SCIENTIFIC REPORTS

OPEN

Current induced polycrystalline-to-crystalline transformation in vanadium dioxide nanowires

Junho Jeong¹, Zheng Yong¹, Arash Joushaghani^{1,†}, Alexander Tsukernik², Suzanne Paradis³, David Alain³ & Joyce K. S. Poon¹

Received: 03 May 2016

Accepted: 27 October 2016

Published: 28 November 2016

Vanadium dioxide (VO₂) exhibits a reversible insulator-metal phase transition that is of significant interest in energy-efficient nanoelectronic and nanophotonic devices. In these applications, crystalline materials are usually preferred for their superior electrical transport characteristics as well as spatial homogeneity and low surface roughness over the device area for reduced scattering. Here, we show applied electrical currents can induce a permanent reconfiguration of polycrystalline VO₂ nanowires into crystalline nanowires, resulting in a dramatically reduced hysteresis across the phase transition and reduced resistivity. Low currents below 3 mA were sufficient to cause the local temperature in the VO₂ to reach about 1780 K to activate the irreversible polycrystalline-to-crystalline transformation. The crystallinity was confirmed by electron microscopy and diffraction analyses. This simple yet localized post-processing of insulator-metal phase transition materials may enable new methods of studying and fabricating nanoscale structures and devices formed from these materials.

Materials capable of insulator-metal phase transitions (IMTs) enable nanoelectronic and nanophotonic devices, such as transistors^{1–3}, memristors^{4–6}, solid-state memory^{7–9}, and optical modulators^{10,11}, with extraordinarily steep turn on characteristics and high on-off ratios that can dramatically improve the energy-efficiency of computing and communications. An example is the correlated electron material, vanadium dioxide (VO₂), which exhibits an IMT that can be induced by temperature, current, voltage, ionic gating, and electromagnetic radiation^{12–18}. In many device applications, epitaxial quality crystalline films are often preferred for improved electrical transport characteristics and spatial homogeneity. Until now, the formation of crystalline VO₂ films by methods such as pulsed laser deposition¹⁷, chemical vapor deposition¹⁹, sputtering²⁰, and vapour transport method^{21,22} has required the seeding from a suitably lattice-matched crystalline substrate, most often Al₂O₃ or TiO₂^{19,20,22,23}. Here, we demonstrate that nanowires etched from polycrystalline VO₂ films sputtered on thermally grown silica (SiO₂) on silicon (Si) can be permanently transformed to crystalline nanowires via the application of electrical currents. Thermal modelling reveals that the applied current causes heating localized to the VO₂ nanowire, which likely led to the rearrangement of grain boundaries in VO₂. Our discovery is a new way to selectively and rapidly post-process nanostructures of polycrystalline VO₂, and potentially other phase transition materials, deposited on amorphous substrates into a crystalline form.

To crystallize VO₂ on amorphous substrates, we drew inspiration from the recrystallization of metals and amorphous Si in which thermal annealing is often used to rearrange existing grain boundaries and promote the growth in size of the newly nucleated grains^{24–26}. We postulated that if sufficient localized heating is applied to polycrystalline VO₂ micro/nanostructures, VO₂ could crystallize upon cooling without the need to heat the entire substrate. Here in our manuscript we define the term “polycrystalline” to be a form when the grain size in the film is less than 100 nm in width and the “crystalline” form is when the grain boundaries are no longer observable through electron microscopy and diffraction analyses. Moreover, if the VO₂ micro/nanostructure is smaller than the final grain size, the resultant structure would effectively be “single crystalline”. A way to deliver heating that is local and specific to the VO₂ micro/nanostructure is by applying an electrical current. For sufficiently small cross-section areas, large current densities, hence sufficient heating, may be achieved at low (<3 mA) applied

¹Department of Electrical and Computer Engineering, University of Toronto, Toronto, Ontario, M5S 3G4, Canada.

