

Experimental demonstration of tunable phase in a thermochromic infrared-reflectarray metamaterial

D.J. Shelton^{1,*}, K.R. Coffey², G. D. Boreman^{1,2}

¹CREOL, University of Central Florida, 4000 Central Florida Blvd., Orlando FL 32816-2700, USA

²AMPAC, University of Central Florida, 4000 Central Florida Blvd., Orlando FL 32816-2700, USA

* dshelton@creol.ucf.edu

Abstract: For the first time, a tunable reflected phase reflectarray is demonstrated in the thermal infrared. This is done using thermochromic VO₂ square-patch elements in a reflectarray metamaterial configuration. A sixty degree change in reflected phase is measured using a Twyman-Green interferometer, and FTIR measurements show that the resonance reflection minima shifts from 9.2 to 11.2 μm as the sample is heated from 45 through 65 °C. These results are in agreement with finite-element method simulations using the optical properties of VO₂ which are measured by infrared ellipsometry.

©2010 Optical Society of America

OCIS codes: (160.3918) Metamaterials; (310.6845) Thin film devices and applications.

References and links

1. D. Shelton, J. Ginn, and G. Boreman, "Bandwidth variations in conformal infrared frequency selective surfaces," *IEEE Antennas Propag. International Symposium*, 3976 (2007).
2. J. Ginn, D. Shelton, P. Krenz, B. Lail, and G. Boreman, "Altering infrared metamaterial performance through metal resonance damping," *J. Appl. Phys.* **105**(7), 074304 (2009).
3. B. Monacelli, J. Pryor, B. A. Munk, D. Kotter, and G. Boreman, "Infrared frequency selective surface based on circuit-analog square loop design," *IEEE Trans. Antenn. Propag.* **53**(2), 745–752 (2005).
4. W. J. Padilla, A. J. Taylor, C. Highstrete, M. Lee, and R. D. Averitt, "Dynamical electric and magnetic metamaterial response at terahertz frequencies," *Phys. Rev. Lett.* **96**(10), 107401 (2006).
5. J. Ginn, B. Lail, J. Alda, and G. Boreman, "Planar infrared binary phase reflectarray," *Opt. Lett.* **33**(8), 779–781 (2008).
6. J. Tharp, B. Lail, B. Munk, and G. Boreman, "Design and demonstration of an infrared meanderline phase retarder," *IEEE Trans. Antenn. Propag.* **55**(11), 2983–2988 (2007).
7. E. Cubukcu, S. Zhang, Y. S. Park, G. Bartal, and X. Zhang, "Split ring resonator for infrared detection of single molecular monolayers," *J. Appl. Phys.* **95**, 043113 (2009).
8. S. Zhang, W. Fan, N. C. Panoiu, K. J. Malloy, R. M. Osgood, and S. R. J. Brueck, "Experimental demonstration of near-infrared negative-index metamaterials," *Phys. Rev. Lett.* **95**(13), 137404 (2005).
9. D. F. Sievenpiper, J. H. Schaffner, H. J. Song, R. Y. Loo, and G. Tangonan, "Two-dimensional beam steering using an electrically tunable impedance surface," *IEEE Tran. Ann. Prop.* **51**(10), 2713–2722 (2003).
10. T. Driscoll, H. T. Kim, B. G. Chae, B. J. Kim, Y. W. Lee, N. M. Jokerst, S. Palit, D. R. Smith, M. Di Ventra, and D. N. Basov, "Memory metamaterials," *Science* **325**(5947), 1518–1521 (2009).
11. M. J. Dicken, K. Aydin, I. M. Pryce, L. A. Sweatlock, E. M. Boyd, S. Walavalkar, J. Ma, and H. A. Atwater, "Frequency tunable near-infrared metamaterials based on VO₂ phase transition," *Opt. Express* **17**(20), 18330–18339 (2009), <http://www.opticsinfobase.org/oe/abstract.cfm?URI=oe-17-20-18330>.
12. F. J. Morin, "Oxides which show a metal-to-insulator transition at the neel temperature," *Phys. Rev. Lett.* **3**(1), 34–36 (1959).
13. P. Laffez, M. Zaghrioui, L. Reversat, and P. Ruello, "Electron doped (Sm_{1-x}Ca_x)MnO₃ perovskite manganite as potential infrared thermochromic switch," *Appl. Phys. Lett.* **89**(8), 081909 (2006).
14. J. B. Goodenough, "The two components of crystallographic transition in VO₂," *J. Solid State Chem.* **3**(4), 490–500 (1971).
15. A. Cavalleri, T. Dekorsy, H. H. W. Chong, J. C. Kieffer, and R. W. Schoenlein, "Evidence for a structurally driven insulator-to-metal transition in VO₂: A view from the ultrafast timescale," *Phys. Rev. B* **70**(16), 161102 (2004).
16. M. H. Lee, and M. G. Kim, "RTA and stoichiometry effect on the thermochromism of VO₂ thin films," *Thin Solid Films* **286**(1-2), 219–222 (1996).

