Optical limiting sensor based on multilayer optimization of Ag/VO₂ phase changing material

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Abstract— An optical limiting sensor working in the infrared was developed to address the need for eye and sensor protection against laser threats. Metallic and dielectric photonic resonators (thin-film multilayers) incorporating phase-change-materials (PCM) like VO₂ were simulated and experimentally realized, with optimization of the deposition procedure by RF magnetron sputtering at low temperature. For the first time, the silver is placed between the substrate and the VO₂, thus improving the device limiting performances. By maximizing the difference of transmittance between the 'ON' and the 'OFF' states at the standard light wavelength for telecom applications (1550 nm), we calculated optimum thickness for VO₂ and Ag films. The deposited thin films were characterized by SEM and Raman spectroscopy, and VO₂ transition temperature was investigated by measuring resistance changes.

As a proof of concept of the device working principle, we calculated transmittance drop of 70% when the sensor is exposed to a laser pulse excitation (20ps, 500 MW/cm²). Our results pave the way for multilayer with optical limiting properties.

Index Terms— light intensity sensor; magnetron sputtering; optical limiting; phase-changing material;VO₂.

I. INTRODUCTION

Infrared (IR) light at high power can cause irreversible injury to the human eye. On the contrary, low power IR radiation is not harmful to the eyesight. High power lasers are widespread nowadays in several fields like spectroscopy, medical applications, production (laser cutting), and defence. It is important to take precautions because infrared light is invisible, so the eyes will not take the protective measures like blinking or closing when a high-intensity beam of infrared radiation shines into them. It is thus of utmost importance to develop a threshold sensor for high optical power that can sense it and switch to non-transmitting state preventing eyes from damaging. An additional application is the protection of detectors in power meters.

In the present work we designed a dynamic device [1] whose transmittance depends on the intensity of the incident radiation, based on the use of a phase changing material (PCM), namely vanadium dioxide (VO₂). The latter can be grown by different techniques [2], among those sputtering is one of the most industrially applicable [3], [4], [5], [6]. We investigated the condition to obtain the VO₂ phase by room temperature deposition and subsequent annealing.

VO₂ is a material endowed with interesting properties. It undergoes a reversible phase transition from monoclinic to tetragonal phase at a temperature around the Insulator to Metal Transition temperature T_{IMT} [5]. When the temperature is low it is in the insulating state, while when the temperature exceeds the transition temperature it is metallic with high reflectance in the IR. The transition can be induced in several ways, by temperature increase and by optical absorption [7], as schematized in Fig. 1. In particular, the former scenario is depicted in Fig 1 (a), in which a device made by a VO₂ thin film transmits all the incident light when the temperature is lower than the T_{IMT}; instead, when the temperature is higher than T_{IMT}, the VO₂ convert to metal and the light is reflected. Fig 1(b) shows the optical limiting concept: the absorption of a very intense light yields a temperature increase, exceeding T_{IMT}, and induces the switch of the device to a low transmission state. In the latter case, a silver (Ag) layer is added to increase the infrared radiation absorption inside the VO₂, thus obtaining light-induced thermal switching of the VO₂ layer.

COMSOL Multiphysics simulations in conjunction with a transfer matrix approach allow to perform the geometrical optimization and the theoretical characterization of the device. The threshold intensity

at which the phase transition in VO_2 occurs strongly depends on the design: at variance with Ref. [7], the silver is placed between the substrate and the VO_2, thus maximizing the device limiting performances. We optimized the deposition condition to obtain a multilayer of Ag and VO_2 films by using magnetron sputtering, also considering the influence of different substrates (fused silica and sapphire).
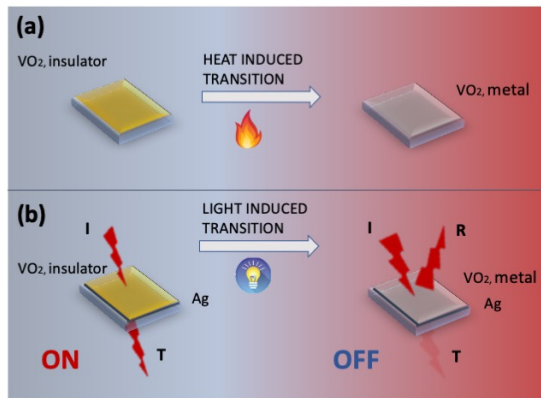

Fig. 1. Concept of optical limiting sensor. (a) VO_2 can be converted from insulator to metal by heating above T_{IMT} (b) By using an excitation with pulsed laser in the IR, at proper value of peak intensity and pulse duration, the infrared absorption by the device raises the temperature in the VO_2 above T_{IMT} and the transmittance drops, so the device is in the OFF state. The sensor is reversible, coming back in the ON state when the stimulus is removed.

