Supporting Information

Highly-Ordered 3D Vertical Resistive Switching Memory Arrays with Ultralow Power Consumption and Ultrahigh Density

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1. ALD deposition

The deposition procedures are fixed for all materials (TiN, TiO_2, and Pt) programmable by using Picosun SUNALETM R150 ALD System software. We can observe the color changing after each deposition step in Figure S1.

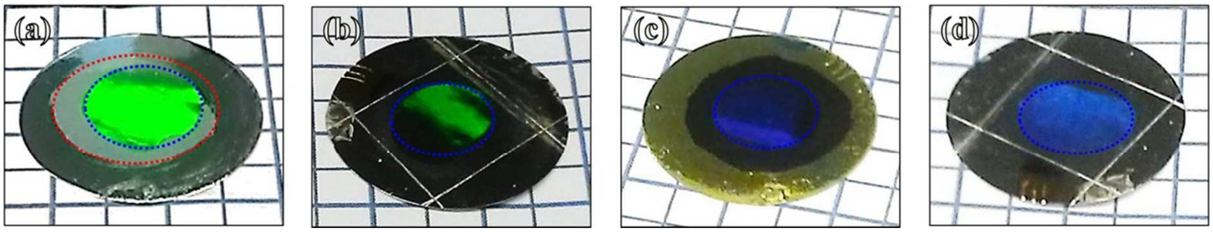

Figure S1. Photographs of the TiN@TiO_2@Pt nanotube/nanowire arrays. (a) As prepared AAO membrane, the area inside the blue marked region (diameter: 1.2 cm) is printed area (perfect ordered nanopore arrays), the area between red and blue marked region is the normal self-organized AAO membrane. (b) After 1700 cycles of TiN deposition by ALD. (c) After 1200 cycles of TiO_2 deposition by ALD. (d) After 500 cycles of Pt deposition by ALD.

2. EDX analysis

Figure S2 shows the line-scan profile of the single nanotube/nanowire which confirms the successful deposition of the three materials and the thickness of 30, 70 and 28 nm for TiN, TiO_2 and Pt (diameter: 55 nm), respectively.

