

Optimizing VO₂ film properties for use in SAW devices

M. E. Kutepov^{*,§}, G. Ya. Karapetyan^{*}, I. V. Lisnevskaya[†],
K. G. Abdulvakhidov[‡], A. A. Kozmin^{*} and E. M. Kaidashev^{*}

^{*}Laboratory of Nanomaterials, Southern Federal University
Stachki, 200/1, 344090 Rostov-on-Don, Russia

[†]Department of Chemistry, Southern Federal University
Zorge, 7, 344090, Rostov-on-Don, Russia

[‡]Smart Materials Research Institute, Southern Federal University
Sladkova, 178/24, 344090, Rostov-on-Don, Russia

[§]kutepov.max@yandex.ru

Received 15 April 2021; Accepted 8 June 2021; Published 18 October 2021

The aim of this study is to optimize the conditions for producing thin VO₂ films for their use in SAW devices. We used the pulsed laser deposition (PLD) method to produce VO₂ films. We used a metallic vanadium target. The dependence of the oxygen pressure during PLD on the resistive metal–insulator transition (MIT) on substrates of *c*-sapphire and LiNbO₃ YX/128° was studied. The resulting films had sharp metal–insulator temperature transitions on *c*-Al₂O₃. The most high-quality films showed resistance change by four orders of magnitude. At the lower point of the hysteresis, the resistance of these samples was in the range of 50–100 Ω. The synthesized VO₂ films had a sharp temperature transition and a relatively small width of thermal hysteresis. The SAW damping during its passage through a VO₂/ZnO/LiNbO₃ YX/128° film at the metal–insulator phase transition temperature was studied. Attenuation SAW decreases with increasing temperature from 52 dB/cm to 0 dB/cm.

Keywords: Surface acoustic wave; VO₂ film; laser pulsed deposition; parameter S₂₁; metal–insulator transition.

1. Introduction

Vanadium dioxide (VO₂) exhibits a metal–insulator transition (MIT) at a temperature of about 69 °C.^{1,2} In this transition, the resistivity of VO₂ changes up to four orders of magnitude.³ The existence of the temperature, light, electrically or mechanically metal–insulator switch in vanadium dioxide (VO₂) at near-room temperature (~68 °C),⁴ combined with a short time of switching from the dielectric state to the metallic state (~26 fs),⁵ a sharp change in resistance (more than four orders of magnitude),⁶ a narrow width of the transition (about $\Delta T_c \sim 1^\circ$ in the best epitaxial films),⁷ the ability to change the capacity of thin-film structures with an electric voltage⁸ allows VO₂ to be considered as a promising material for use in controlled devices based on surface acoustic waves (SAW), broadband SAW photodetectors (from UV to IR range), controlled by light and electrically SAW couplers, switches and reflectors. Various methods such as magnetron sputtering^{11,12} sol–gel method,¹³ chemical vapor deposition,^{14,15} molecular beam epitaxy (MBE)^{16,17} and pulsed laser deposition (PLD)^{7,18,19} are used to make VO₂ thin films. Among all these methods, PLD is one of the most successful methods.

VO₂ films fabricated by PLD at different O₂ pressures on Al₂O₃ (0001) substrates were studied in Ref. 18. All films

exhibited thermal hysteresis behavior. The greatest change in resistance (~5 × 10³ times) was demonstrated by a film obtained at an oxygen pressure of 2.7 Pa. The film resistance varies from 7 × 10⁴ to 150 Ohm.

Similar studies were carried out by the authors of Ref. 19, using (1010) Al₂O₃ M-cut substrates, different deposition temperatures and different cooling times of the substrates after deposition. The smallest resistance was observed for samples that were synthesized at 630 °C and cooled for 1 and 3 h. The resistance of the samples varied from 2 × 10⁵ to 4.1 Ohm.

Quite similar results were obtained in Ref. 20 for VO₂ films fabricated by PLD, also at 630 °C on Al₂O₃ C (0001) and M (1010) cuts. The resistances of the samples varied approximately 10⁵ times from 4 × 10⁵ to 4 Ohm.

2. Synthesis of VO₂ Films

To create VO₂ films on a *c*-Al₂O₃ substrate, we used the method of laser pulsed deposition. A metal V target was placed at a distance of 5 cm from the substrate. Radiation from a KrF (248 nm, 10 Hz) laser was focused on the surface of a rotating target with a power density of 2.3 J/cm². The substrate temperature was maintained at (550–650 °C), and the oxygen atmosphere in the vacuum chamber was (3–6) × 10⁻² mbar.