II.   METHODS

A.   Device realization

Thin films multilayers were deposited by a modular sputtering system equipped with RF and DC generators (Advanced Energy). VO_2 thin film was grown starting from a pure vanadium target, with RF magnetron sputtering in oxidizing conditions. The Oxygen/Ar ratio during deposition was 50% and the substrates were kept at room temperature. Glass and sapphire substrate were tested and the influence on VO_2 phase analysed. To realize the optical sensor, a thin Ag film was deposited by DC magnetron sputtering between the substrate and the VO_2 to control the incident intensity threshold power at which the switching occurs. Annealing procedure was optimized to achieve the correct crystalline monoclinic phase: air or vacuum from a primary pump were investigated as annealing atmospheres.

PCM phase and morphology were investigated by Raman Spectroscopy (RS) and Field Emission Scanning Electron Microscope (FE-SEM) Leo 1525. For RS a modular micro-Raman confocal system from Horiba with laser excitation at 532 nm, 100X objective and 1800 l/mm grating was used.

B.   Simulation and test

Optimization of the geometrical parameters was carried out employing a transfer matrix approach, where the whole VO_2 layer was assumed to be in its metallic or dielectric phase, corresponding to 'OFF' and 'ON' state, respectively. The theoretical characterization was achieved with transient opto-thermal simulations performed with COMSOL Multiphysics software [8], [9], [10], [11].

For testing the sensor performances, a home-built microscopy setup integrated with a tunable pulsed laser from an optical parametric oscillator was used. The transmission through the samples is recorded both on a power meter and on an InGaAs camera.

III.   RESULTS AND DISCUSSIONS

A.   Simulations for geometrical optimization.

Before the experimental deposition of the multilayer, the device simulation was carried out to optimize the geometrical parameters (thickness of Ag and VO_2 layer) in order to have maximum change in transmittance $\Delta Tr = Tr_{diel} - Tr_{met}$, where Tr_{diel} (Tr_{met}) is the transmittance in the dielectric (metallic) state. All calculations were made at fixed wavelength at 1550 nm (value of interest for telecom applications), using T_{IMT} taken from literature [12]. A transfer matrix approach was implemented assuming that the whole VO_2 layer was assumed to be in its metallic or dielectric phase. For the optimal configuration, a transmittance drop of more than 70% is expected when the device is in the 'OFF' state. The optimum thickness obtained for Ag/VO_2 are 5/52 nm respectively.

B.   Sensor characterization.

SEM microscopy revealed the morphology of the single and multilayer film (Fig. 2). Polycrystalline structure can be observed for VO_2 thin film - Fig.2 (a) - many rounded particles with typical dimensions between 150 and 250 nm coalesce in a brain-shaped fashion. For multilayer film Ag/ VO_2 - Fig. 2 (b) - the mean diameter of the observed particles is much smaller, being around 100 nm. Very porous structuring of the surface is observable at higher magnification. Despite the observed strong influence of the Ag layer on the VO_2 film morphology, the correct phase was observed both for VO_2 and for Ag/VO_2 multilayer, as confirmed by Raman Spectroscopy.
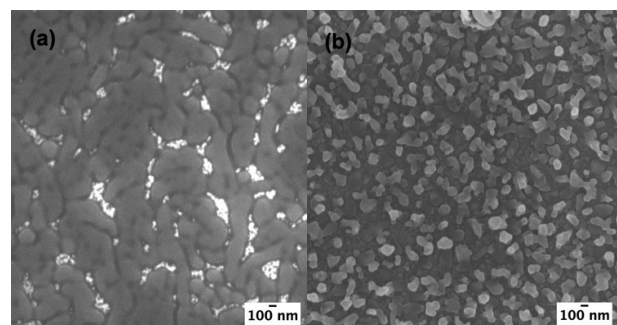

Fig.2. SEM images of the (a) VO_2 thin film compared to (b) Ag/VO_2 thin films acquired at 50kX magnification. Grain size observed in VO_2 thin film is much bigger that the one of the Ag/VO_2 multilayer (100nm).

Raman Spectroscopy was useful to characterize the quality of produced thin films. It is well-known that the V_2O_5 phase is unwanted

and the procedural window to obtain the pure VO_2 phase is quite narrow. Reactive sputtering is needed when starting from a metal target, followed by annealing at 550°C in vacuum. Air atmosphere during annealing produced the unwanted V_2O_5 phase. Influence of the substrate for deposition is shown in Fig. 3, by comparing thin films deposited on sapphire and glass substrate. Lattice vibrations typical of the VO_2 (M1) insulating phase [13], [14] can be observed for the thin film deposited on sapphire substrate (except for the peak of the sapphire substrate, visible at 417 cm^{-1}). On the contrary, when a glass substrate was used, the correct phase was not achieved, and broad peaks of V_2O_5 appears superposed to amorphous band of the glass substrate. VO_2 (M1) phase is achieved as well for multilayer Ag/VO_2.

