Supplemental Material: Optical second harmonic signature of phase coexistence in ferroelectric|dielectric heterostructures

Nives Strkalj[1],* Amadé Bortis[1], Marco Campanini[2],
Marta D. Rossell[2], Manfred Fiebig[1], and Morgan Trassin[1]
[1] *Department of Materials, ETH Zurich, 8093 Zurich, Switzerland*
[2] *Electron Microscopy Center, Swiss Federal Laboratories for Materials Science and Technology, Empa, Dübendorf, 8600, Switzerland*

This supplemental contains time-dependent RHEED intensity during PLD, topography, lateral PFM response of the as-grown state and XRD characterization of the phase coexistence in the PTO|STO superlattice on SRO-buffered DSO. It further includes SHG polarimetry of uncapped PTO layer and PTO|STO|PTO multilayer.

I. GROWTH MONITORING AND CHARACTERIZATION OF THE PTO|STO SUPERLATICE

The time-dependent RHEED intensity for the growth of the PTO and STO layers, and surface topography of the finalized $(PTO_{12}|STO_{12})_8PTO_{12}$ superlattice are shown in Supplementary Fig. 1. Image of lateral PFM response in Supplementary Fig. 2(a) shows that the a_1/a_2-phase and the vortex phase are separated in stripes with a width of about 500 nm along [010], and a length of about 5 µm along [100], as previously reported for the system [1]. Idealized sketch of the domain configuration is provided in Supplementary Fig. 2(b).

Reciprocal space maps (RSM) in Supplementary Fig. 3 show the diffuse scattering corresponding to phase coexistence of the a_1/a_2-phase and the vortex phase in the PTO|STO superlattice. Diffuse scattering for the DSO 002 reflection in Supplementary Fig. 3(a) corresponds to the size of a vortex-antivortex pair of 10 nm along the [010] direction, in agreement with the STEM analysis in Fig. 2. The reciprocal space map for the DSO 113 reflection in Supplementary Fig. 3(b) shows that the overall structure is coherently tensile-strained to the DSO substrate and that an in-plane periodicity of 20 nm appears along ⟨110⟩ directions.

II. SHG POLARIMETRY OF UNCAPPED PTO LAYER AND PTO|STO|PTO MULTILAYER

The polar dependence of the SHG intensity at fixed angles of β for a uncapped PTO layer and a PTO|STO|PTO multilayer is shown in Supplementary Fig. 4. The SHG response of the uncapped PTO layer is well-described with the one-phase model indicating absence of the vortex phase. The SHG response of the PTO|STO|PTO multilayer is very weak and therefore difficult to model. Fit of the two-phase model shows better matching with the SHG data of

* ns851@cam.ac.uk

the multilayer than the fit of the one-phase model.

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[1] A. R. Damodaran, J. D. Clarkson, Z. Hong, H. Liu, A. K. Yadav, C. T. Nelson, S.-L. Hsu, M. R. McCarter, K.-D. Park, V. Kravtsov, A. Farhan, Y. Dong, Z. Cai, H. Zhou, P. Aguado-Puente, P. Garcia-Fernandez, J. Iñiguez, J. Junquera, A. Scholl, M. B. Raschke, L.-Q. Chen, D. D. Fong, R. Ramesh, and L. W. Martin, Phase coexistence and electric-field control of toroidal order in oxide superlattices, Nat. Mater. **16**, 1003 (2017).