17. F. Guinneton, L. Sauques, J. C. Valmalette, F. Cros, and J. R. Gavarri, "Optimized infrared switching properties in thermochromic vanadium dioxide thin films: role of deposition process and microstructure," *Thin Solid Films* **446**(2), 287–295 (2004).
18. M. Soltani, M. Chaker, E. Haddad, R. V. Kruzelecky, and D. Nikanpour, "Optical switching of vanadium dioxide thin films deposited by reactive pulsed laser deposition," *J. Vac. Sci. Technol. A* **22**(3), 859 (2004).
19. F. C. Case, "Influence of ion beam parameters on the electrical and optical properties of ion-assisted reactively evaporated vanadium dioxide thin films," *J. Vac. Sci. Technol. A* **5**(4), 1762 (1987).
20. D. J. Shelton, J. Sharp, G. Zummo, W. Folks, and G. D. Boreman, "Fabrication of periodic microstructures on flexible polyimide membranes," *J. Vac. Sci. Technol. B* **25**(6), 1827 (2007).
21. B. A. Munk, *Frequency Selective Surfaces: Theory and Design* (Wiley, 2000), Chap. 9.
22. E. E. Chain, "Optical properties of vanadium dioxide and vanadium pentoxide thin films," *Appl. Opt.* **30**(19), 2782 (1991).
23. F. Guinneton, L. Sauques, J. C. Valmalette, F. Cros, and J. R. Gavarri, "Role of surface defects and microstructure in infrared optical properties of thermochromic VO₂ materials," *J. Phys. Chem. Solids* **66**(1), 63–73 (2005).
24. A. Cavalleri, C. Tóth, C. W. Siders, J. A. Squier, F. Ráksi, P. Forget, and J. C. Kieffer, "Femtosecond structural dynamics in VO₂ during an ultrafast solid-solid phase transition," *Phys. Rev. Lett.* **87**(23), 237401 (2001).

1. Introduction

In the infrared (IR) band metamaterials have been fabricated for IR systems to control spectral reflection [1], transmission [2], emission [3-4], reflected phase [5], as broad-band wave plates [6], and for molecular detection [7]. Metamaterials are also known to produce unique capabilities for the manipulation of radiation at an artificially engineered surface, such as left handed refraction [8] or focusing of emitted, reflected, or transmitted radiation in the thermal infrared band. In a static metamaterial these capabilities are limited to a small frequency bandwidth and a limited spatial configuration. A tunable metamaterial would be able to scan a specific resonance across the thermal infrared to effectively broaden the useful frequency bandwidth of the device and allow the metamaterial to function at a wider range of angles of incidence or to steer a beam. Tunable reflectarray metamaterials have been used for beam-steering in the radio frequency band [9]. The development of beam steering capabilities without moving parts can be used to improve efficiency and lower costs in solar and infrared energy applications.

Tunable metamaterials have been demonstrated in the THz band [10] and in the near IR [11]. In this article the ability to control reflected phase in a tunable metamaterial is demonstrated for the first time. The results also mark the first tunable metamaterial demonstrated in the thermal infrared band, and the largest resonance shift measured to date in the infrared. This was accomplished using a reflectarray metamaterial configuration [5] consisting of thermochromic VO₂ square-patch elements on an amorphous silicon (a-Si) microcavity as shown in Fig. 1. The reflection spectrum was measured by FTIR at 20 and 70 °C, and the reflected phase was measured at 10.6 μm by a Twyman-Green interferometer as a function of temperature. The reflectarray consisted of an array of square-patches 1.7 μm wide spaced at a 2.1 μm periodicity populating a 15 mm by 4 mm array.

