

VO₂: a material for high-performance micro/nanoactuators

Nicola Manca,^{1,2,3} Luca Pellegrino,² Teruo Kanki,⁴ Warner J. Venstra,³ Giordano Mattoni,³ Yoshiyuki Higuchi,⁴ Hidekazu Tanaka,⁴ Andrea D. Caviglia,³ and Daniele Marre^{1,2}

¹Physics Department, University of Genova, Genova, Italy

²CNR-SPIN, Genova, Italy

³Kavli Institute of Nanoscience, Delft University of Technology, Delft, The Netherlands

⁴Institute of Scientific and Industrial Research, Osaka University, Osaka, Japan

Vanadium Dioxide (VO₂) is an interesting material for micro/nanoactuators due to its fast and reversible Solid State Phase Transitions (SSPT) at 68 ° that in single crystals shows strain values up to 1% and large applied forces. This transition is hysteretic and occurs in a temperature window of 5–10 °C. In thin films it shows percolative behavior, spreading with the switching of single domains having micrometric or nanometric size, depending on the growing conditions. We present two VO₂-based microdevices exploiting the domain structure of this SSPT. A programmable mechanical resonator having mechanical memory capabilities and a mechanical oscillator excited only by a DC source are discussed. VO₂-based microactuators could add unique features to the field of nanoactuators thanks to its peculiar properties and to the possibility to preserve and control the SSPT of this material down to submicrometric scale. Details on the project “solid state actuators for micro/nanorobotics” can be found at <http://www.vo2actuators.spin.cnr.it>.

Contribution presented at ACTUATOR 2018 (Bremen, Germany), see www.actuator.de.

Keywords: VO₂, microactuators, phase transition, mechanical resonators

Introduction

Solid State Phase Transitions (SSPT) have intrinsically high work energy densities and are employed as driving mechanisms in actuating devices, like the case of the well-known class of Shape Memory Alloys (SMA) [1]. Some transition metal oxides also show SSPT and can be competitive with SMA in terms of miniaturization for the development of micro and nanoactuators. Vanadium Dioxide (VO₂) has been recently considered as an optimum active material in micro/nanoactuators, due to its sharp and reversible SSPT at 68°C from a monoclinic to a tetragonal phase that in single crystal shows strain values up to 1% and large applied forces [2, 3]. The work energy density associated with the SSPT of VO₂ is 0.63 J/cm³ for thin films and theoretically 7 J/cm³ for single crystals [4]. Remarkably, its phase transition is intrinsically fast (ps) and occurs in a sharp temperature window of 5–10 °C, with hysteresis between the heating and cooling branches. The SSPT, illustrated in Figure 1(a), results in a change of the electrical resistance up to four orders of magnitude from a low temperature insulating phase to a high temperature metallic phase, together with a modification of the optical reflectance. These additional elements are used to drive and detect the SSPT in VO₂-based microstructures. In this work, we present some examples of micromechanical devices based on VO₂ thin films whose working principle is based on the control of the SSPT in free-standing micrometric structures.

Experimental

The fabrication process of our device is described in Figure 1(b). VO₂ films are grown by Pulsed Laser Depo-

sition on top of a TiO₂-buffered MgO(001) single crystal substrate (I). Film thickness varies between 50 and 200 nm, while the thickness of the buffer layer, required for optimal epitaxial growth and to control the device mechanical characteristics, spans between 5 and 70 nm. The devices' geometries are realized by optical or e-beam lithography (II) and different etching techniques (III) (Ar milling, acid bath, exposure to acid vapours). After a cleaning step in acetone to remove the fabrication residues (IV), the structures are made free-standing by selective chemical etching of the MgO substrate in H₃PO₄ (V). As final step, the structure are dried in a critical point dryer to prevent stiction (Figure 1(c)). Electrical and mechanical measurements are performed in a custom sample holder with controlled temperature and gas pressure. Mechanical characterizations are performed using optical lever technique by focusing a laser on the micro-structures.