[§]Corresponding author.

After the deposition of the films, the samples were kept in an O₂ atmosphere at 650 °C for 20 min, and then they were also cooled in an O₂ atmosphere.

The VO₂ layer was deposited by the PLD method from a metallic vanadium target using the above-described technique of *in situ* annealing of the preliminary layer. The first thin layer of VO₂ was deposited in 700 laser pulses with a laser pulse repetition rate of 5 Hz. The substrate temperature was 620 °C, and the O₂ partial pressure was 5×10^{-2} mbar. Then, *in situ* annealing was performed for 5 min. P (O₂) was 5×10^{-2} mbar, substrate temperature was 620 °C. Then, *in situ* annealing was performed for 5 min. P (O₂) was 5×10^{-2} mbar, the substrate temperature was 620 °C. Then the main VO₂ layer was deposited (laser pulse repetition rate 10 Hz, substrate temperature 620 °C, oxygen pressure 5×10^{-2} mbar). After the deposition of the main layer, one more *in situ* annealing was carried out without changing the O₂ pressure and temperature for 10 min. Then the sample was cooled to a temperature of ~150 °C in an oxygen atmosphere of 5×10^{-2} mbar.

3. Results and Discussion

3.1. Structural properties of VO₂ films

The X-ray diffraction (XRD) measurements were carried out using an ARL XTRA diffractometer with CuK α -radiation at 35 kV and 30 mA. The scan step was 0.02°. The XRD pattern of a typical VO₂ film deposited on *c*-Al₂O₃ substrate shown in Fig. 1 reveals peaks at 39.9° and 86° which correspond to (020) and (040) reflexes of monoclinic VO₂ phase. No other peaks were observed which claims that the films are epitaxial with (010) planes oriented parallel to *c*-Al₂O₃ substrate. FWHM of (002) and (040) peaks were measured to be as narrow as 0.18° and 0.20°, correspondingly, which claims on a high crystalline quality of prepared films. The unit cell

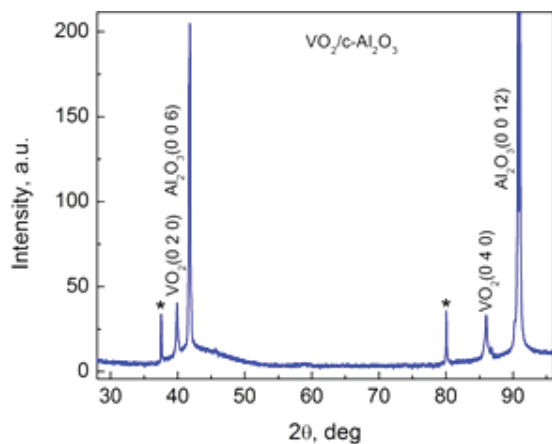


Fig. 1. XRD pattern of VO₂ film on *c*-Al₂O₃ substrate deposited at 650 °C and oxygen pressure of $6 \cdot 10^{-2}$ mbar; *denote parasite peaks of a sapphire substrate.

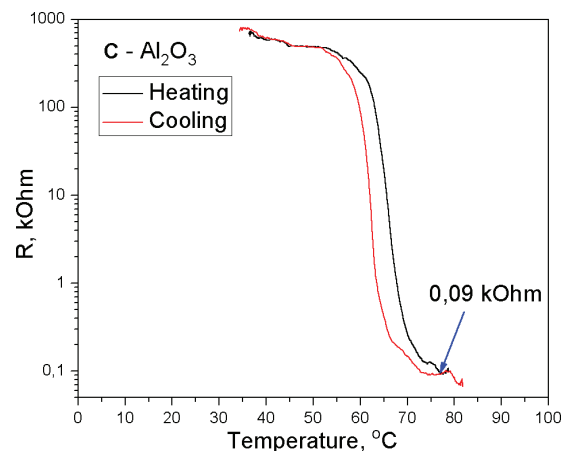


Fig. 2. Dependency of resistivity from temperature in VO₂ films on *c*-Al₂O₃.

parameter *c* calculated from the positions of (020) and (040) reflexes is 0.4521 nm which is slightly different from *c* = 0.4532 nm for monoclinic VO₂ powder sample, exhibiting minor compressive stresses due to film and substrate lattice parameters mismatch.