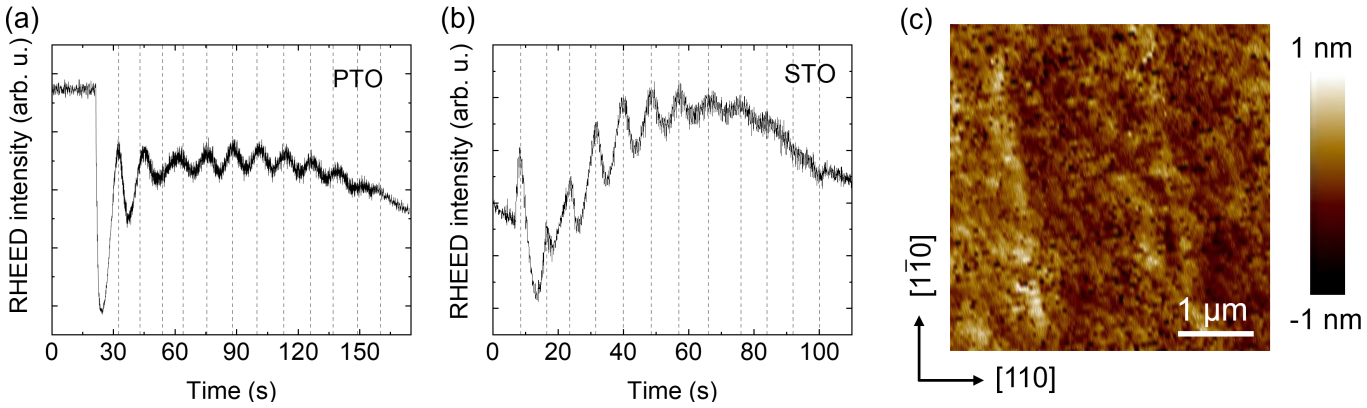

FIG. 1. (a,b) Time-dependent RHEED monitoring of the PTO and STO layers in the PTO|STO superlattice. (c) Topography image.

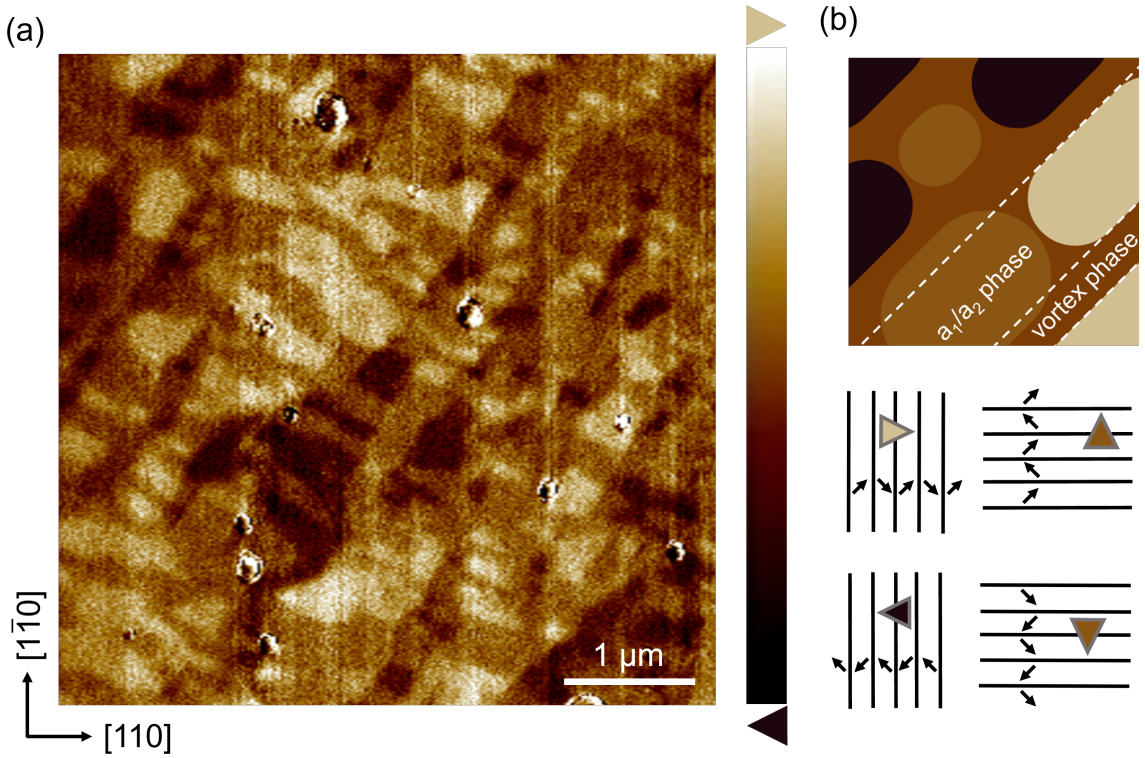

FIG. 2. (a) Image of the lateral PFM response. (b) Idealized sketch of the polarization direction in a sample with phase coexistence of the a_1/a_2-phase and the vortex phase (top). Polarization direction is indicated by the color and direction of triangles in the schematics (bottom). In the a_1/a_2-phase, superdomains with a net polarization along [110] and [1$\bar{1}$0] are observed because the individual a_1- and a_2-domains have a lateral size below the resolution limit of the PFM method.