Results & Discussions

During the SSPT, VO₂ shows phase separation in domains whose size can vary from nanometric to micrometric dimensions, depending on the substrate of growth. Apart from observing the phase transition by increasing the temperature of the specimen, the SSPT itself can be induced by electro-thermal heating, exploiting Joule effect through a proper electrical bias. If a current is applied across a micrometric VO₂-based structure, progressive formation of metallic submicrometric VO₂ regions is detected both optically [5] and by resistive measurements citePellegrino2012,Yamasaki2014. If the microstructure is thermalized within the hysteretic temperature window of VO₂ (width of few degree), elec-

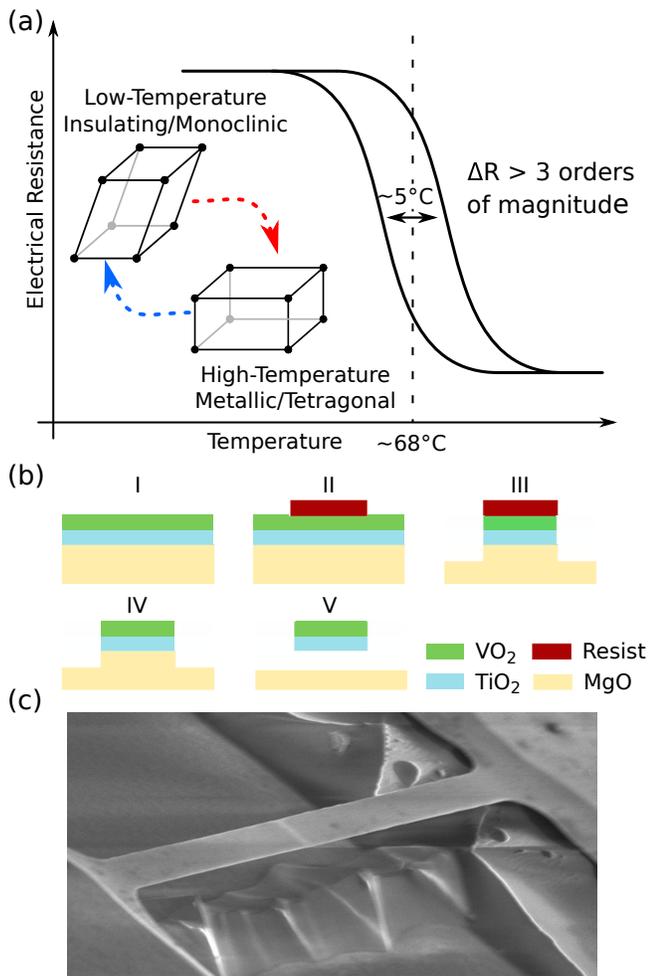


Figure 1. (a) Illustration of the phase transition in VO₂. Upon heating, at around 68 °C the electrical resistance drops by more than three orders of magnitude, concurrently with a change of lattice symmetry from monoclinic to tetragonal. (b) Fabrication steps of a VO₂ free-standing device. (c) Scanning Electron Microscope image of a VO₂-based micro-bridge ($100 \times 10 \mu\text{m}^2$).

trical current pulses can be employed to change the ratio between metallic and insulating VO₂ domains. The written state can be then erased by simply cooling the system below the hysteretic region [6]. The possibility to change progressively the resistive, optical and internal stress states of microstructures with memory capabilities offers intriguing perspectives for device engineering. The presence of hysteresis can be undesirable for applications concerning actuators. However, a self-sensing technique has been used by Merced et al. to achieve better control of electrothermal actuators based on VO₂ [7]. Another aspect is related to the design of the microstructures. Electrothermally-driven SSPT on a shaped microstructure depends on the thermal dissipation environment that has to be properly designed. For example, freestanding structures need much less electrical power for their heating with respect to the clamped