²Toronto Nanofabrication Centre, University of Toronto, Toronto, Ontario, M5S 3G4, Canada. ³Defence Research and Development Canada - Valcartier, Quebec, Quebec G3J 1X5, Canada. [†]Present address: Intel, 2501 NW 229th Avenue, Hillsboro, OR 97124, USA. Correspondence and requests for materials should be addressed to J.J. (email: dave.jeong@mail.utoronto.ca)

Figure 1. The fabricated device and the as-sputtered VO₂ film. The (a) top and (b) cross-section view schematics of a VO₂ device with Pd contacts. (c) SEM images of a device at two magnifications. (d) X-ray diffraction pattern of the starting VO₂ film before crystallization.

currents and the crystallization process can be extremely rapid. This process is in contrast to the typical thermal annealing of VO₂, in which the film is heated gradually and uniformly to reduce V₂O₅ into VO₂ or create oxygen vacancies^{27–29}.

Results

Two terminal device and electrical characterization. Figure 1(a) and (b) show the top and cross-section view schematics of the two terminal VO₂ nanowire devices fabricated to demonstrate and investigate this phenomenon. A 120 nm thick VO₂ film was deposited onto a 2 μm thick thermally grown SiO₂ on a Si substrate using radio-frequency (RF) magnetron sputtering¹¹. The deposition results in polycrystalline films. Then we fabricated sets of VO₂ nanowires with varying widths and lengths using electron beam lithography and reactive ion etching. The two lateral electrodes were formed by electron-beam lithography and lift-off of a 100 nm thick palladium (Pd) film. The contacts were separated by a gap of length L varying between 0.75 μm and 7.5 μm, and the VO₂ wire width, W , varied between 0.40 μm and 50 μm. We chose to use the two-contact geometry, rather than the four-point probe geometry in ref. 30, because it allows for the straightforward measurement of the voltage vs. current (VI) characteristics before and after the crystallization using the same devices. Details of the fabrication are described in the Methods section.

To ensure that the contact resistance between the Pd contact pads and VO₂ film would not have a significant contribution to the total measured resistance, the surface area of the VO₂ film under each Pd contact was kept to a large area of 4 μm × 50 μm. This surface area was identical for all the devices such that their contact resistances would be nominally identical. The H shaped VO₂ region is indicated by the long dashed line in Fig. 1(a). Figure 1(c) shows a scanning electron microscope (SEM) images of a device with $L = 7.5$ μm and $W = 5.0$ μm. The X-ray diffraction (XRD) scan of the VO₂ film in Fig. 1(d) shows that the film was dominantly VO₂ but also had some contribution of V₂O₅. The calculated weight percentage of VO₂ and V₂O₅ was 68.9 wt.% and 31.1 wt.%, respectively. The broad low amplitude peak between 15 °2θ and 47 °2θ with a maximum around 22 °2θ in the scan is due to the amorphous SiO₂ under the VO₂ film.

The fabricated devices were measured by contacting tungsten probes to the two Pd pads. Using a precision sourcemeter with a 1 kΩ resistor in series, current was applied to the devices. This resistor protected the device from being overdriven during the IMT. First, we obtained the “before” VI relations of the polycrystalline VO₂ by sweeping the applied current between 0 mA and 0.3 mA with 1 μA increments, which is applied for 2.7 μs. We ensure that the devices undergo the nominal current-induced two-step phase transition in VO₂^{31,32} before increasing the maximum current limit. A swept voltage measurement would result in only a single step transition^{12,15,33}. Two example VI plots are shown in blue in Fig. 2(a) and (b) for VO₂ nanowires with two different dimensions. The “before” VI curve has a large hysteresis in the second step due to the Joule heating assisted phase transition¹⁵. The width of the hysteresis, as well as the current and voltage of the first and second transition depends on the dimension of the VO₂ wires³².

Figure 2. VI and resistivity measurements before and after crystallization. VI measurements for (a) $L = 0.77 \mu\text{m}$ and $W = 2.4 \mu\text{m}$ and (b) $L = 0.99 \mu\text{m}$ and $W = 0.93 \mu\text{m}$. (c) The resistivity vs. current corresponding to (a) and the inset is the magnified region in dashed lines. (d) The resistivity vs. temperature for the “before” and “after” states showing the reduced hysteresis and resistivity in the “after” state.

