun, Hyogo 679-5198, Japan*
[6]*RIKEN SPring-8 Center, 1-1-1 Kouto, Sayo-cho, Sayo-gun, Hyogo 679-5148, Japan*
[7]*Linac Coherent Light Source, SLAC National Accelerator Laboratory, Menlo Park, 94025, USA*
[8]*Institute for Solid State Physics, The University of Tokyo, Kashiwa, Chiba 277-8581, Japan*
[9]*Department of Materials Science and Engineering, Stanford University, Stanford, 94305, USA*
[10]*Center for Nanoscale Materials, Argonne National Laboratory, Lemont, 60439, USA*
*E-mail: vxs30@psu.edu

Multiple order parameters can coexist in complex materials, whereas their cooperativity or competition can be difficult to disentangle under equilibrium conditions. Photoinduced transitions (PIPT) can be used to directly accesss multiple coexisting orders during the excitation and relaxation processes. Two significant challenges remain in this area of research: a) creation of novel orders and b) stimulation of persistent phases to amend the equilibrium phase diagrams. Of particular interest is the prospect of controlling topological defects in nano-materials that can be written and erased on demand using ultrafast external stimuli. A novel class of topological defects has recently emerged in nanoscale ferroelectrics including vortices, skyrmions, merons and vortex supercrystals, which present emerging properties: collective dynamics, negative capacitance, chirality and topological transformations. Furthermore, the inherent multiplicity of the periodic arrangements formed from these topological defects opens the avenue to explore order-disorder transformations in these systems.
We previously demonstrated that femtosecond optical pulses can be used to convert disordered-like phase mixtures into vortex supercrystals with long range order that remain stable at room temperature until heating is applied for restoring the ground state [1]. Here, we will present XFEL and synchrotron measurements of vortex supercrystal ordering and its mechanisms. The real-time evolution of the photoinduced irreversible transformation is resolved by single-shot XFEL measurements. The excitation dependency of the ground state collapse into polar disorder on fs-ps timescale is subsequently followed by ns nucleation of different types of topological defects, which ultimately transform into supercrystals at the longer timescales. Dynamical phase field modeling [2] is employed to visualize the evolution of 3D structure relative to the experiments, including the evolution of polar, strain and charge order of the system. The underlying trigger of this persistent PIPT mechanism is correlated with transient charging and modification of the internal electrostatic field, which is followed up by a much slower thermal relaxation on > 100 ns, which greatly strengthens the supercrystal polar distortions. Moreover, we will present the ultrafast access to reversible supercrystal ordering while using smaller excitation strengths, which reveals the synergy between electronic and phononic excitations that stabilizes the supercrystal ordering. These results captured different regimes of dynamical transformation of topological defects, suggesting new perspectives in exploring related ultrafast phenomena.

**References:**

[1] V.A. Stoica et al., Nat. Mater. 18, 377–383 (2019).
[2] T. Yang et al., Phys. Rev. Lett. 124, 107601 (2020).