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Supporting Information for:

Induced Ferroelectric Phases in $SrTiO_3$ by a Nanocomposite Approach

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Phase field simulation

Phase-field modeling was performed using a self-developed module within the framework of a commercial finite-element method (FEM) software (Comsol Multiphysics v5.2). The free-energy functional F derived for a [001]-oriented $SrTiO_3$ thin film has the following form:

$$F = \alpha_{ij} P_i P_j + \alpha_{ijkl} P_i P_j P_k P_l + \frac{1}{2} g_{ijkl} P_{i,j} P_{k,l} + \frac{1}{2} c_{ijkl} \varepsilon_{ij} \varepsilon_{kl} - q_{ijkl} \varepsilon_{ij} P_k P_l - \frac{1}{2} \kappa_0 E_i E_i - E_i P_i \quad (1)$$

where all repeating subscripts imply summation over the Cartesian coordinate components x_i (i = 1, 2 and 3) and '$,i$' denotes the partial derivative operator with respect to x_i (that is, $\partial / \partial x_i$); P_i is a component of the polarization vector **P**, ε_{ij} mechanical strain and E_i electric field; α's are the Landau expansion coefficients (up to the fourth-order for STO here), g_{ijkl} the polarization gradient coefficients related to the energy cost for domain wall formation, c_{ijkl} the elastic stiffness tensor, q_{ijkl} the electrostrictive coefficients and κ_0 the background dielectric permittivity. The material coefficients and related model parameters used are provided in Supplementary Table 1. Note that for the present model, we have neglected the weak coupling between the polarization and structural order parameters of STO at room temperature; the latter is related to the antiferrodistortive phase transition of STO which occurs at a much lower temperature (~105 K for the bulk phase).

The whole film was assumed to be homoepitaxially constrained with zero misfit strain (neglecting the experiment value of -0.0038). The displacements of the bottom surface were set to conform to the substrate while the top surface was set mechanically free. Short-circuit electrical boundary conditions were applied for both top and bottom surfaces. A typical model size was 70 × 70 × 60 nm^3 (*xyz*), which was discretized 0.7×0.7×2 nm^3 second-order cuboid elements. A 65 × 65 nm^2 region randomly selected from the HAADF-STEM images was binarized and then used to designate the *xy*-locations of MgO pillars with constant cross-sections across the thickness; note

that such smaller region than the lateral size of the model was chose to facilitate the application of in-plane (*xy*) continuity periodic boundary conditions. An initial out-of-plane strain of 0.0786 was assigned to the MgO pillars due to the lattice parameter difference between bulk MgO and STO (assuming no strain relaxation along the vertical interface). The elastic parameters of MgO were set for the pillars.

The spatio-temporal evolution of the STO:MgO nanocomposite system's field variables is governed by the time-dependent LGD polarization order parameter equation coupled with the continuum mechanical equilibrium equation and Poisson's equation of electrostatics:

$$\frac{\partial P_i(\boldsymbol{x},t)}{\partial t} = -L\frac{\delta F}{\delta P_i(\boldsymbol{x},t)} \tag{2}$$

$$\sigma_{ij,j} = \left(\partial F/\partial \varepsilon_{ij}\right)_{,j} = 0 \tag{3}$$

$$D_{i,i} = \left(\partial F/\partial E_i\right)_{,i} = 0 \tag{4}$$

in which t is time and L the kinetic coefficient related to the domain wall mobility; σ_{ij} and D_i are mechanical stress and electric displacement. An initial polarization state in the STO consisting of small random values (< 0.001 C/m^2) was assumed, and quasi-steady state solutions of the system were obtained using the time-dependent solver.

Table S1 Parameters and energy coefficients of $SrTiO_3$/MgO used in the phase-field simulation.

Coefficients	Value	Units	Coefficients	Value	Units
α_1	1.83×10^{8}	$C^{-2}\,m^{2}\,N$	Q_{11}	0.0458	$C^{-2}\,m^{4}$
α_{11}	1.70×10^{9}	$C^{-4}\,m^{6}\,N$	Q_{12}	-0.0135	$C^{-2}\,m^{4}$
α_{12}	1.37×10^{9}	$C^{-4}\,m^{6}\,N$	Q_{44}	0.0096	$C^{-2}\,m^{4}$
${c_{11}}^{STO}$	335	GPa	g_{11}	1.0×10^{-11}	$J\,m^{3}\,C^{-2}$
${c_{12}}^{STO}$	100	GPa	g_{12}	0.5×10^{-11}	$J\,m^{3}\,C^{-2}$
${c_{44}}^{STO}$	125	GPa	g_{44}	0.5×10^{-11}	$J\,m^{3}\,C^{-2}$
${c_{11}}^{MgO}$	300	GPa	κ_r^b	20	1
${c_{12}}^{MgO}$	100	GPa			
${c_{44}}^{MgO}$	155	GPa			