Electronic Supplementary Material (ESI) for Nanoscale.


# High performance electroformed Single-crystallite $VO_2$ threshold switch

Xin Zhou[a], Deen Gu[a*], Yatao Li[a], Haoxin Qin[a], Yadong Jiang[a*], Jimmy Xu[b]

[a] School of Optoelectronic Information, University of Electronic Science and Technology of China (UESTC), Chengdu, 610054, China

[b] School of Engineering, Brown University, 184 Hope Street, Providence, Rhode Island 02912, USA

* gudeen@uestc.edu.cn; jiangyd@uestc.edu.cn

## Supporting Information

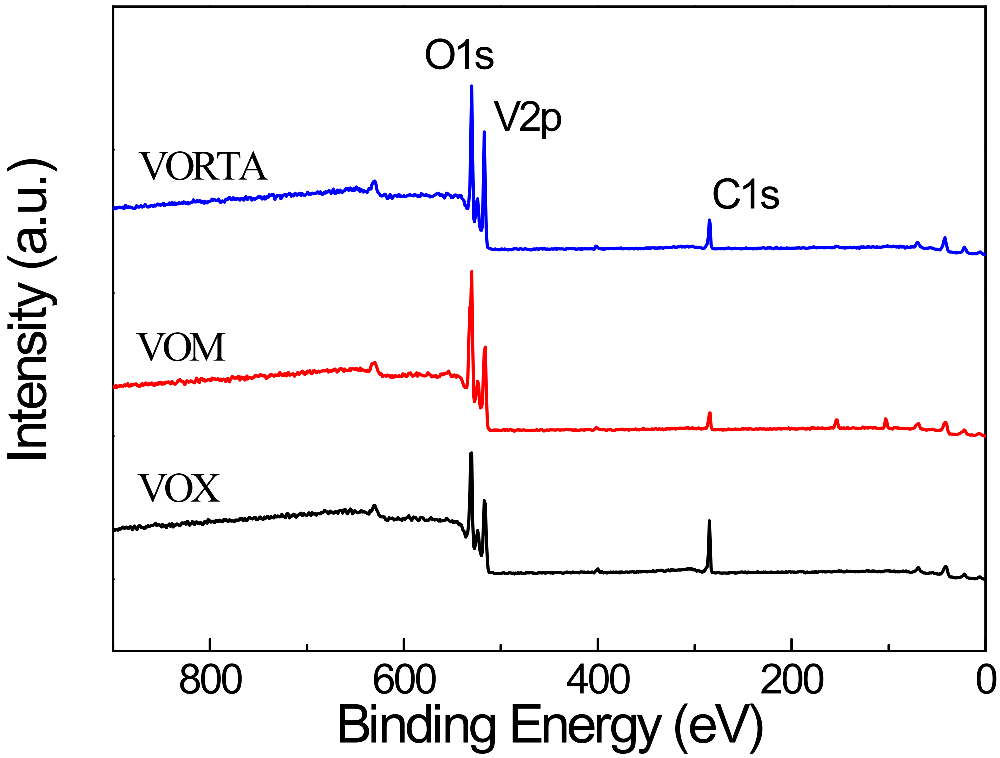


Figure S1. (a) Survey XPS spectra for VOX, VOM and VORTA.

The survey XPS spectra shows that all the samples are composed of vanadium and oxygen, no other element has been found in our samples.

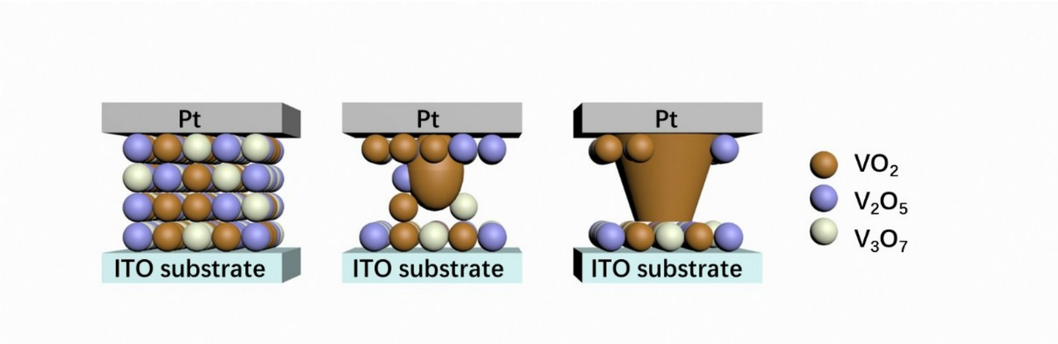


Figure S2. Schematic illustration of the electroforming process in the VORTA.

