

## Supplementary Materials

**Micro-electromechanical system-based cryogenic and heating *in situ* transmission electron microscopy for investigating phase transitions and domain evolution in single-crystal BaTiO<sub>3</sub>**

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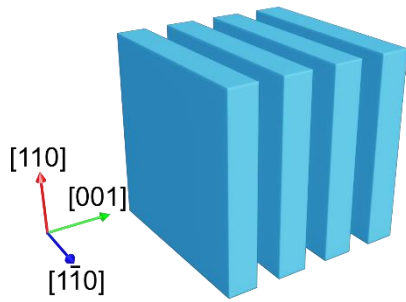
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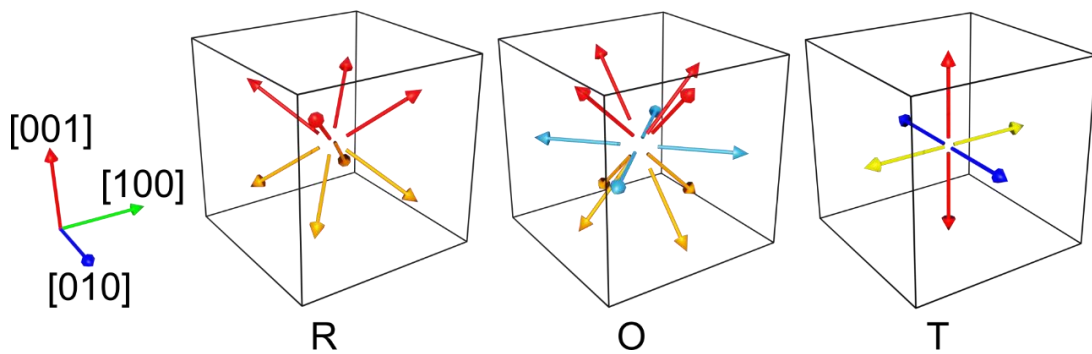
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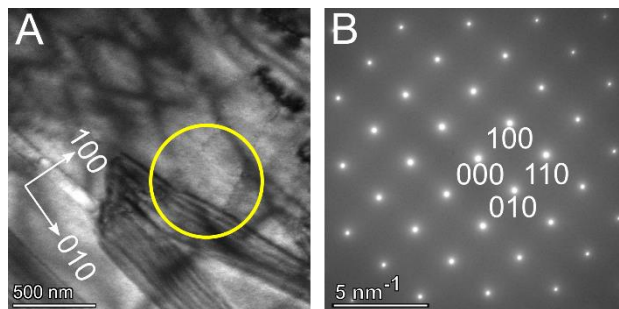
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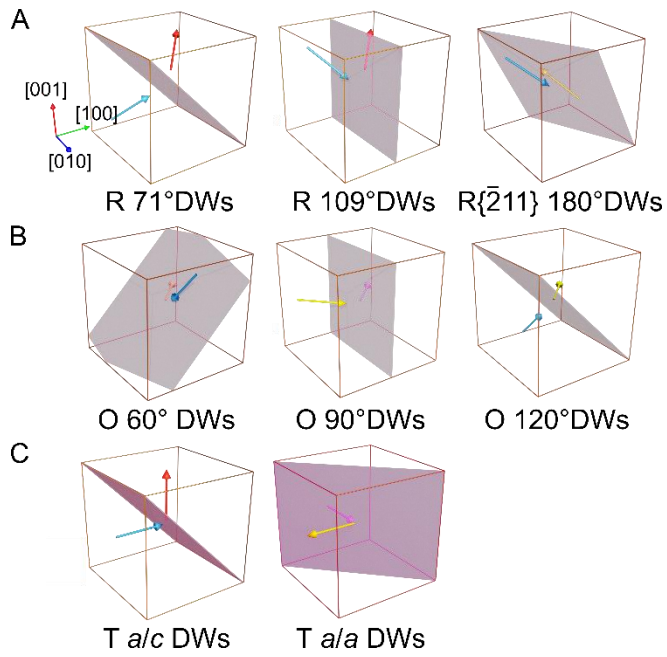
**Supplementary Figure 1.** Schematic of FIB-prepared TEM specimen along the (001) plane of [110]-orientated single-crystalline BTO.



