

# Highly controllable and stable quantized conductance and resistive switching mechanism in single-crystal TiO<sub>2</sub> resistive memory on silicon

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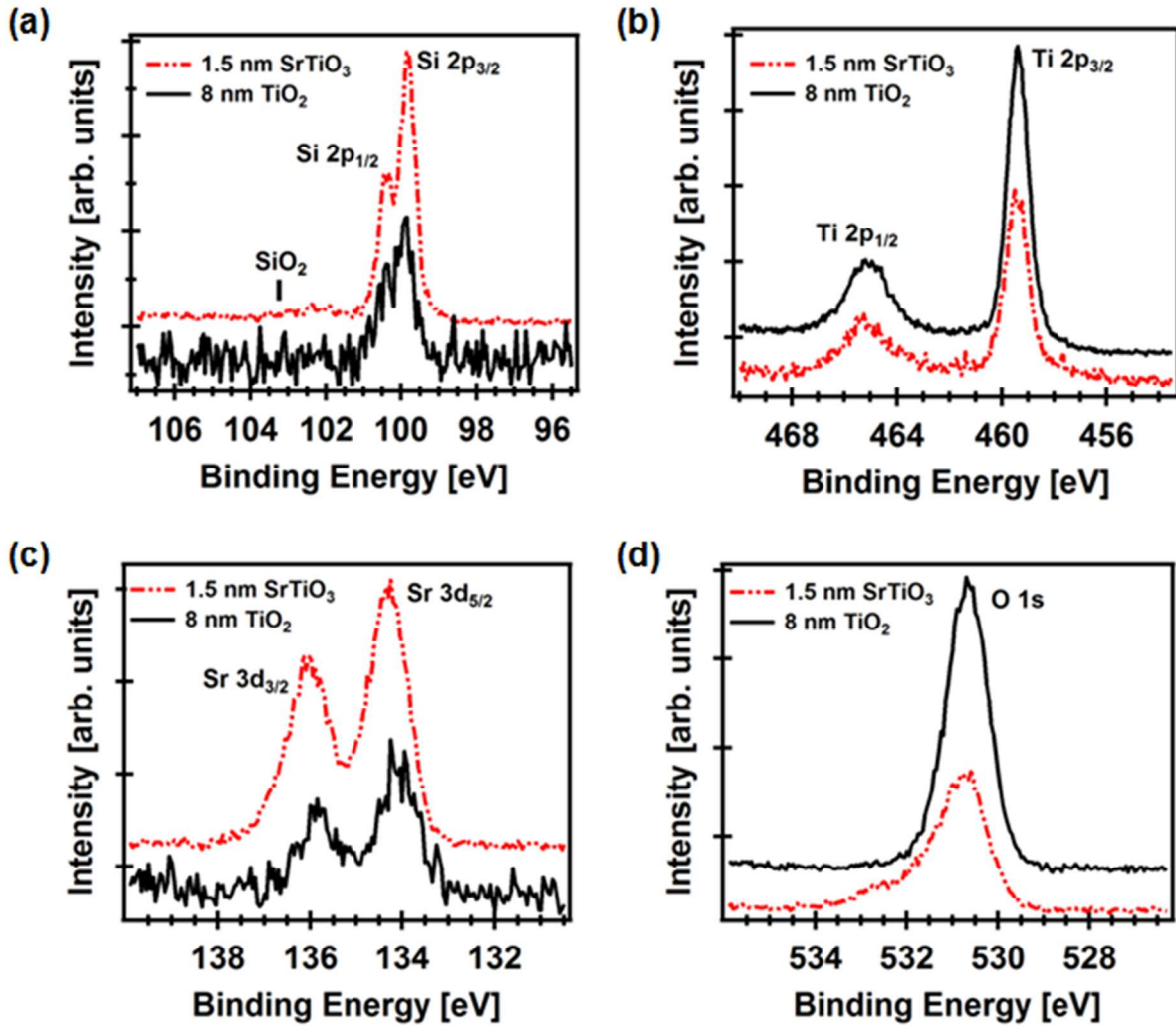
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## XPS characterization of the epitaxial single-crystal TiO<sub>2</sub> films



**Figure S1.** X-ray photoelectron spectra taken of a four-unit-cell (1.5 nm) SrTiO<sub>3</sub> buffer layer on Si (001) (dashed red line) and after 8-nm-thick epitaxial TiO<sub>2</sub> grown by ALD (solid black line). The high resolution core-level scans are shown for (a) Si 2p, (b) Ti 2p, (c) Sr 3d, and (d) O 1s.

The chemical composition of the crystalline TiO<sub>2</sub> films was characterized by *in situ* XPS. High resolution core-level spectra were collected for Si 2p, Ti 2p, Sr 3d, and O 1s, as shown in Fig. S1(a)-(d).

The Ti 2p<sub>3/2</sub> peak is located at 459.4 eV for the 8 nm TiO<sub>2</sub> film, indicating that the Ti is fully oxidized.

The Si  $2p$  spectrum verifies that there is negligible  $\text{SiO}_2$  formation after MBE growth of the single-crystal STO buffer layer, with only a small presence of silicon suboxide ( $\text{SiO}_x$ ) is observed at  $\sim 102.2$  eV. The Sr  $3d$  signal is greatly suppressed after growth of the  $\text{TiO}_2$  layer, but still visible. The O  $1s$  peak can be fit with a single Voigt function after  $\text{TiO}_2$  growth, which strongly indicates that no secondary phases are present in the epitaxial  $\text{TiO}_2$  film. For the 8 nm  $\text{TiO}_2$  film, the ALD growth was continuous and no post-deposition vacuum annealing was performed. The resulting heterostructure for the 8 nm  $\text{TiO}_2$  film is composed of  $\text{TiO}_2$  (8 nm) /  $\text{SrTiO}_3$  (1.5 nm) /  $\text{SiO}_x$  ( $\sim 0.5$  nm) / Si (001).