We deduce that when the voltage applied on the multiphase VORTA reaches a certain level, some $VO_2$(M) particles begin to grow. As the voltage continues to increase, these particles form $VO_2$(M) channels and the formation process ends.

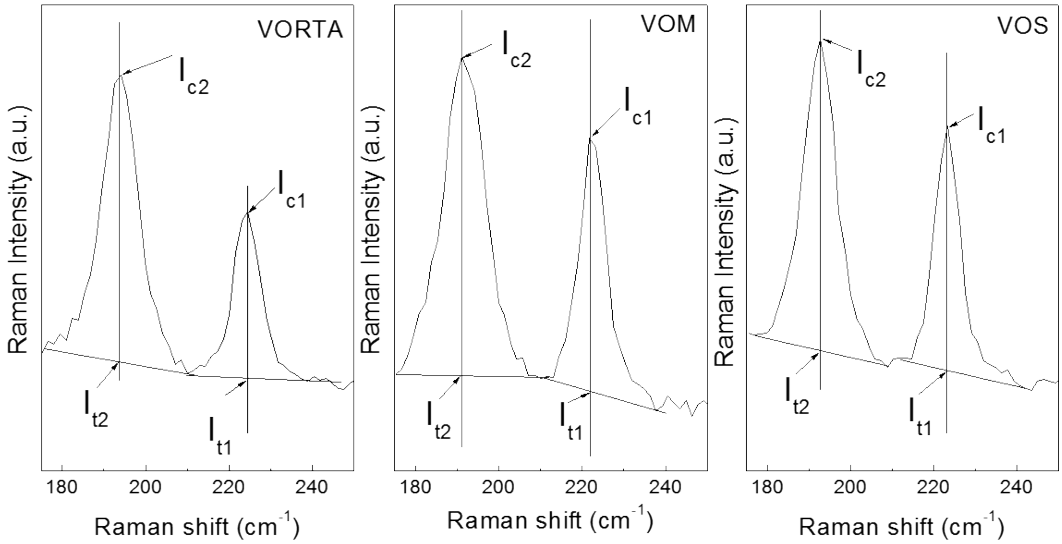


Figure S3. Crest value ($I_c$) and trough value ($I_t$) of each sample.

We find that the relative intensity (RI) of Raman peaks at 193 cm$^{-1}$ and 224 cm$^{-1}$ are closely related to the crystalline orientation of $VO_2$. The RI is calculated through the formula $RI=(I_{c1}-I_{t1})/(I_{c2}-I_{t2})$. Where the $I_c$ is the crest intensity value of Raman peak, $I_t$ is the intersection at the vertical line of $I_c$ and peak-valley line.