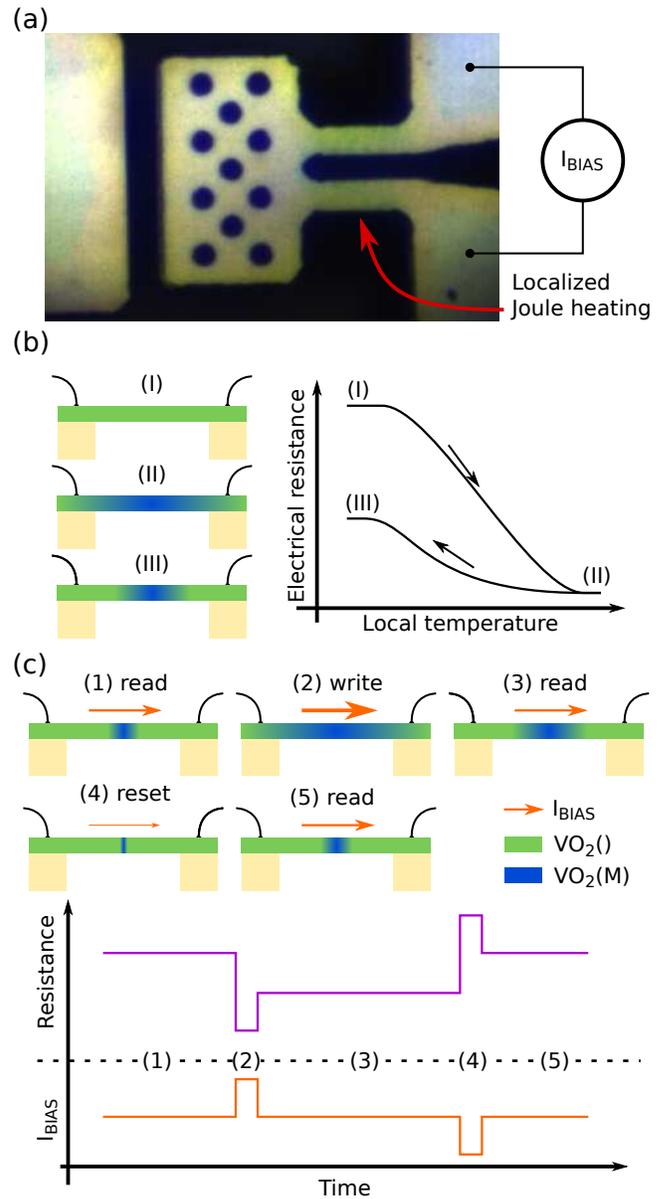


Figure 2. (a) Optical image of a VO-based cantilever. The current bias ($40 \mu\text{A}$) produces a local heating along the arms, triggering the phase transition (visible as a slight change of color). (b) Illustration of the “non-volatile” memory effect. When the sample temperature is within the hysteresis region, an electrical current determines the semi-permanent formation of metallic regions (blue) in the insulating (green) VO₂. (c) Illustration of the “volatile” memory effect. Current pulses of different amplitudes (orange arrows) modify the amount of metallic regions, that are then maintained by the heat provided by the “read” current.

ones [8] and geometrical shaping can be employed to concentrate Joule heating in targeted regions [9].

The way thermal flow is designed also affects the switching behavior of the device and its repeatability. SSPT of a microstructure (i.e. microbridge) under a progressive electrical bias can in fact occur smoothly or through sharp jumps or by a series of minor jumps. This

depends on the thermal boundary conditions that must be carefully taken into account when designing VO₂-based switching devices [5]. For the case of micromechanical devices, the magnitude of the mechanical actuation and the built-in strain state also depend on the crystallographic properties of the films that might be epitaxial, textured or polycrystalline, depending on the growing conditions. For these reasons, the use of heteroepitaxial growth on different substrates is a useful tool to tune the crystallographic properties of VO₂ films. In the next sections we describe two applications related to the manipulation of VO₂ domains with possible applications for the development of actuators.