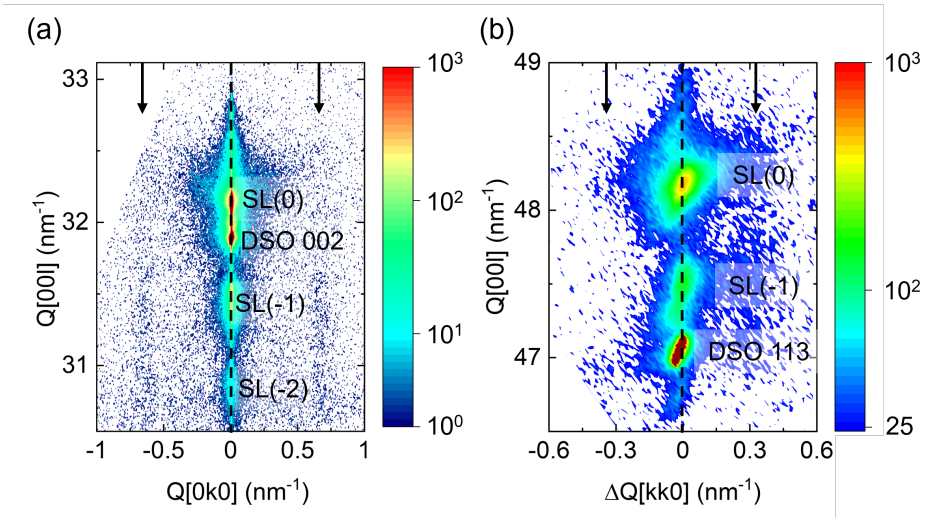

FIG. 3. RSM for (a) 002 and (b) 113 reflections of the DSO substrate. Arrows indicate the positions of the diffuse scattering corresponding to (a) periodicity of vortex-antivortex pairs of 10 nm in the vortex phase and (b) domain-wall periodicity of 20 nm in the a_1/a_2-phase.

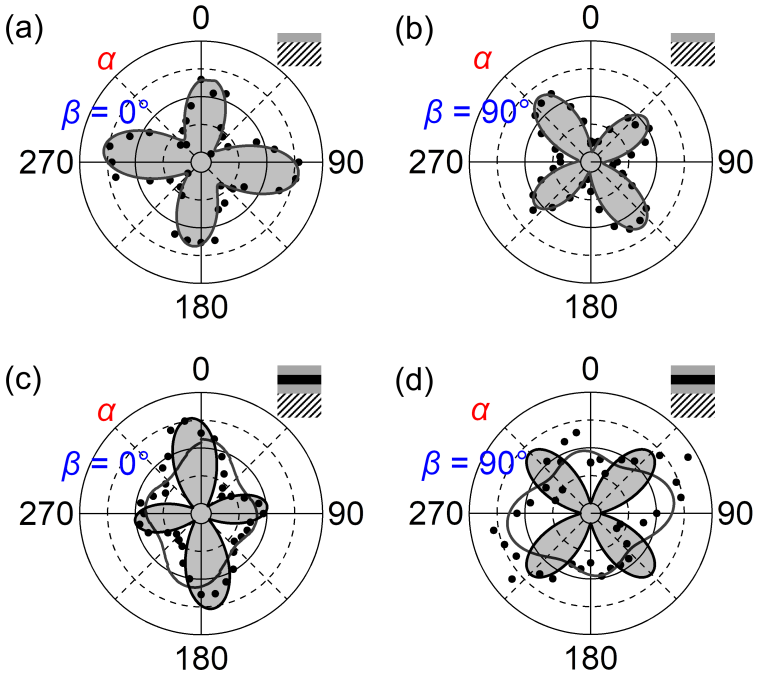

FIG. 4. (a-d) Polar plots of SHG intensity (points) versus the incident-light polarization angle α for (a,b) a uncapped PTO layer and (c,d) a PTO|STO|PTO multilayer for (a,c) $\beta = 0°$ and (b,d) $\beta = 90°$. Grey lines are fits of the one-phase model and black lines are fits of the two-phase model. A schematic of the heterostructure is given as inset on the top right. Grey denotes PTO, black denotes STO, and upward hatching denotes the SRO-buffered DSO substrate. The intensity scale is normalized for each sample.