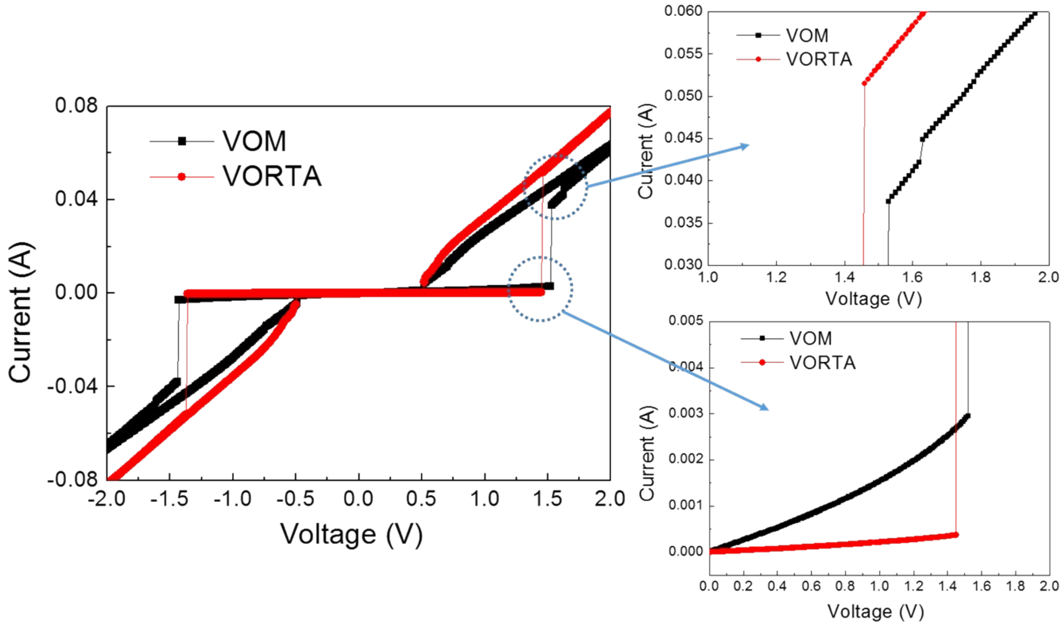


Figure S4. Detail information of the I-V curve for VOM and VORTA around the turn-on voltage. The equivalent resistance of the $VO_2$ channel in our samples are estimated by V/I, where the V is the voltage and I is the current. As shown in figure S4, the equivalent resistance of VORTA is larger than that of VOM before switching, while it became smaller than VOM after switching.

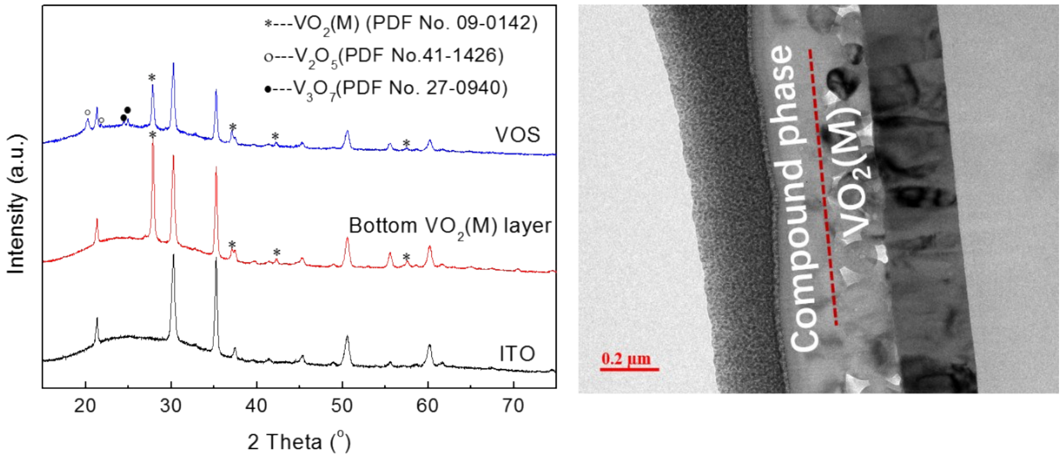


Figure S5. Left: XRD pattern for ITO substrate, bottom $VO_2$(M) layer and two layer structure VOS. Right: TEM image of two layer structure VOS.

The main diffractive peak of bottom $VO_2$(M) layer and VOS located at 27.9 °, indicated the $VO_2$(M) are (011)-oriented. TEM image reveal the two layer structure of VOS.