Programmable MEMS mechanical resonator [9]

This device exploits confined Joule heating in order to change the ratio between metallic/tetragonal (M) VO₂ domains and insulating/monoclinic (I) ones. The microresonator consists of a cantilever geometry with narrow constrictions. In Figure 2(a) we show a VO₂ free-standing structure biased with 40 μ A. The balance between Joule heating and heat dissipation determines an increase of temperature along the arms, inducing a localized phase transition visible in Figure 2(a) as a dim green/blue coloration. In analogy to what has been shown in reference [6], the hysteresis of the SSPT gives the device itself a memory and thus multiple domain configurations can be prepared by proper choice of the current pulse that determines the amount of heating. We observe two kinds of memory effects, illustrated in Figures 2(b) and (c). The first occurs when a pristine structure, thermalized within the hysteresis windows (I), is heated by a current pulse. Depending on the magnitude of the electrical current, some VO₂ domain will switch from the I to the M phase, lowering the electrical resistance (II). Even after the current has been nullified, some metallic domains persist, resulting in a semi-permanent change of the resistance value (III). Subsequent current pulses of smaller magnitude do not perturb this configuration that can be erased only by cooling the system. We call this “non-volatile” memory effect. The other type of memory scheme is instead achieved by maintaining a constant supply current (of the order of few tens of microamperes) across the sample (always kept within the hysteretic temperature region (1)). In this case, current pulses of different magnitude (2) change the M/I configurations into multiple states that are stable until the supply current is maintained (3) and can be erased by nullifying this current for a short time (4-5). We called this process “volatile” memory effect. This effects can be detected both resistively, by measuring the electrical resistance of the microstructure and mechanically. In VO₂ bulk, at transition the *c*-axis is contracted by $\approx 0.8\%$. The *a* and *b* axis are instead elongated by $\approx 0.5\%$. In our deposited VO₂ films the SSPT produces in-plane stresses. Because

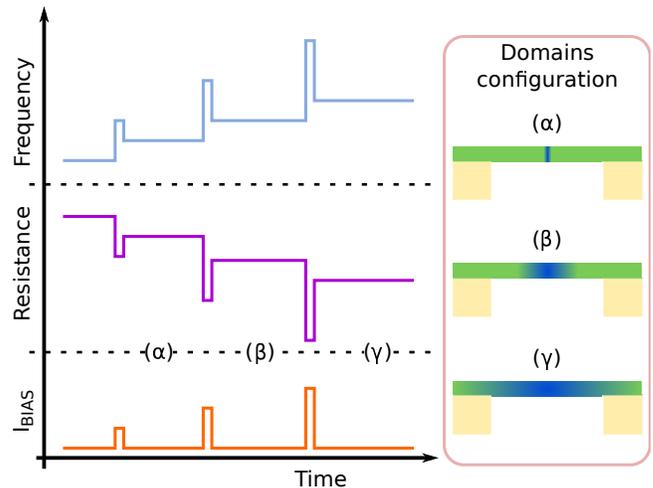


Figure 3. Multiple resistive and eigenfrequency states programmed by current pulses. Each state is selected by the current magnitude and corresponds to a different ratio between metallic and insulating areas.

of the crystallographic orientation of VO₂ films and the growth on TiO₂, the stress of the two orthogonal domains compensates, limiting the maximum achievable stress below the theoretical value expected for a VO₂ single crystal. The stress originating from different M/I configurations also produces bending of the microstructure. Stress-stiffening and stress-related deformation of the microstructure profoundly affect its mechanical resonance. The value of the mechanical eigenfrequency of the microstructure itself changes with temperature, increasing of about 40% when heating the whole sample above 68 °C. Similarly to what has been achieved with the realization of multiple resistive states, the progressive tuning of internal stress with diverse M/I configurations changes the mechanical eigenfrequency of the resonator in a reproducible manner. VO₂ can be thus employed to fabricate a resonator whose mechanical frequency is programmed into multiple and reversible states, as illustrated in Figure 3. This smart resonator may be part of more complex vibrating systems taking advantages of its simple memory capabilities.