3.2. Study of temperature induced resistance alteration in VO₂ films

The most high-quality VO₂ films on *c*-Al₂O₃ showed a change in resistance by four orders of magnitude. At the lower hysteresis point the resistance of these samples was in the range (40–100) Ohms (Fig. 2). It should be noted that our films exhibit a rather sharp transition and a relatively small hysteresis width, which indicates the high quality of the obtained material.

For VO₂ films on a LiNbO₃ YX/128° piezoelectric substrate with a buffer ZnO sublayer, the temperature dependence of the resistance is shown in Fig. 3.

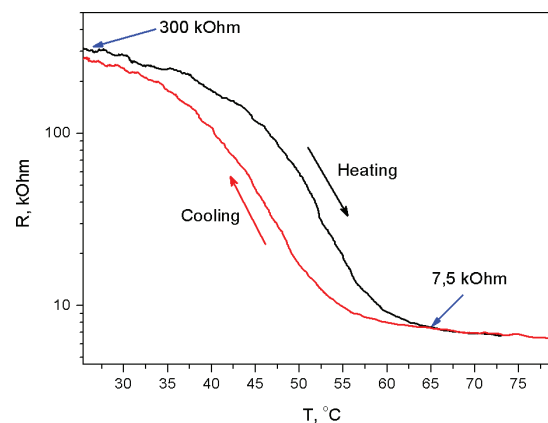


Fig. 3. Dependency of resistivity from temperature in VO₂ on LiNbO₃ YX/128° substrate with ZnO buffer layer.

At room temperature, the resistance of the VO₂ film was ~300 kOhm, and at a temperature of 65 °C, it was 7.5 kOhm.

4. SAW Measurements

To study the attenuation of a surface acoustic wave (SAW) when passing through a thin VO₂ film in the temperature range of the metal-semiconductor phase transition, a special stand was made, the design of which is shown in Fig. 4. It contains two YX/128° cut lithium niobate substrates, each of which has a unidirectional IDT with internal reflectors⁹ at a central frequency of 120.7 MHz. The ends of the substrates are rounded so that a SAW absorber can be applied to them. IDTs contain 17 unidirectional sections with an aperture of 1.2 mm. The substrate, which is placed on top, provides the transition of the SAW from one position with one IDT to another substrate with another IDT. This is due to the fact that SAWs are accompanied by an electric field and excite a SAW in the upper piezoelectric substrate. A vanadium dioxide film is deposited on the surface of the upper substrate. The absence of IDTs on this substrate significantly expands the possible parameters of the synthesis of the vanadium oxide film and makes it possible to study the vanadium oxide film immediately after its deposition. Next, the SAWs by their electric field excite the SAWs in another substrate, on which one more IDT is located. Piezoelectric substrate without IDT, but with a film of vanadium oxide is pressed against piezoelectric substrates with IDT using a clamping screw, through a heater. S-parameters were recorded by using a network analyzer OBZOR 304/1. The electromagnetic signal is converted into a SAW using an IDT, then the SAWs propagate to another IDT, passing first to a piezoelectric substrate with a vanadium oxide film, and then to another substrate with an IDT. In this case, the distance traveled by the SAW is 58 mm, which corresponds to a delay of 14.28 μs.

Figure 5 shows the change in the amplitude–frequency characteristic of SAW S₂₁ when the sample is heated from 30 to 60 °C. It can be seen that at a temperature of 30 °C, the SAW attenuation is 17 dB greater than at 60 °C. This is due

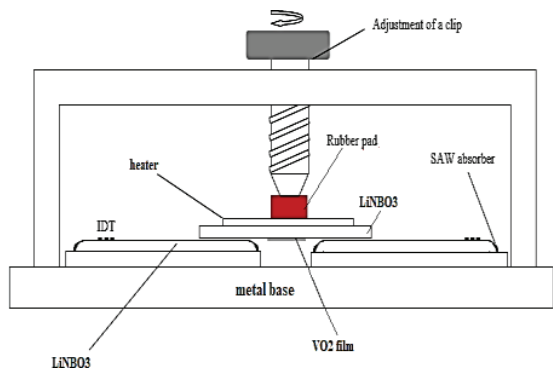


Fig. 4. Experimental setup for measuring the attenuation of a surfactant in a vanadium oxide film.