To investigate the T_{IMT}, we placed the device on a Peltier cell (Linkam THMS600) for controlled temperature change. The Raman-
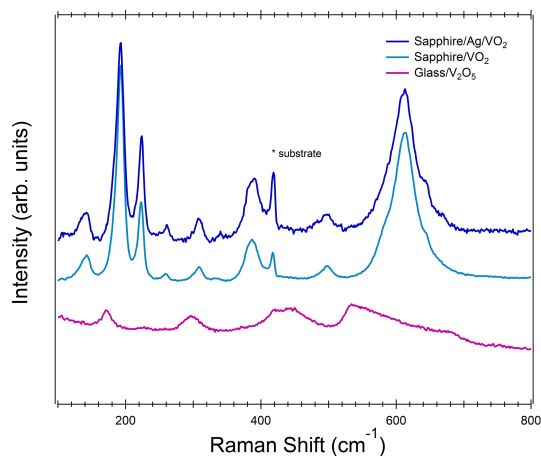

Fig. 3. Raman spectra acquired on vanadium oxide layers for different substrates: for sapphire substrate both VO_2 and Ag/VO_2 layers (blue and light blue lines) presents only vibrations from VO_2, (M) phase, and sapphire substrate; (glass-magenta line) the phase obtained on glass substrate is poorly crystalline, with few peaks ascribable to V_2O_5.

active phonon modes of the VO_2 disappear across the semiconductor–metal phase transition because the unit cell of VO_2 changes to the rutile phase. The transition was tested also by measuring the resistivity of the Ag/VO_2, confirming that T_{IMT}=65°C during the heating cycle and T_{IMT}=59°C during the cooling cycle. The cycle is fully reversible and was repeated several times with no observable changes. Moreover, stability tests performed over samples kept in ambient air at room temperature for up to 4 months showed no variation in the phase by Raman spectroscopy.

The insulator to metal transition was confirmed by transmittance measurements performed at 1550 nm for VO_2 and Ag/VO_2 sample, with calculation of the hysteresis cycle.

C. Simulations to investigate the thermal dynamics.

We studied the transient response of the sensor device when illuminated by a laser pulse of duration 20 ps and peak intensity 500 MW/cm^2 (details of the modelling are reported in [15] for Au/VO_2 multilayer). Fig. 4 reports the transient response of the sensor. As we can see, after the pulse arrival (occurring at 200 ps) the temperature in the VO_2 layer (blue curve) increases and overcomes the critical
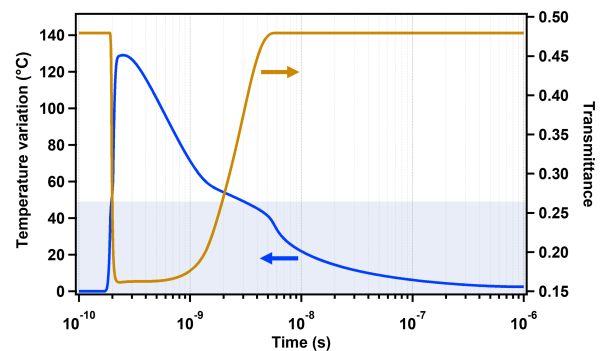

Fig. 4. Simulated temperature variation (blue) and transmittance (brown) temporal dynamics following the excitation from a single laser pulse (1550 nm, 20 ps FWHM, peak intensity 500 MW/cm^2). The shaded area represents the region with temperature lower than T_{IMT}. The device undergoes a change of optical properties only when $T>T_{IMT}$.

value, hence the transmittance (brown curve) drops from 0.47 to 0.15. The response time of the layer, which is the time interval necessary to heat the sensor to the maximum temperature, is the same as the pulse duration (20 ps at FWHM). Afterwards, the device cools down and after 3 ns the temperature becomes lower than the critical value, hence restoring the high transmittance state. The complete cooling occurs on longer time scales, as shown in the Fig. 4. We noticed that the T_{IMT}, and thus the OFF state, is reached for the high intensity source. In the case of a low intensity light, the equilibrium temperature does not exceed the critical one and thus the sensor is not activated.

IV. CONCLUSION

We designed and realized a planar multilayer sensor with vanadium dioxide to achieve a self-activating optical limiter with ON-OFF behaviour. The mechanism is activated by the temperature increase induced by high power laser in the IR (1550 nm).