A DC-powered mechanical oscillator [10]

The combination of non-linear electrical response and structural changes in a VO₂ micro-sized area has been employed to realize a novel high-frequency mechanical actuation scheme, showing direct conversion from a DC voltage to mechanical excitation of a freestanding microstructure in the MHz range. To this purpose, it is sufficient to apply a DC voltage bias (V_0) to a VO₂ element in series with a load resistor (R_0). The non-linear relationship between resistance, temperature and Joule heating in VO₂ can trigger an electrothermal instability, resulting in a periodic phase transition of the VO₂ element between the insulating and

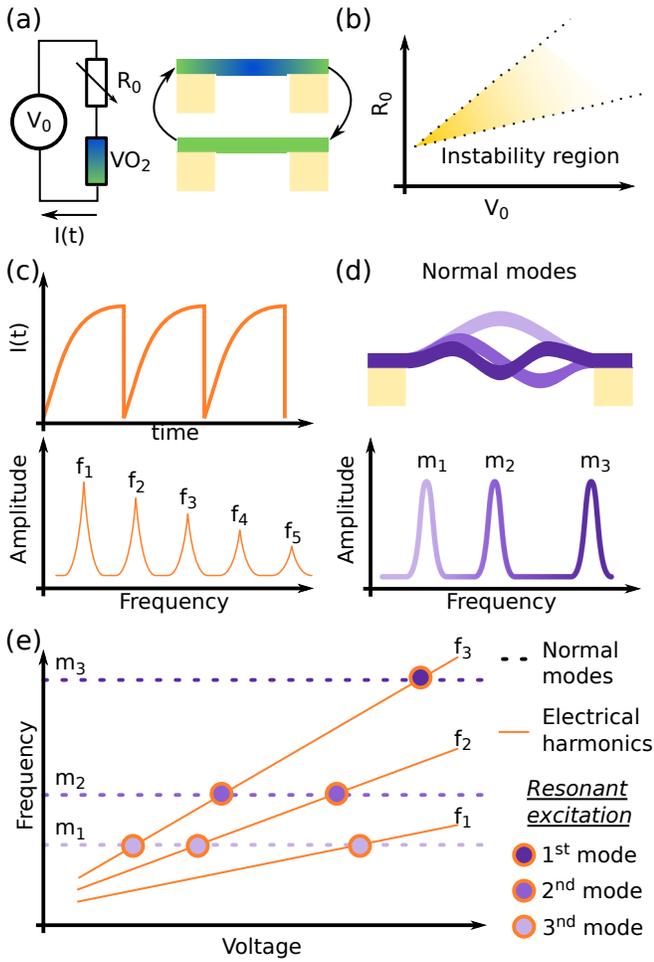


Figure 4. (a) Schematic of the circuit used to trigger the electrical oscillations (EO) and the corresponding periodic phase change in the microbridge. (b) Illustration of the space of parameters where the EO are self-sustained. (c) Time plot and power spectrum of the current flowing in the circuit (d) Mode shape and spectral response of the normal modes of a microbridge. (e) Working mechanism of the mode excitation. The harmonics of the EO shift with the voltage bias (solid orange lines), when one of them crosses one of the normal modes (dashed lines) resonant excitation occurs (dots).