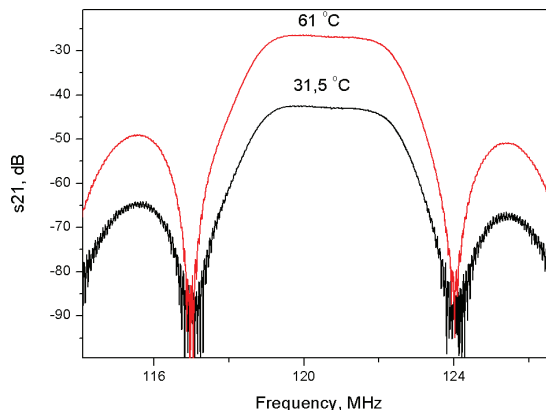


Fig. 5. Change in the amplitude–frequency characteristic of SAW S₂₁ when the sample is heated from 31.5 °C to 60 °C.

to the fact that at a temperature of 30 °C the resistance of the film is about 300 kΩ, while the film is in a semiconducting state. At the same time, at 60 °C, the film resistance is less than 10 kOhm and it is in a state close to metallic. Therefore, the SAW, passing under the film at 30 °C, experiences acoustoelectronic interaction^{20,21} with charge carriers in the film, which leads to a significant acoustoelectronic SAW. When the film is in a state close to metallic, it screens the electric field of the SAW and the acoustoelectronic interaction is small. In this case, acoustoelectronic damping is also small. Figure 6 shows the dependence of the SAW decay at temperatures near the metal-semiconductor VO₂ phase transition. Such dependence was constructed from the frequency dependences of the parameter S₂₁, taken at different temperatures, both in the direction of increasing the temperature and in the direction of decreasing it. In this case, the SAW attenuation was measured by the maximum amplitude–frequency characteristic of the parameter S₂₁ at a frequency of 120.7 MHz, taking into account the length of the VO₂ film equal to 3 mm.

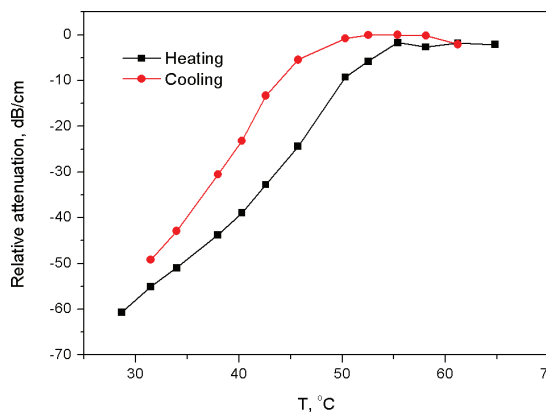


Fig. 6. Changes in the attenuation of a SAW when passing through a thin VO₂ film in the temperature range of the metal-semiconductor phase transition.

5. Conclusions

An investigation was made of vanadium dioxide films obtained by PLD from a metal target. A narrow semiconductor-metal junction, a large change in resistance (change by four orders of magnitude) and a low resistance value at the end of the junction (90 Ohm) determine the prospects for using VO₂ films on c-Al₂O₃ in impedance-dependent SAW devices.¹⁰ A significant change in SAW attenuation (~19 dB) in VO₂ films on piezoelectric substrates at semiconductor-metal phase transition temperatures determines the prospects for using VO₂ in temperature-controlled or light-controlled SAW devices.

Acknowledgment

This research work is supported by Southern Federal University, project No.07/2020-06-MM.