After the geometrical optimization of the device to maximize the change of the transmitted power, we obtained a reduction up to 70% after transition in the OFF state. The device was realized by magnetron sputtering deposition of multilayer Ag/VO_2. The characterization via Raman spectroscopy and SEM allowed to obtain an optimized device that shows good agreement with the theoretical predictions. Our results open the opportunity for novel optical sensor with optical limiting properties.

ACKNOWLEDGMENT

REFERENCES

[1] L. Carletti *et al.*, "Reconfigurable nonlinear response of dielectric and semiconductor metasurfaces," *Nanophotonics*, vol. 10, no. 17, pp. 4209–4221, 2021, doi: 10.1515/nanoph-2021-0367.

[2] T. C. Chang, X. Cao, S. H. Bao, S. D. Ji, H. J. Luo, and P. Jin, "Review on thermochromic vanadium dioxide based smart coatings: from lab to commercial application," *Adv. Manuf.*, vol. 6, no. 1, pp. 1–19, 2018, doi: 10.1007/s40436-017-0209-2.

[3] S. S. Maklakov *et al.*, "Post-deposition annealing of thin RF magnetron sputter-deposited VO2 films above the melting point," *J. Alloys Compd.*, vol. 763, pp. 558–569, 2018, doi: 10.1016/j.jallcom.2018.06.014.

[4] D. Ruzmetov, K. T. Zawilski, V. Narayanamurti, and S. Ramanathan, "Structure-functional property relationships in rf-sputtered vanadium dioxide thin films," *J. Appl. Phys.*, vol. 102, no. 11, 2007, doi: 10.1063/1.2817818.

[5] H. Y. Xu, Y. H. Huang, S. Liu, K. W. Xu, F. Ma, and P. K. Chu, "Effects of annealing ambient on oxygen vacancies and phase transition temperature of VO$_2$ thin films," *RSC Adv.*, vol. 6, no. 83, pp. 79383–79388, 2016, doi: 10.1039/c6ra13189a.

[6] C. Baratto, "Growth and properties of ZnO nanorods by RF-sputtering for detection of toxic gases," *RSC Adv.*, vol. 8, no. 56, pp. 32038–32043, 2018, doi: 10.1039/c8ra05357j.

[7] Y. Wang *et al.*, "Electrical tuning of phase-change antennas and metasurfaces," *Nat. Nanotechnol.*, vol. 16, no. 6, pp. 667–672, 2021, doi: 10.1038/s41565-021-00882-8.

[8] B. Li *et al.*, " Fundamental limits for transmission modulation in VO 2 metasurfaces ," *Photonics Res.*, vol. 11, no. 1, p. B40, 2023, doi: 10.1364/prj.474328.

[9] D. De Ceglia *et al.*, "Transient guided-mode resonance metasurfaces with phase-transition materials," *Opt. Lett.*, vol. 48, no. 11, pp. 2961–2964, 2023.

[10] M. Celebrano *et al.*, "Optical tuning of dielectric nanoantennas for thermo-optically reconfigurable nonlinear metasurfaces," *Opt. Lett.*, vol. 46, no. 10, p. 2453, 2021, doi: 10.1364/ol.420790.

[11] A. Ronchi *et al.*, "Light-Assisted Resistance Collapse in a V2 O3 -Based Mott-Insulator Device," *Phys. Rev. Appl.*, vol. 15, no. 4, p. 1, 2021, doi: 10.1103/PhysRevApplied.15.044023.

[12] C. Wan *et al.*, "On the Optical Properties of Thin-Film Vanadium Dioxide from the Visible to the Far Infrared," *Ann. Phys.*, vol. 531, no. 10, pp. 1–7, 2019, doi: 10.1002/andp.201900188.

[13] M. Pan *et al.*, "Raman study of the phase transition in VO2 thin films," *J. Cryst. Growth*, vol. 268, no. 1–2, pp. 178–183, 2004, doi: 10.1016/j.jcrysgro.2004.05.005.

[14] S. S. Majid, A. Ahad, F. Rahman, V. Sathe, and D. Shukla, "Unveiling the Role of VO2 (B) Polymorph in the Insulator–Metal Transition of VO2 (M1) Thin Films," *Phys. Status Solidi Basic Res.*, vol. 259, no. 10, pp. 1–6, 2022, doi: 10.1002/pssb.202200108.

[15] A. Tognazzi *et al.*, " Opto-thermal dynamics of thin-film optical limiters based on the VO 2 phase transition ," *Opt. Mater. Express*, vol. 13, no. 1, p. 41, 2023, doi: 10.1364/ome.472347.