After the “before” measurements, for each sample, we increased the applied current beyond 0.3 mA in increments of 0.2 mA until we observed a rapid, abrupt drop in the voltage at a critical current, I_C (Supplementary Figure S1). The voltage drop occurred on time-scales shorter than the data acquisition time in our measurement setup of about $2.7 \mu\text{s}$. After the voltage drop, the current was ramped down. The change in the resultant VI was permanent, and we obtained the “after” VI relation by sweeping the applied current between 0 mA and 0.3 mA. The red lines in Fig. 2(a) and (b) show the “after” VI curves of the VO_2 wires after applying I_C of 2.3 mA and 0.68 mA, respectively. As seen in both Fig. 2(a) and (b), the transition voltage at the first step decreased by approximately 50% and the hysteresis in the second step, from the thermal part of the phase transition¹¹, was significantly reduced. The hysteresis in Fig. 2(a) has nearly collapsed completely. As discussed in the Supplementary Information, we found that the contact resistances in the VO_2 insulating and metallic phases, $R_{\text{con},i}$ and $R_{\text{con},m}$, in the “before” and “after” states were within the same order of magnitude (“before”: $R_{\text{con},i} = 62.2 \pm 9.7 \text{ k}\Omega$, $R_{\text{con},m} = 580 \pm 53 \Omega$; “after”: $R_{\text{con},i} = 54.6 \pm 7.1 \text{ k}\Omega$, $R_{\text{con},m} = 336 \pm 49 \Omega$). This means the change in the VI characteristics was not due to the contacts but rather the VO_2 wire. Furthermore, this suggests the crystallinity of the VO_2 film under the Pd contacts experienced minimal change.

Figure 2(c) compares the “before” and “after” resistivity, ρ , as a function of current for the device of Fig. 2(a) (details on the resistivity calculations are described in the Supplementary Information). The “after” plot shows that the hysteresis has been dramatically reduced and large resistivity discontinuities are absent. The almost continuous resistivity vs. current characteristics and low hysteresis width are useful for devices requiring continuous control over resistance and would suggest improved switching speeds. The reduction in the resistivity change is also observed in the ρ vs. temperature plot in Fig. 2(d). The resistivity of the “after” state is about 2.5 times lower than the “before” state for both the insulator and metallic phases of the VO_2 . This is consistent with an increased carrier scattering time or mean free path in the material by at least 2.5 times. However, the overall change in resistivity across the phase transition was reduced. The ratio between the metallic and insulator phase resistivity in the “before” case is 2.3×10^{-2} , and the resistivity ratio in the “after” case is 3.6×10^{-2} . The “before” and “after” phase transition temperatures were about the same at approximately 343 K, but in the “after” case, the hysteresis width was reduced to about 3 K, identical to that of single crystal VO_2 ^{19,20,23,34}. Furthermore, all the VI and ρ vs. temperature measurements were repeatable in both the “before” and “after” case. Device failure occurred when a current beyond I_C was injected into the device.

Due to the small size of the wires and the device geometry, it is difficult to directly determine the material composition, stoichiometry, or the vanadium valence state in the “after” state using standard techniques such as XRD or X-ray photoelectron spectroscopy (XPS). However, the “after” state should be dominantly VO_2 since the phase transition temperature in the “before” and “after” cases are similar (Fig. 2(d)). It is unlikely that the VO_2 wires were converted to other vanadium oxides, such as VO , V_2O_3 , or V_2O_5 , which have drastically different phase

Figure 3. Electron microscopy images and diffraction patterns before and after the crystallization. Top view SEM images of the VO_2 nanowire device with $L = 7.5 \mu\text{m}$ and $W = 0.91 \mu\text{m}$ (a) before and (b) after the crystallization. XTEM images of a VO_2 nanowire (c) before and (d) after crystallization showing the absence of grains in the “after” state. (e) is the high-resolution TEM and (f) the SAED pattern corresponding to the “before” state in (c). The polycrystallinity is evident in the non-uniform plane orientations in (e) and randomness of the diffraction pattern in (f). (g) is the high-resolution TEM and (h) the SAED pattern corresponding to the “after” state in (d) taken in the VO_2 metallic (rutile) phase. The atomic arrangement is more periodic and the diffraction pattern exhibits clear symmetries that can be matched to the (i) computed diffraction pattern of VO_2 with a rutile crystal structure at view direction $[210]$ using the lattice parameters from ref. 38.

transition temperatures compared to VO_2 (for example, the transition temperature of V_2O_3 and V_2O_5 are around 150 K and 553 K, respectively^{35,36}). The reduction in the resistivity change in the “after” state may be due to the small quantity of V_2O_5 (Fig. 1(d)) or changes in the strain in the VO_2 wires³⁷.