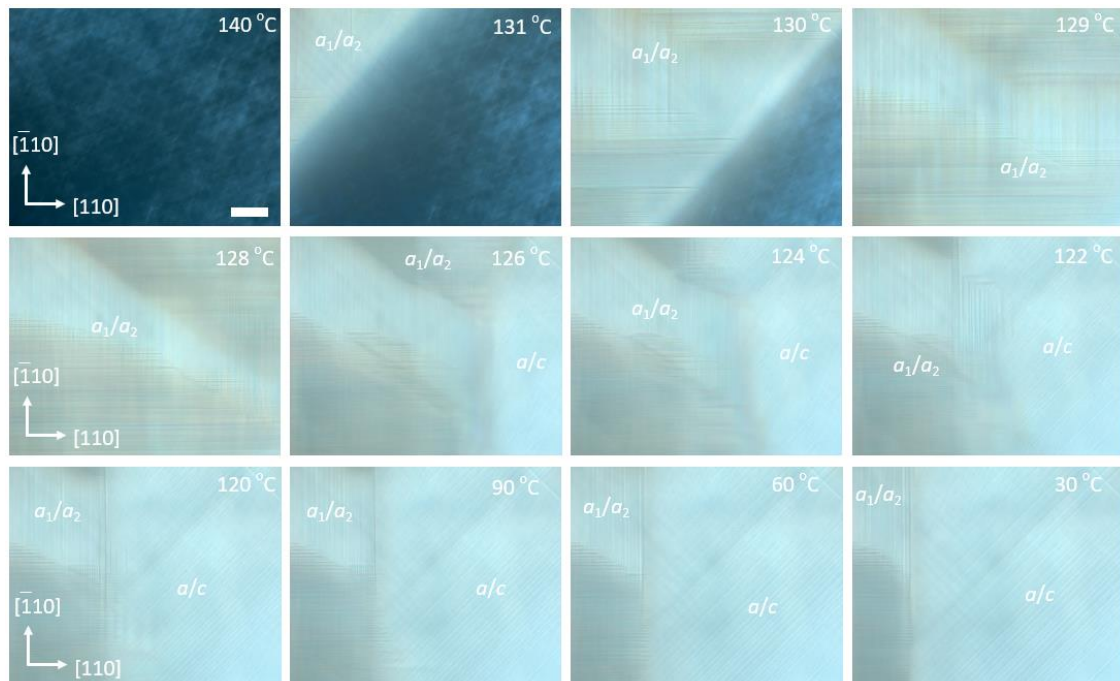
**Supplementary Figure 2.** Spontaneous polarizations in rhombohedral (denoted as R), orthogonal (O), and tetragonal (T) phases of  $\text{BaTiO}_3$ , respectively.



**Supplementary Figure 3.** (A) BF-TEM image obtained at  $-160\text{ }^\circ\text{C}$ . (B) SAED pattern corresponds to the area highlighted by the yellow circle in (A). Scale bars are (A) 500 nm and (B)  $5\text{ nm}^{-1}$ , respectively.



**Supplementary Figure 4.** The observed domain wall configurations in (A) rhombohedral (denoted as R), (B) orthogonal (O), and (C) tetragonal (T) phases of BaTiO<sub>3</sub>, respectively.



**Supplementary Figure 5.** Optical images obtained from the cooling process at different temperatures. The scale bar is 50  $\mu\text{m}$ .

**Supplementary Table 1. Material parameters of BaTiO<sub>3</sub> for bulk free energy calculation**

Parameters	Symbol	Value	Ref.
Landau-Devonshire potential coefficients	$\alpha_1$	$4.124 \times 10^5 (T - T_c)$ $\text{Nm}^2/\text{C}^2$	[1]
	$\alpha_{11}$	$-2.097 \times 10^8 \text{ Nm}^6/\text{C}^4$	
	$\alpha_{12}$	$7.974 \times 10^8 \text{ Nm}^6/\text{C}^4$	
	$\alpha_{111}$	$1.294 \times 10^9 \text{ Nm}^{10}/\text{C}^6$	
	$\alpha_{112}$	$-1.950 \times 10^9 \text{ Nm}^{10}/\text{C}^6$	
	$\alpha_{123}$	$-2.500 \times 10^9 \text{ Nm}^{10}/\text{C}^6$	
	$\alpha_{1111}$	$3.863 \times 10^{10} \text{ Nm}^{14}/\text{C}^8$	
	$\alpha_{1112}$	$2.529 \times 10^{10} \text{ Nm}^{14}/\text{C}^8$	
	$\alpha_{1122}$	$1.637 \times 10^{10} \text{ Nm}^{14}/\text{C}^8$	
	$\alpha_{1123}$	$1.367 \times 10^{10} \text{ Nm}^{14}/\text{C}^8$	

## REFERENCES

1. Li YL, Cross LE, Chen LQ, A phenomenological thermodynamic potential for BaTiO<sub>3</sub> single crystals. *J Appl Phys* 2005; 98: 064101.