

SUPPLEMENTARY MATERIAL TO:

A low-temperature route for producing epitaxial perovskite superlattices on (001)-oriented SrTiO₃/Si substrates

Aleksandr V. Plokhikh,^{1*} Iryna S. Golovina,^{1*} Matthias Falmbigl,¹ Igor A. Karateev,² Alexander L. Vasiliev,³ Jason Lapano,⁴ Roman Engel-Herbert,^{4,5,6} and Jonathan E. Spanier^{1,7,8}

¹*Department of Materials Science & Engineering, Drexel University, Philadelphia, Pennsylvania 19104, USA*

²*National Research Center “Kurchatov Institute”, Kurchatov Square 1, Moscow 123182, Russia*

³*Moscow Institute of Physics and Technology (State University), MIPT, 9 Institutskiy per., Dolgoprudny, 141701 Moscow Region, Russia*

⁴*Department of Materials Science & Engineering, Pennsylvania State University, University Park, Pennsylvania 16802, USA*

⁵*Department of Chemistry, Pennsylvania State University, University Park, Pennsylvania 16802, USA*

⁶*Department of Physics, Pennsylvania State University, University Park, Pennsylvania 16802, USA*

⁷*Department of Mechanical Engineering and Mechanics, Drexel University, Philadelphia, Pennsylvania 19104, USA*

⁸*Department of Physics, Drexel University, Philadelphia, Pennsylvania 19104, USA*

* Authors with equal contribution.

An analysis of the (BaTiO₃)_m/(SrTiO₃)_n superlattice structure peaks reveals a systematic change of the superlattice repeat unit thickness with a change in the deposition sequence. The slope for an increasing number of BaTiO₃-layers from $m/n = 1/5, 3/5$ to $5/5$ (BaTiO₃/SrTiO₃ cycles), and vice versa for the SrTiO₃-layers reveals slopes of 4.08 nm and 3.89 nm, respectively, which are both close to the expected unit cell thicknesses of the individual constituents (see Fig. S1). This observation indicates a minor intermixing of Ba and Sr in the individual layers.

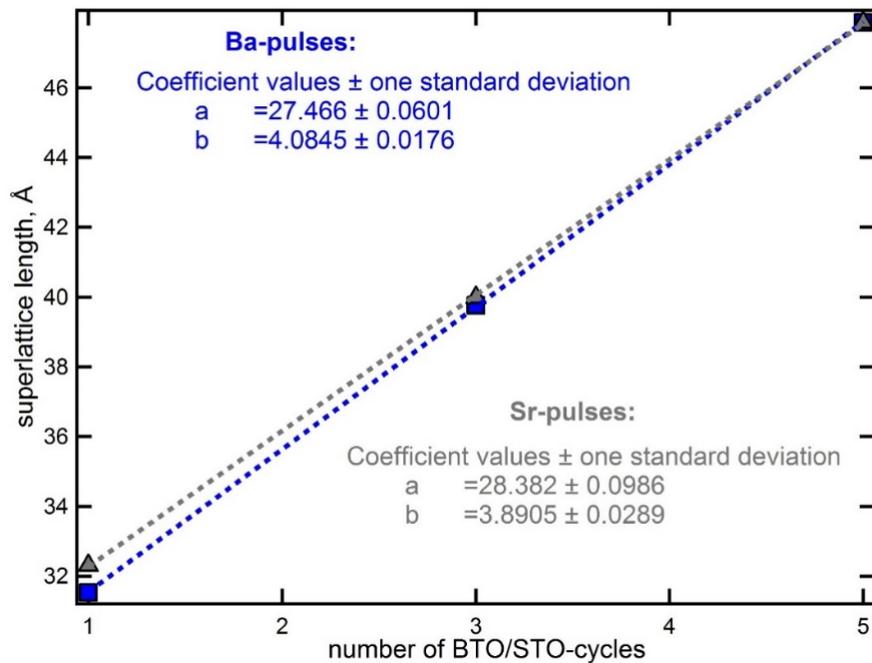


Figure S1. Superlattice structure repeat unit thickness as a function of increasing BaTiO₃ or SrTiO₃ cycles for a constant number of 5 repeat cycles for the other constituent.