Electron microscopy and diffraction analyses. The “after” state of the devices was found to be due to a permanent structural transformation of the polycrystalline VO_2 into crystalline VO_2 . Figure 3 shows SEM, cross-sectional transmission electron microscope (XTEM) and selected area electron diffraction (SAED) images of the VO_2 nanowires in the “before” and “after” states. Figure 3(a) and (b) show the “before” and “after”, respectively, top-view SEM images of the device with $L = 7.5 \mu\text{m}$ and $W = 0.91 \mu\text{m}$. The top surface of the VO_2 in Fig. 3(a) is rough with visible grain boundaries. From the XTEM image of the VO_2 film in the “before” state in Fig. 3(c), the grain sizes are estimated to be about $75 \pm 25 \text{ nm}$ in width. In Fig. 3(b), the VO_2 surface is smooth with no visible grain boundaries. The expanded width on the right end of the wire is suggestive of material reflow. The XTEM of the “after” state in Fig. 3(d) also shows the absence of grain boundaries. Because only a small region could be imaged, grain boundaries could exist along the VO_2 wire width and length and we were not able to confirm whether the wire was entirely single crystal. For devices with $W > 5 \mu\text{m}$, only a narrow region in the

VO₂ wire appeared smooth, suggesting that the current was concentrated through a filament that had a minimum resistance between the two contact pads (Supplementary Figure S3). The width of the narrow, smooth region is consistent with our previous measurements of the width of thermal filaments in ref. 32.

Figure 3(e) and (f) respectively show high-resolution XTEM and SAED images of the VO₂ in a device at room temperature. The images were taken over a sample diameter of 150 nm. Numerous crystal orientations are evident in Fig. 3(e) and the randomness of the diffraction pattern in Fig. 3(f) confirm the polycrystallinity of the VO₂. In contrast, the high-resolution XTEM and SAED images of VO₂ in Fig. 3(g) and (h), respectively, clearly shows crystalline characteristics. These images have been taken at the metallic phase of the VO₂ at 80 °C and were taken from a device different from that in Fig. 3(e) and (f) due to the need for sample preparation (Supplementary Figure S4). In Fig. 3(g), the atomic arrangement is spatially regular without any grain boundaries, and the clear diffraction pattern exhibiting symmetries in Fig. 3(h) further suggests the VO₂ is crystalline. Using the SingleCrystal software package, we matched the observed diffraction pattern with the expected diffraction pattern using the lattice parameters of VO₂ in the metallic phase (rutile structure) from previous studies³⁸. The calculated diffraction pattern is shown in Fig. 3(i). A challenge in matching the computed diffraction pattern with the measurement was the unknown orientation of the VO₂ sample relative to the incident electron beam. To determine the incident direction of the beam, in Fig. 3(h), we first identified a separation of 1.42 Å between the center of the diffraction pattern and the nearest diffraction spot in the horizontal direction. This distance is similar to half of the *c*-axis lattice constant of VO₂ (*c* = 2.85 Å), so we identified the diffraction spot to be from the (002) plane and varied the electron beam incident direction until the rest of the diffraction pattern was matched. A confirmation of the fitting is that with the correct view direction of the diffraction pattern, [210], the simulated diffraction pattern aligns with the SAED pattern. The faint spots in the diffraction pattern that do not match with the VO₂ are likely due to the residual V₂O₅. The complete fittings of the diffraction patterns for both the monoclinic and rutile phases and the contribution from V₂O₅ are included in the Supplementary Information.