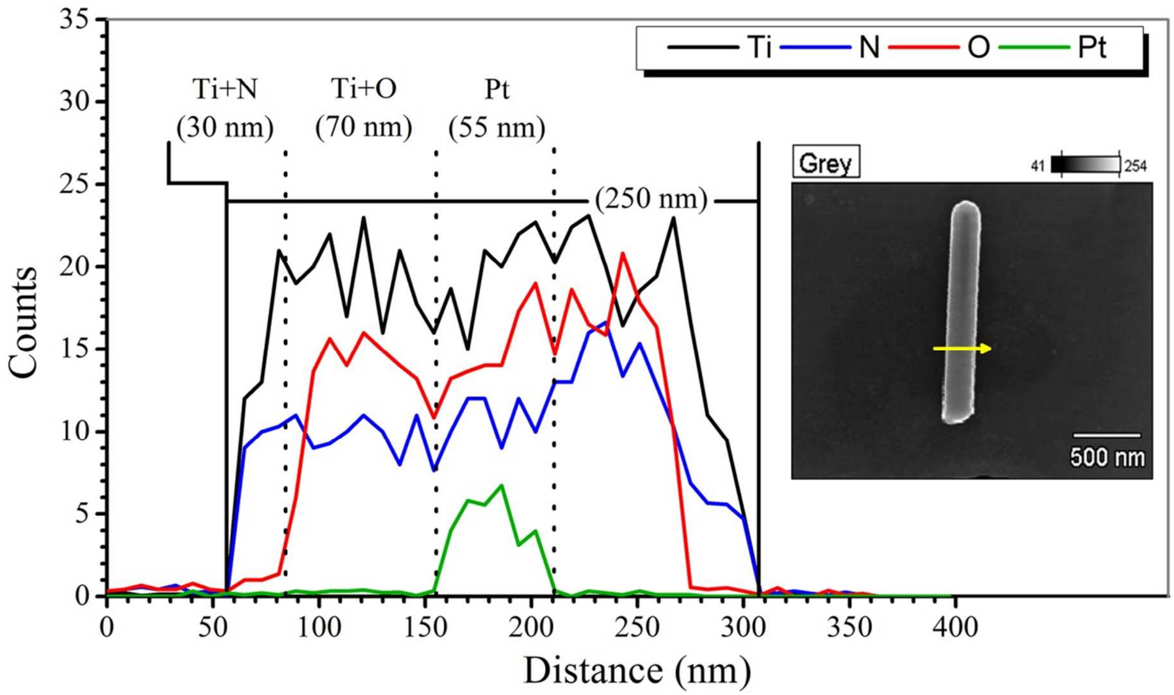

Figure S2. EDX line-scan profile of the single nanotube/nanowire along the line in the SEM image (inset).

Figure S3 shows the EDX analysis of TiN/TiO_2/Pt nanotube/nanowire. The sample prepared on Cu grid after removing the AAO template to avoid any additional counts of O atoms. The inset table represents the weight and atoms of the elements of the TiN/TiO_2/Pt nanotube/nanowire on Cu grid of the selected area of the inset SEM image.

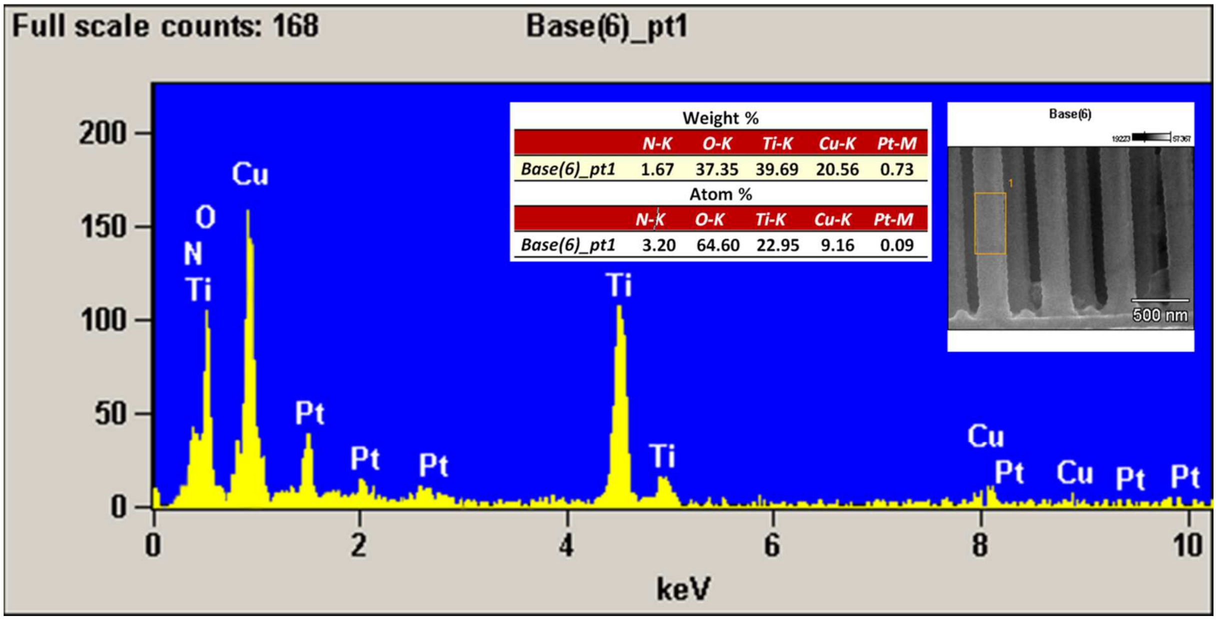

Figure S3. EDX profile of the prepared TiN/TiO_2/Pt nanotube/nanowire on Cu. The inset table represents the weight and atoms of the elements of the TiN/TiO_2/Pt nanotube/nanowire of the selected area of the inset SEM image.