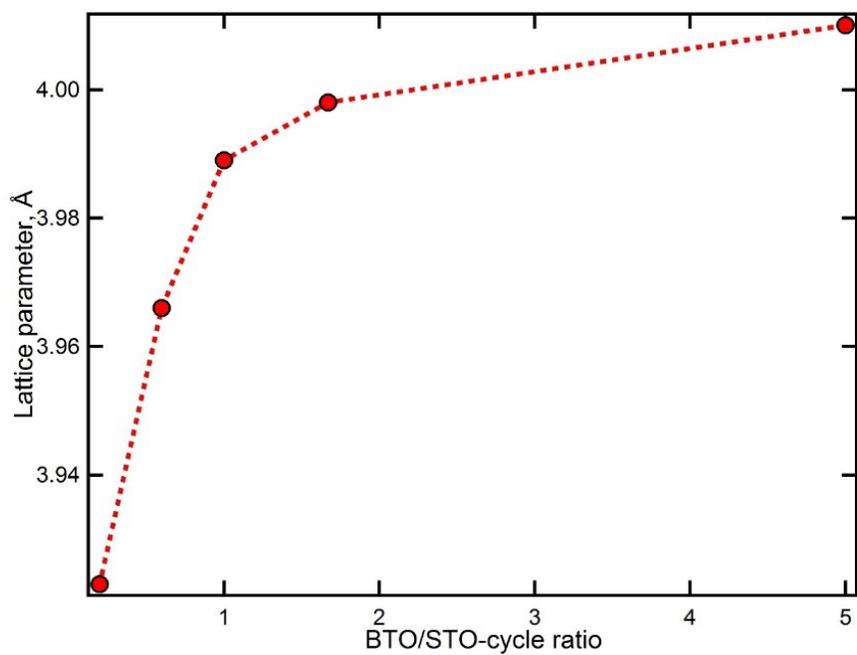


Figure S2. The average *c*-lattice parameters of the superlattices obtained from XRD data as a function of increasing BaTiO₃/SrTiO₃-cycle ratio.

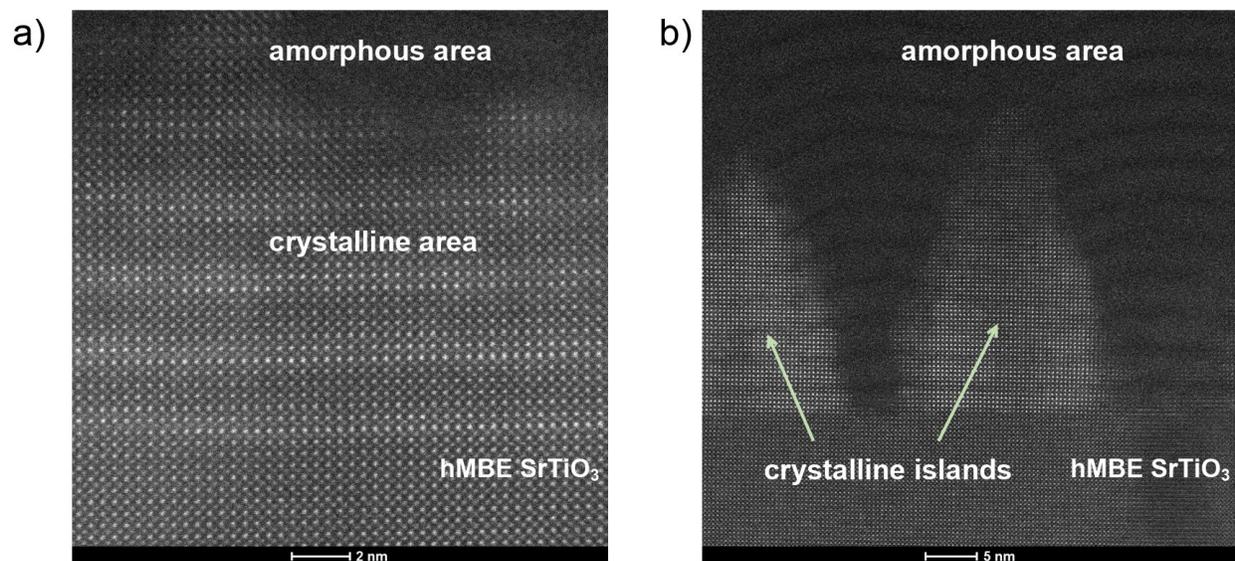


Figure S3. HAADF STEM images of **a)** $(\text{BaTiO}_3)_1/(\text{SrTiO}_3)_5$ and **b)** $(\text{BaTiO}_3)_5/(\text{SrTiO}_3)_1$ superlattices with a total number of 10 repeats.