Thermal modelling. Through measurements of devices with varying *W* and *L* combined with heat transport simulations, we can ascribe the observed polycrystalline-to-crystalline transformation to a thermally activated process, akin to recrystallization in metals and amorphous semiconductors^{24–26}. Applying Fourier's law of heat conduction ($\vec{q} = -k\nabla T$, where $\vec{q}$ is the heat flow, *k* is the thermal conductivity, and *T* is the temperature) to the VO₂ nanowire, in the one-dimensional steady-state, if $\vec{q}$ is taken to be the dissipated power (*P*) over cross-section area (*Wt*), then

$$P = \frac{kWt\Delta T}{L} \quad (1)$$

where ΔT is the change in temperature. Therefore, the dissipated power is expected to be linearly proportional to the geometric parameter, *W/L*. In our experiment, $P = I_C^2 R$, with $R = R_{tot} - 2R_{con,m}$ where R_{tot} is the total measured resistance of the device. Figure 4(a) shows the measured dissipated electrical power, *P*, at the critical current for the structural change is indeed a linear function of *W/L*. Using the thermal conductivity of VO₂ in ref. 39, the measured data is fitted to Equation 1 with $\Delta T = 1470$ K as shown in Fig. 4(a) in red. This result suggests that the local temperature of the VO₂ wire was around 1770 K for the polycrystalline-to-crystalline transformation to occur. This temperature is about 80% of the melting point of VO₂ of 2240 K, which is consistent with the requirements of recrystallization processes^{24,25}. For example, amorphous Si can be recrystallized at around 773–913 K even though the melting point of Si is 1687 K^{24,25}. In recrystallization, the energy required for nucleation and grain growth responsible for the change in crystallinity is less than the energy required for melting^{25,26}.

To obtain a more accurate estimate of the temperature for the polycrystalline-to-crystalline transformation and to confirm whether such temperatures could have been generated in our experiment, we simulated the temperature profile in VO₂ nanowires, including the Pd contacts and substrate, at the critical current. The calculations were performed using Joule heating module of COMSOL Multiphysics a stationary study in three dimensions using a non-uniform mesh (minimum mesh size of 5 nm) for different VO₂ wire dimensions. The top surfaces of the structure are modelled as convective cooling surfaces allowing thermal transport into the air. The side and bottom boundaries were set to room temperature and were sufficiently far away from the VO₂ wire such that they did not influence the results of the heat transport in the wire. The heat capacity of VO₂ was modelled as ref. 40 and the thermal conductivity values were taken from ref. 39. In the thermal simulations, the applied voltage across the contact pads is varied to match the current density through the VO₂ nanowire with the experimental critical current density required for crystallization. Figure 4(b) and (c) show the matched simulated current density and corresponding temperature profile, respectively, for the VO₂ wire with *L* = 0.78 μm and *W* = 0.41 μm. The current density is the highest near the interface between the Pd contacts and VO₂ wire. The centre of the VO₂ wire reaches the highest temperature of 1780 K, similar to the value extrapolated from the one dimensional Fourier model and Fig. 4(a) of 1770 K. Different wire dimensions resulted in similar maximum temperature values to within 10%. Interestingly, the highest temperature region is localized to the centre of the nanowire, which may explain why the contact resistance was unchanged in the polycrystalline-to-crystalline transformation as the temperature may not have been sufficiently high at the Pd contacts for VO₂ to crystallize. Moreover, at these high temperatures it is unlikely that VO₂ will further oxidize with the oxygen in the air to form V₂O₅. This is because the boiling temperature of V₂O₅ is 1750 K⁴¹ and at around 1770 K it will most likely thermally decompose.

Discussion

In summary, we have demonstrated the transformation of polycrystalline VO₂ into crystalline VO₂ in nanowire geometries by injecting electrical currents of the order of 1 mA to reach local temperatures near 1780 K. The reconfiguration is likely through a process akin to recrystallization in metals and semiconductors by thermal annealing, with a key difference that the transformation here was rapid, occurring within microseconds. The final

Figure 4. Thermal modeling of the crystallization. (a) The measured dissipated power at the critical current compared to the power required to heat the VO₂ nanowires by $\Delta T = 1470$ K calculated using (a) 1D Fourier model for devices with varying W/L . The result of a thermal transport simulation (COMSOL Multiphysics) showing (b) the current density and (c) temperature distribution for VO₂ wire of dimensions $L = 0.78 \mu\text{m}$ and $W = 0.41 \mu\text{m}$ at the critical current, I_C .