metallic state (Figure 4(a)). Such instability can be triggered for specific combinations of V_0 and R_0 , as shown in Figure 4(b). The frequency of these electrical oscillations (EO) depends, in general, from the thermal and electrical boundary conditions. Practically, it can be controlled by tuning the value of V_0 and R_0 . As illustrated in Figure 4(c), the wave-shape of the EO is double-exponential, which is typical for self-oscillating non-linear systems (relaxation-oscillators). As a consequence, several evenly-spaced harmonic components constitute the signal. This is the key-element for this actuation scheme. If we trigger the EO using a VO_2 element with a micro-bridge geometry, these oscillations can couple with the normal modes of the structure. For this kind of geometry, the mechanical eigenfrequencies

correspond to the different flexural modes of the structure which are not evenly spaced in frequency, contrary to the power-spectrum of the EO (Figure 4(d)). It is thus possible to realize a single-matching condition between the harmonic components of the electrical oscillations and the mechanical modes of the microbridge to achieve selective resonant excitation. This is illustrated in Figure 4(e). By controlling one of the parameters of the oscillating circuit (in this case V_0), the values of the harmonics of the EO change (solid orange lines). If one of these components has the same value of one of the mechanical eigenfrequencies of the structure (dashed lines), the corresponding mechanical mode is excited. This scheme thus allows the selective excitation of different mechanical modes of the device, just by turning the knob of a DC voltage generator. This approach to resonant actuation could enable the design of simple and autonomous micro/nano electro-mechanical devices with integrated DC power sources, like solar cells or small batteries. Our scheme can be employed in a variety of fields of applications, like robotics, environmental monitoring or biomedical sensors. VO_2 -based microactuators are promising to the field of micro and nanoactuators, thanks to the above described properties and to the possibility to preserve and control the SSPT of this material down to submicrometric scale.

Acknowledgments

This research was supported by the Dutch Foundation for Fundamental Research on Matter (FOM), a Grant-in-Aid for Scientific Research A (No.26246013), a Grant-in-Aid for Scientific Research B (No.16H03871) from the Japan Society for the Promotion of Science (JSPS) and the Executive programme of cooperation between Italy and Japan by the Directorate General for Cultural and Economic Promotion and Innovation of the Ministry of Foreign Affairs and International Cooperation, of the Italian Republic.

- [1] J. Mohd Jani, M. Leary, A. Subic, and M. A. Gibson, *Mater. Des.* **56**, 1078 (2014).
- [2] K. Liu, C. Cheng, Z. Cheng, K. Wang, R. Ramesh, and J. Wu, *Nano Lett.* **12**, 6302 (2012).
- [3] K. Liu, C. Cheng, J. Suh, R. Tang-Kong, D. Fu, S. Lee, J. Zhou, L. O. Chua, and J. Wu, *Adv. Mater.* **26**, 1746 (2014).
- [4] E. Merced, X. Tan, and N. Sepúlveda, *Sensors Actuators A Phys.* **196**, 30 (2013).
- [5] N. Manca, L. Pellegrino, and D. Marré, *Appl. Phys. Lett.* **106**, 203502 (2015), arXiv:1702.00826.
- [6] L. Pellegrino, N. Manca, T. Kanki, H. Tanaka, M. Biasotti, E. Bellingeri, A. S. Siri, and D. Marré, *Adv. Mater.* **24**, 2929 (2012).
- [7] E. Merced, D. Torres, X. Tan, and N. Sepulveda, *J. Microelectromechanical Syst.* **24**, 100 (2015).

- [8] S. Yamasaki, T. Kanki, N. Manca, L. Pellegrino, D. Marré, and H. Tanaka, *Appl. Phys. Express* **7**, 023201 (2014).
- [9] N. Manca, L. Pellegrino, T. Kanki, S. Yamasaki, H. Tanaka, A. S. Siri, and D. Marré, *Adv. Mater.* **25**, 6430 (2013), [arXiv:1011.1669v3](https://arxiv.org/abs/1011.1669v3).
- [10] N. Manca, L. Pellegrino, T. Kanki, W. J. Venstra, G. Mattoni, Y. Higuchi, H. Tanaka, A. D. Caviglia, and D. Marré, *Adv. Mater.* **29**, 1701618 (2017), [arXiv:1704.06594](https://arxiv.org/abs/1704.06594).