References

- ¹N. Bahlawane and D. Lenoble, Vanadium oxide compounds: Structure, properties, and growth from the gas phase, *Chem. Vap. Depos.* **20**, 299 (2014).
- ²F. J. Morin, Oxides which show a metal-to-insulator transition at the Neel temperature, *Phys. Rev. Lett.* **3**, 34 (1959).
- ³D. P. Partlow, S. R. Gurkovich, K. C. Radford and L. J. Denes, Switchable vanadium-oxide films by a sol-gel process, *J. Appl. Phys.* **70**, 443 (1991).
- ⁴Y. Ke, S. Wang, G. Liu, M. Li, T. J. White and Y. Long, Vanadium Dioxide: The multistimuli responsive material and its applications, *Small* **14**, 39 (2018).
- ⁵M. F. Jager, C. Ott, P. M. Kraus, C. J. Kaplan, W. Pouse, R. E. Marvel, R. F. Haglund, D. M. Neumark and S. R. Leone, Tracking the insulator-to-metal phase transition in VO₂ with few-femtosecond extreme UV transient absorption spectroscopy, *Proc. Natl. Acad. Sci. USA* **114**, 36 (2017).
- ⁶M. Borek, F. Qian, V. Nagabushnam and R. K. Singh, Pulsed laser deposition of oriented VO₂ thin films on R-cut sapphire substrates, *Appl. Phys. Lett.* **63**, 24 (1993).
- ⁷D. H. Kim and H. S. Kwok, Pulsed laser deposition of VO₂ thin films, *Appl. Phys. Lett.* **65**, 3188 (1994).
- ⁸V. S Aliev, S. G. Bortnikov and I. A. Badmaeva, Anomalous large electrical capacitance of planar microstructures with vanadium dioxide films near the insulator-metal phase transition, *Appl. Phys. Lett.* **104**, 13 (2014).
- ⁹G. Y. Karapetyan, V. E. Kaydashev, D. A. Zhilin, T. A. Minasyan, K. G. Abdulvakhidov and E. M. Kaidashev, Use of multiple acoustic reflections to enhance SAW UV photo-detector sensitivity, *Smart Mater. Struct.* **26**, 035029 (2017).
- ¹⁰G. Ya. Karapetyan, V. E. Kaydashev, D. A. Zhilin, M. E. Kutepov, T. A. Minasyan and E. M. Kaidashev, A surface acoustic wave impedance-loaded high sensitivity sensor with wide dynamic range for ultraviolet light detection, *Sens. Actuat. A* **296**, 70 (2019).
- ¹¹A. Lafort et al., Optical properties of thermochromic VO₂ thin films on stainless steel: Experimental and theoretical studies, *Thin Solid Films* **519**, 3283 (2011).
- ¹²J. B. K. Kana et al., Thermochromic VO₂ thin films synthesized by rf-inverted cylindrical magnetron sputtering, *Appl. Surf. Sci.* **254**, 3959 (2008).
- ¹³T. Driscoll, H. T. Kim, B. G. Chae, M. Di Ventra and D. N. Basov, Phase-transition driven memristive system, *Appl. Phys. Lett.* **95**, 043503 (2009).
- ¹⁴S. Mathur, T. Ruegamer and I. Grobelsek, Phase-selective CVD of vanadium oxide nanostructures, *Chem. Vap. Depos.* **13**, 42 (2007).
- ¹⁵M. B. Sahana, M. S. Dharmaprasanth and S. A. Shivashankar, Microstructure and properties of VO₂ thin films deposited by MOCVD from vanadyl acetylacetonate, *J. Mater. Chem.* **12**, 333 (2002).
- ¹⁶L. Dillemans, R. R. Lieten, M. Menghini, T. Smets, J. W. Seo and J. P. Locquet, Correlation between strain and the metal-insulator transition in epitaxial V₂O₃ thin films grown by molecular beam epitaxy, *Thin Solid Films* **520**, 4730 (2012).
- ¹⁷A. D. Rata, A. R. Chezan, M. W. Haverkort, H. H. Hsieh, H. J. Lin, C. T. Chen, L. H. Tjeng and T. Hibma, Growth and properties of strained VO_x thin films with controlled stoichiometry, *Phys. Rev. B* **69**, 075404 (2004).
- ¹⁸S. Fan, L. Fan, Q. Li, J. Liu and B. Ye, The identification of defect structures for oxygen pressure dependent VO₂ crystal films, *Appl. Surf. Sci.* **321**, 464 (2014).
- ¹⁹B. J. Kim, Y. W. Lee, S. Choi, B. G. Chae and H. T. Kim, Analysis of the surface morphology and the resistance of VO₂ thin films on M-plane Al₂O₃, *J. Korean Phys. Soc.* **50**, 653 (2007).
- ²⁰M. E. Kutepov, V. E. Kaydashev, G. Y. Karapetyan, T. A. Minasyan, A. V. Chernyshev, K. G. Abdulvakhidov, S. I. Shevtsova, E. V. Glazunova, V. A. Irkha and E. M. Kaidashev, Deep UV light sensitive Zn_{1-x}-yMg_xAl_yO films with fast photoelectric response for SAW photodetectors, *Smart Mater. Struct.* **28**, 065024 (2019).
- ²¹R. R. Hemphill, Effect of a thin-film phase-transition material on surface acoustic wave propagation, *IEEE Ultrason. Symp.* (Dallas, TX, USA, 1984), pp. 1006–1010.