crystal orientation of the VO₂ nanowires was not controlled in these experiments, but it may be possible to induce a preferential orientation by introducing strain or patterning the substrate, for example. The crystallized devices exhibited reduced hysteresis across the phase transition and reduced resistivity, essential features for common electronic and photonic devices. Because polycrystalline VO₂ films can be deposited at low temperatures (less than around 450–500 °C) on amorphous dielectrics or silicon, compatible with back-end of line complementary metal oxide semiconductor (CMOS) fabrication processing^{17,28}, our findings open the potential for highly dense, three-dimensional integration of crystalline VO₂ and other IMT materials with CMOS devices. Further, the simplicity of the current induced crystallization can broaden the investigation of crystalline VO₂ and IMT nanostructures on unconventional substrates, including those that are lattice-mismatched, amorphous, or flexible.

Methods

VO₂ deposition. The VO₂ film was deposited using reactive RF magnetron sputtering of a 50 mm vanadium target (99.7% purity). A 4 cm × 4 cm piece of Si with a 2 μm thick of thermally grown SiO₂ was placed on a 150 mm diameter Si wafer, which served as the substrate holder. During the deposition, the substrate holder was held at 500 °C and the chamber was filled with high purity argon (Ar) and oxygen (O₂) at flow rates of 80.0 sccm and 2.0 sccm, respectively at a total pressure of 10 mT. The RF power was ramped to 200 W and the O₂ flow rate was closely monitored to keep the DC bias of the sample constant. The deposition rate was approximately 0.3 Å/s.

Device Fabrication. The devices were fabricated using aligned electron-beam lithography, dry etching, and metal deposition. First, 20 μm × 20 μm tungsten alignment markers for the electron-beam lithography were formed onto the VO₂ film. Then, the patterns for the VO₂ nanowires were written using an electron-beam writer (Vistec EBPG 5000, at a dosage of 250 C/cm² and a current of 500 pA) in a 450 nm thick layer of ZEP-520A resist. The chip was then developed in ZED-N50 for 60 s and then in methyl isobutyl ketone (MIBK) for 30 s. Prior to etching, the resist was baked for 4 min at 100 °C. The sample was then etched using an inductively coupled plasma reactive ion etcher (ICP-RIE). The sample was placed on an aluminum chuck and pre-heated on a hotplate to 80 °C prior to insertion into the ICP-RIE, so the VO₂ was in its metallic phase during the etch. A gas mixture of Cl₂ (6 sccm), H₂ (6 sccm), and Ar (9 sccm) at a total pressure of 5 mT was used for the etch. At a RF power of 125 W and ICP power of 500 W, 50 s was required for a full etch of the VO₂. After etching, the ZEP resist was removed by immersing the sample in ZDMAC for 10 min at 70 °C followed by 1 min of sonication in the same solution. With this fabrication process, the minimal feature size of the VO₂ nanowire obtained was 400 nm in width and 750 nm in length. Finally, contact pads were formed on the VO₂ nanowires by aligned electron-beam lithography (with ZEP-520A resist), thermal evaporation of a 3 nm thick chromium adhesion layer and a 100 nm thick Pd layer (deposition rate: 1.5 Å/s), and lift-off.