3. TEM lamella Preparing

TEM lamella was prepared in a Zeiss NVision 40 CrossBeam™ focused ion beam (FIB) instrument equipped with a Ga liquid metal ion source. The energy of the Ga ions can be adjusted from 1 keV to 30 keV. In order to obtain ultrathin lamella, we have applied double-tilt method as described in the previous literature.[1] Figure S4 shows the main steps of the TEM lamella preparation: (1) deposition of amorphous carbon, (2) rough milling and lift-out and (3) double-tilt thinning. Prior to the rough thinning, an amorphous carbon layer with a thickness of ~ 2 µm was deposited using ion beam induced deposition (IBID) to protect the nanowires from subsequent Ga ion bombardment. Rough milling was carried out using 30 keV Ga ions to remove materials around the region of interest. For lift out, the lamella was attached to a manipulator using IBID and then cut free from the bulk. The dimension of the lamella was approximately 10 µm wide, 10 µm high and 2 µm thick. After lift out process, the lamella was transferred to a TEM grid using the manipulator and IBID. The TEM grid was mounted on a special rotation-tilt holder which allows double-tilt thinning to be carried out. As shown in the lower-right figure in Figure S4, the backside of the lamella was thinned perpendicular to the nanotubes. The thinning of the backside was stopped when the depth of the groove reached approximately half of the lamella thickness. Subsequently, the front side was thinned orthogonally to the first milling groove. At the end, a thin window containing a several nanowires was created where the two milling grooves overlap. During thinning, the Ga ion energy was gradually reduced from 30 keV to 1 keV to reduce ion-bombardment induced damage. The glancing angle was kept at a low value between 1° to 3° to obtain flat surfaces. The thickness of the thin window was monitored by measuring the secondary electron (SE) intensity. Figure S5 represents a 2 kV SE image of the lamella after double-tilt thinning. The thin window is indicated by the rectangle. Figure S6 shows a 300 kV TEM image of the lamella, six nanowires can be clearly seen within the thin window.