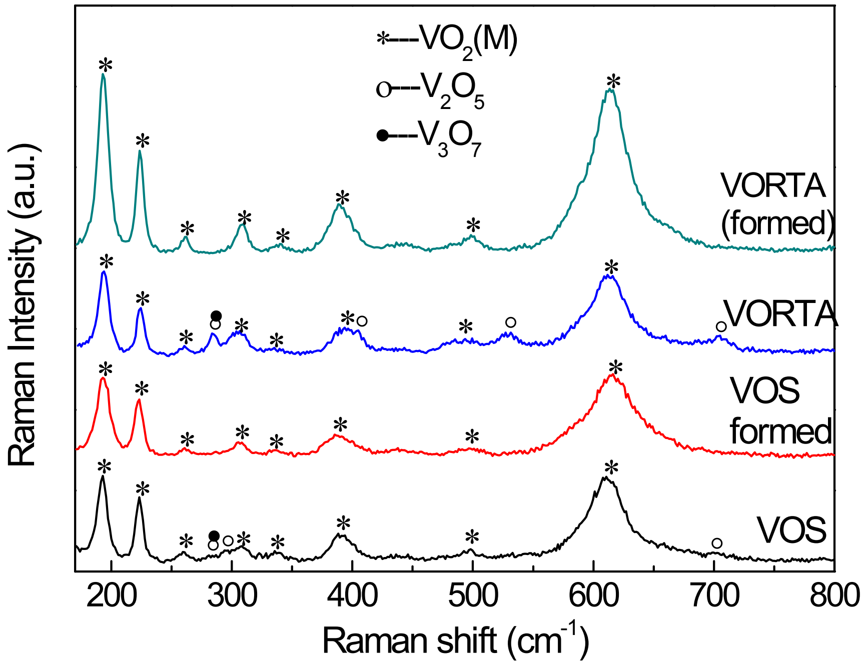


Figure S6. Raman spectra of unformed and formed area for VORTA and VOS.

The relative intensity between 224 $cm^{-1}$ and 193 $cm^{-1}$ in Raman spectra is closely relate to the crystalline orientation of $VO_2$. As shown in figure S6, the relative intensity between 224 $cm^{-1}$ and 193 $cm^{-1}$ indicated the $VO_2$ are (-211)-orientated.

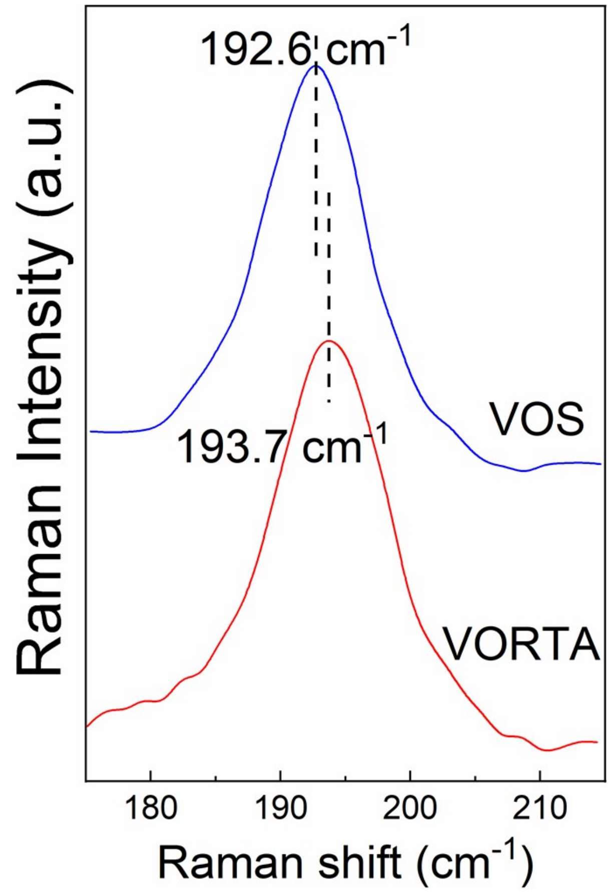


Figure S7. Raman spectra for the VORTA and VOS in the range of 175-215 $cm^{-1}$.

Figure S7 shows the Raman spectra for the VORTA and VOS in the range of 175-215 $cm^{-1}$, the low-frequency Raman spectrum has been demonstrated to be highly sensitive to the strain state of $VO_2$(M) lattice. VOS presents a lower frequency of low-frequency phonon mode ($\omega_1$) than VORTA, which indicates the (011)-orientated $VO_2$ crystallite is under more compressive strain.