References

- Shibuya, K., Ohnishi, T., Lippmaa, M., Kawasaki, M. & Koinuma, H. Single crystal SrTiO₃ field-effect transistor with an atomically flat amorphous CaHfO₃ gate insulator. *Applied Physics Letters* **85**, 425–427 (2004).
- Zhou, Y. & Ramanathan, S. Correlated Electron Materials and Field Effect Transistors for Logic: A Review. *Critical Reviews in Solid State and Materials Sciences* **38**, 286–317 (2013).
- Shukla, N. *et al.* A steep-slope transistor based on abrupt electronic phase transition. *Nature communications* **6**, 7812 (2015).
- Driscoll, T., Kim, H. T., Chae, B. G., Di Ventra, M. & Basov, D. N. Phase-transition driven memristive system. *Applied Physics Letters* **95**, 2–4 (2009).
- Ha, S. D., Aydogdu, G. H. & Ramanathan, S. Metal-insulator transition and electrically driven memristive characteristics of SmNiO₃ thin films. *Applied Physics Letters* **98**, 98–101 (2011).
- Pickett, M. D., Medeiros-Ribeiro, G. & Williams, R. S. A scalable neuristor built with Mott memristors. *Nature materials* **12**, 114–7 (2013).
- Watanabe, Y. *et al.* Current-driven insulator-conductor transition and nonvolatile memory in chromium-doped SrTiO₃ single crystals. *Applied Physics Letters* **78**, 3738–3740 (2001).
- Kwon, D.-H. *et al.* Atomic structure of conducting nanofilaments in TiO₂ resistive switching memory. *Nature nanotechnology* **5**, 148–153 (2010).
- An electrically tuned solid-state thermal memory based on metal-insulator transition of single-crystalline VO₂ nanobeams. *Advanced Functional Materials* **21**, 1602–1607 (2011).
- Kawakami, Y. *et al.* Optical modulation of effective on-site coulomb energy for the Mott transition in an organic dimer insulator. *Physical Review Letters* **103**, 2–5 (2009).
- Joushaghani, A. *et al.* Wavelength-size hybrid Si-VO₂ waveguide electroabsorption optical switches and photodetectors. *Optics Express* **23**, 3657 (2015).
- Stefanovich, G., Pergament, A. & Stefanovich, D. Electrical switching and Mott transition in VO₂. *J. Phys.: Condens. Matter* **12**, 8837–8845 (2000).
- Liu, M. *et al.* Terahertz-field-induced insulator-to-metal transition in vanadium dioxide metamaterial. *Nature* **487**, 345–8 (2012).
- Aetukuri, N. B. *et al.* Control of the metal-insulator transition in vanadium dioxide by modifying orbital occupancy. *Nat Phys* **9**, 661–666 (2013).
- Joushaghani, A. *et al.* Voltage-controlled switching and thermal effects in VO₂ nano-gap junctions. *Applied Physics Letters* **104** (2014).
- Martens, K. *et al.* Field Effect and Strongly Localized Carriers in the Metal-Insulator Transition Material VO₂. *Physical Review Letters* **115**, 2–7 (2015).
- Jeong, J. *et al.* Suppression of Metal-Insulator Transition. *Science* **339**, 1402–1405 (2013).
- Morrison, V. R. *et al.* A photoinduced metal-like phase of monoclinic VO₂ revealed by ultrafast electron diffraction. *Science* **346**, 445 (2014).
- Kim, H., You, H., Chiarello, R. & Chang, H. Finite-size effect on the first-order metal-insulator transition in VO₂ films grown by metal-organic chemical-vapor deposition. *Physical Review B* **47**, 900–907 (1993).
- De Natale, J. F., Hood, P. J. & Harker, A. B. Formation and characterization of grain-oriented VO₂ thin films. *Journal of Applied Physics* **66**, 5844–5850 (1989).
- Guiton, B. S., Gu, Q., Prieto, A., Gudiksen, M. S. & Park, H. Single-Crystalline Vanadium Dioxide Nanowires with Rectangular Cross Sections. *Journal of the American Chemical Society* **127**, 498–499 (2005).
- Wu, J. *et al.* Strain-induced self organization of metal-insulator domains in single-crystalline VO₂ nanobeams. *Nano Letters* **6**, 2313–2317 (2006).
- Nag, J. & Haglund, R. F. Jr. Synthesis of vanadium dioxide thin films and nanoparticles. *Journal of Physics: Condensed Matter* **20**, 264016 (2008).
- Roth, J. A. & Anderson, C. L. Silicon epitaxy by solid-phase crystallization of deposited amorphous films. *Applied Physics Letters* **31**, 689–691 (1977).
- Iverson, R. B. & Reif, R. Recrystallization of amorphized polycrystalline silicon films on SiO₂: Temperature dependence of the crystallization parameters. *Journal of Applied Physics* **62**, 1675–1681 (1987).
- Humphreys, F. & Hatherly, M. *Recrystallization and Related Annealing Phenomena*, 2 edn (Elsevier, 2004).
- Suh, J. Y., Lopez, R., Feldman, L. C. & Haglund, R. F. Semiconductor to metal phase transition in the nucleation and growth of VO₂ nanoparticles and thin films. *Journal of Applied Physics* **96**, 1209–1213 (2004).
- Rathi, S. *et al.* Postfabrication annealing effects on insulator-metal transitions in VO₂ thin-film devices. *ACS Applied Materials and Interfaces* **6**, 19718–19725 (2014).
- Marvel, R. E., Harl, R. R., Craciun, V., Rogers, B. R. & Haglund, R. F. Influence of deposition process and substrate on the phase transition of vanadium dioxide thin films. *Acta Materialia* **91**, 217–226 (2015).
- Gu, W., Choi, H. & Kim, K. K. Universal approach to accurate resistivity measurement for a single nanowire: Theory and application. *Applied Physics Letters* **89** (2006).
- Kim, H. T. *et al.* Electrical oscillations induced by the metal-insulator transition in VO₂. *Journal of Applied Physics* **107** (2010).
- Joushaghani, A. *et al.* Characteristics of the Current-Controlled Phase Transition of VO₂ Microwires for Hybrid Optoelectronic Devices. *Photonics* **2**, 916–932 (2015).
- Zimmers, A. *et al.* Role of thermal heating on the voltage induced insulator-metal transition in VO₂. *Physical Review Letters* **110**, 1–5 (2013).
- Zhong, X., Zhang, X., Gupta, A. & Leclair, P. Avalanche breakdown in microscale VO₂ structures. *Journal of Applied Physics* **110**, 1–6 (2011).
- Ji, Y. D. *et al.* Epitaxial growth and metal-insulator transition of vanadium oxide thin films with controllable phases **101** (2012).
- Kang, M. *et al.* Metal-insulator transition without structural phase transition in V₂O₅ film Metal-insulator transition without structural phase transition in V₂O₅ film. *Applied Physics Letters* **98** (2011).
- Nakano, M. *et al.* Collective bulk carrier delocalization driven by electrostatic surface charge accumulation. *Nature* **487**, 459–462 (2012).
- Kück, S. & Werheit, H. V₂O₅: crystal structure, lattice parameters, density. In Madelung, O. (ed.) *Non-Tetrahedrally Bonded Binary Compounds II*, 1 edn (Springer-Verlag Berlin Heidelberg, 2000).
- Oh, D. W., Ko, C., Ramanathan, S. & Cahill, D. G. Thermal conductivity and dynamic heat capacity across the metal-insulator transition in thin film VO₂. *Applied Physics Letters* **96**, 1–4 (2010).
- Berglund, C. N. Thermal Filaments in Vanadium Dioxide. *IEEE Transactions on Electron Devices* **16**, 432–437 (1969).
- Greenwood, N. & Earnshaw, A. In *Chemistry of the Elements*, 2 edn (Butterworth-Heinemann, Oxford, 1997).