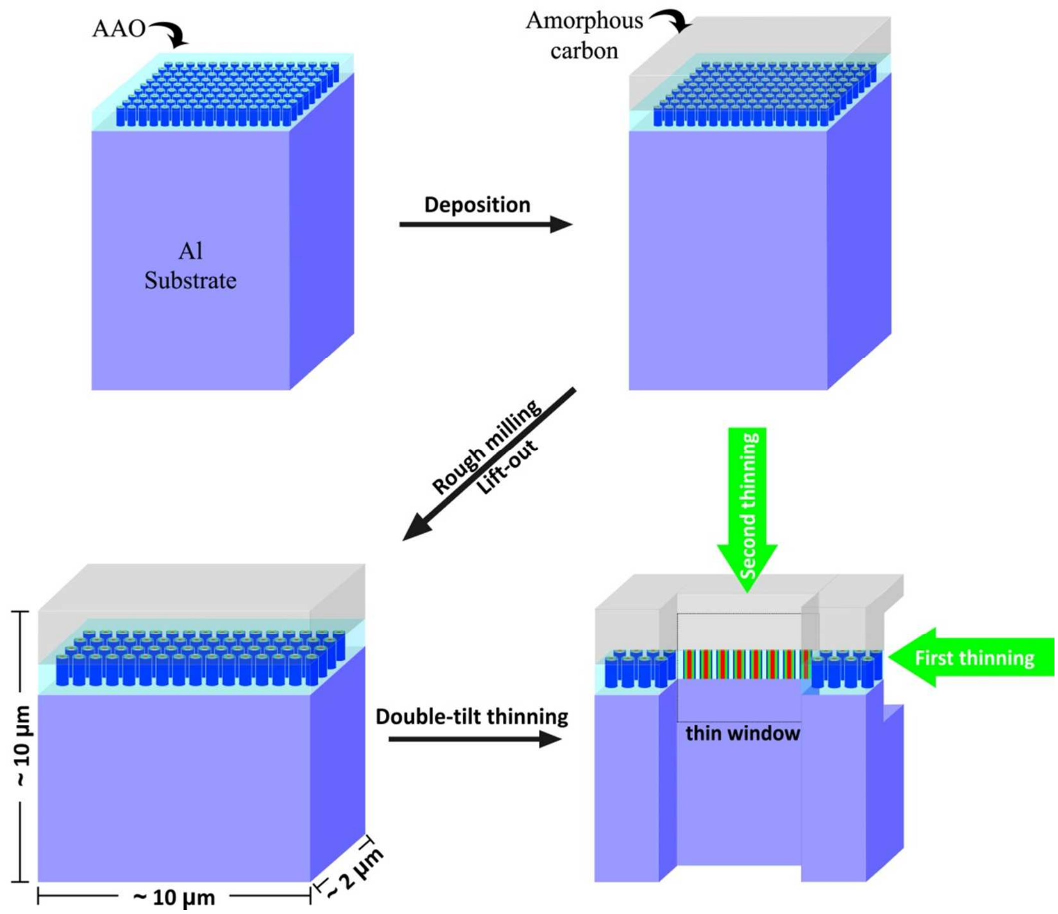

Figure S4. Main steps of TEM lamella preparation.

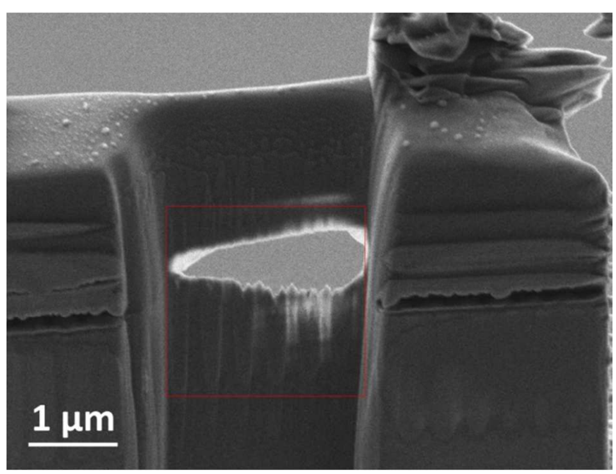

Figure S5. 2 kV SE image of the lamella after double-tilt thinning. The image was acquired by using Everhart-Thornley detector. The thin window is indicated by the red rectangle.

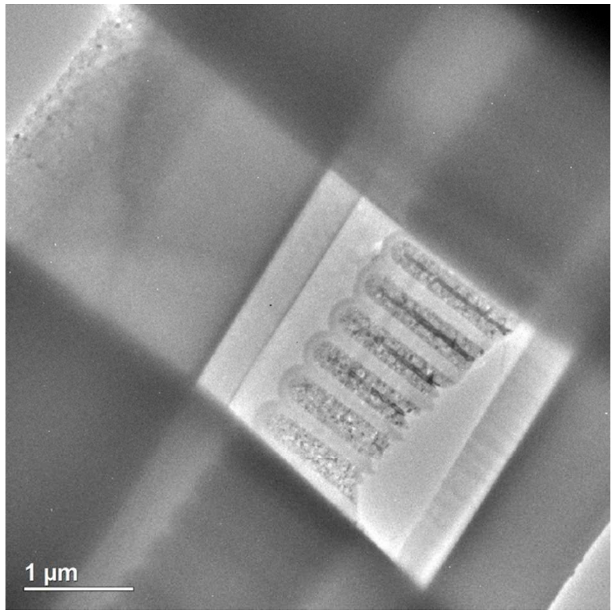

Figure S6. 300 kV TEM image showing six nanowires within the thin window

4. Analyzing the HRTEM images of TiN-TiO_2 interface

The HRTEM images in Figure S7a shows the region I representing a mixed interfacial layer between region III (TiN) and region IV (TiO_2). The thickness of the region I was around 3 nm and it is formed as a result of the sequence depositions of TiN and TiO_2. On the other side, we can confirm from FFT pattern in the Figure S7b that the regions III and IV are TiN and TiO_2, respectively. By using Gatan-DigitalMicrograph software, the calculated inverse FFT images for the two kinds of patterns in Figure S7b are shown in Figure S7c, d. By calculating the lattice space of the peaks in these figures (S7c-d), we find that lattice fingerprints are 3.525 and 2.437 Å, which are identified to TiN (111) and TiO_2 (101), respectively.