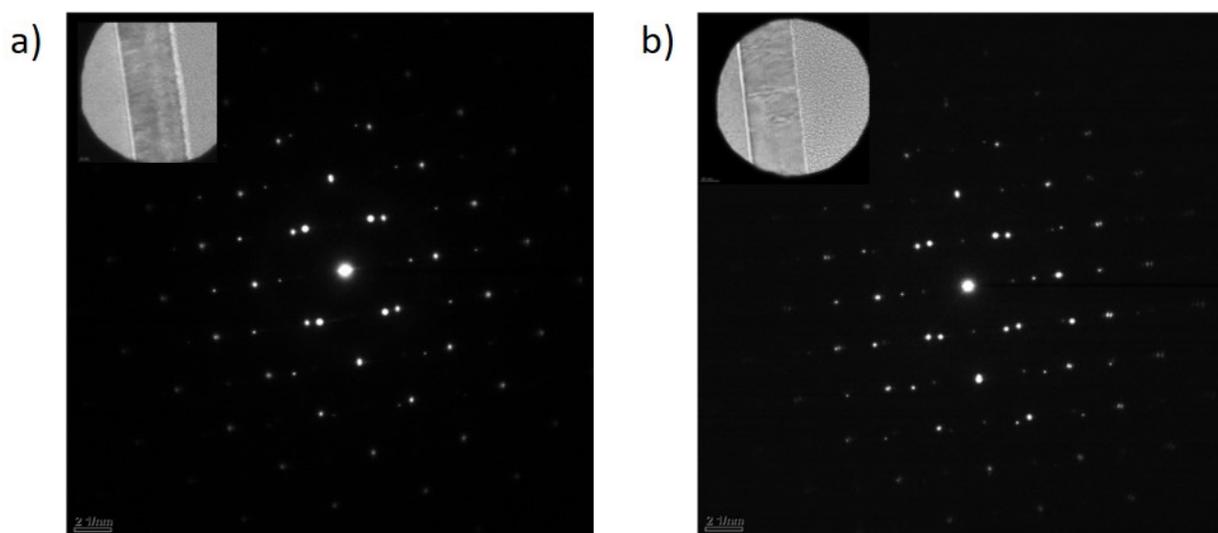


Figure S4. SAED image taken on the $(\text{BaTiO}_3)_3/(\text{SrTiO}_3)_5$ **(a)** and $(\text{BaTiO}_3)_5/(\text{SrTiO}_3)_3$ **(b)** superlattices.

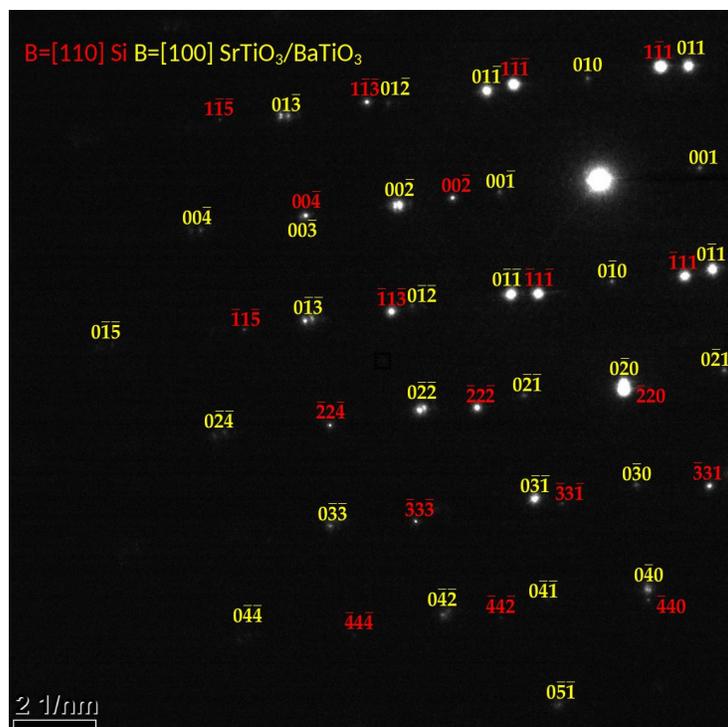


Figure S5. Indexed SAED image (1/4 part) taken on a $(\text{BaTiO}_3)_5/(\text{SrTiO}_3)_3$ superlattice.

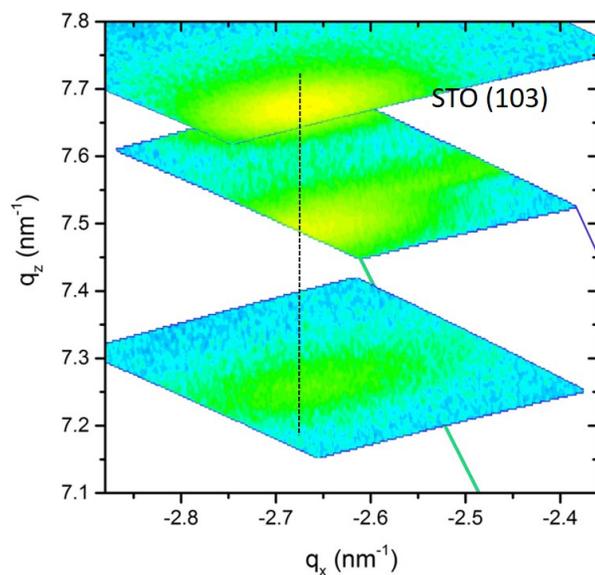


Figure S6. RSM of an asymmetric scan around the (103) peak of the *h*MBE SrTiO_3 -substrate layer. The dashed line indicates the epitaxial strain of the superlattice structure to the in-plane lattice dimensions of the SrTiO_3 -substrate layer.