Acknowledgements

Funding from the Natural Sciences and Engineering Research Council of Canada and the Canada Research Chairs program is gratefully acknowledged. The authors thank Joel Loh for assistance in the XRD measurement, and the Canadian Centre for Electron Microscopy for the SAED and TEM measurements.

Author Contributions

J.J., A.J. and J.K.S.P. designed the experiment. J.J. and Z.Y. fabricated the devices, performed electron microscopy, and conducted the electrical experiments. A.T. executed the e-beam lithography. S.P. and D.A. deposited the VO₂ film. J.J. and J.K.S.P. wrote the manuscript and all authors have given approval to the final version of the manuscript.

Additional Information

Supplementary information accompanies this paper at <http://www.nature.com/srep>

Competing financial interests: The authors declare no competing financial interests.

How to cite this article: Jeong, J. *et al.* Current induced polycrystalline-to-crystalline transformation in vanadium dioxide nanowires. *Sci. Rep.* **6**, 37296; doi: 10.1038/srep37296 (2016).

Publisher's note: Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

This work is licensed under a Creative Commons Attribution 4.0 International License. The images or other third party material in this article are included in the article's Creative Commons license, unless indicated otherwise in the credit line; if the material is not included under the Creative Commons license, users will need to obtain permission from the license holder to reproduce the material. To view a copy of this license, visit <http://creativecommons.org/licenses/by/4.0/>

© The Author(s) 2016