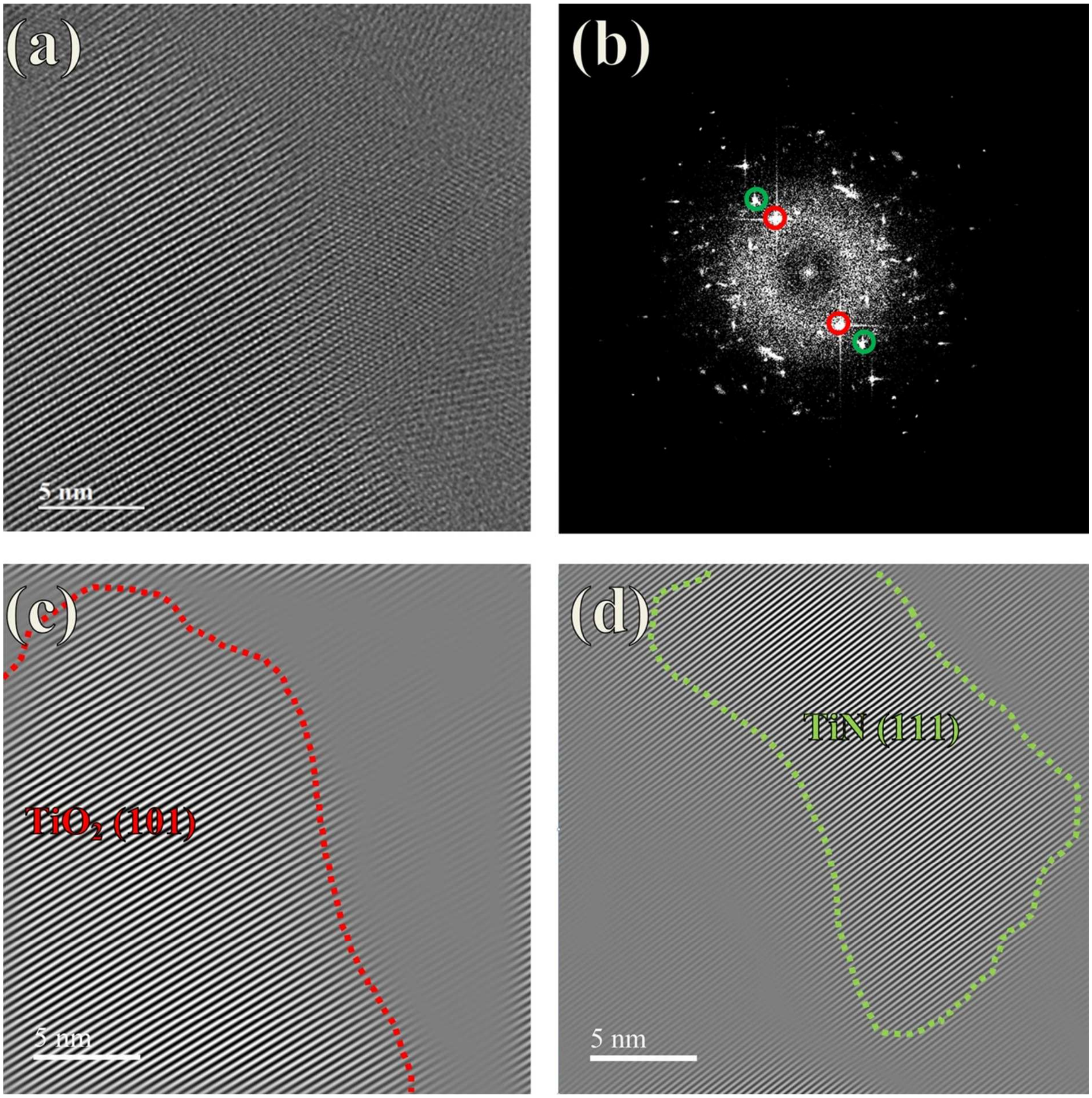

Figure S7. (a) HRTEM image of the interfacial layer between TiN and TiO_2. (b) FFT pattern of image (a). (c, d) The inverse FFT images calculated from the peaks in the image (b).

Reference

(1) Lechner, L.; Biskupek, J.; Kaiser, U., Improved Focused Ion Beam Target Preparation of (S) Tem Specimen—a Method for Obtaining Ultrathin Lamellae. *Microsc. Microanal.* **2012**, *18*, 379-384.