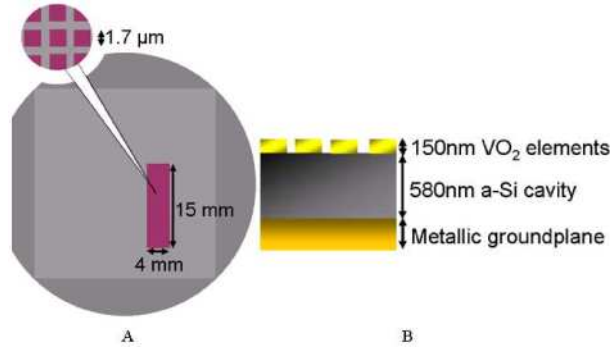


Fig. 1. (Color online) Reflectarray metmaterial diagram, A: patterned stripe with 1.7 μm square-patch VO_2 elements, B: reflectarray cross section.

2. Thermochromic VO_2 phase transition

Vanadium dioxide (VO_2) was first observed in Ref. 12 to undergo a transition from a semiconducting to metallic phase at 67°C . Such thermochromic behavior has been observed in other transition metal oxides such as MnO_3 [13], but VO_2 has produced the best results to date and may be integrated into metamaterials with a relatively low transition temperature. The metal-to-insulator transition in VO_2 is the result of a diffusionless phase transformation from an insulating monoclinic phase to a metallic rutile phase [14]. This phase transformation occurs on a picosecond time scale [15], and thus the transition rate of a thermochromic-tunable metamaterial is effectively limited only by the rate at which the elements may be heated and cooled.

VO_2 thin films may be fabricated by reactive electron-beam evaporation [16], reactive sputtering [17], pulsed laser deposition [18], or thermal oxidation of metallic vanadium [19]. The VO_2 elements used in this article were made using a reactive-ion version of the thermal oxidation process in Ref. 19 to convert metallic V elements to VO_2 . V square-patch elements were fabricated using standard electron-beam lithography methods as in Refs. 5 and 20. Figure 2 shows the measured electrical resistivity as a function of temperature and optical constants at 20 and 70°C for the VO_2 used in the square-patch elements. As the resistivity decreases exponentially from 45 through 65°C there is a change in the optical properties of VO_2 resulting in the contrast between 20 and 70°C . In the thermal IR the index of refraction increases from around 2 to 5, but this is accompanied by an increase in the extinction coefficient. The rutile phase of VO_2 does not

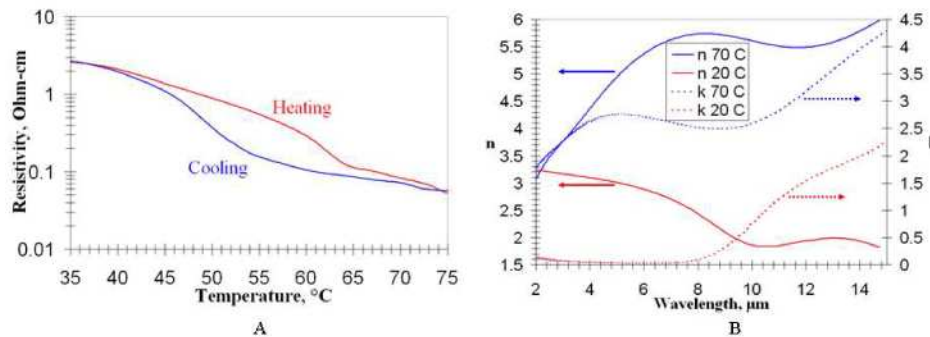


Fig. 2. (Color online) Measured material parameters for VO_2 , A: Resistivity as a function of temperature, B: IR optical properties (index of refraction n , extinction coefficient k) for VO_2 measured by ellipsometry.

have sufficient conductivity to serve as metamaterial elements in traditional split-ring resonator (SRR) designs, and thus the use of VO₂ in such designs will only result in a small shift in the resonance frequency. This can be seen in Ref. 11. The transition from a transparent to absorbing state that occurs in VO₂ may be best exploited in a metamaterial by coupling VO₂ elements to a resonating microcavity which is similar to an absorber frequency selective surface [21].

3. FTIR results

The reflectarray metamaterial from Fig. 1 was fabricated by depositing 100 nm of Cr on a Si wafer followed by the deposition of 580 nm of a-Si by electron-beam evaporation. V elements were fabricated on the a-Si cavity using electron-beam lithography. Following liftoff processing the reflectarray was annealed to convert the V elements to VO₂. Results from the FTIR measurement compared to finite-element method (FEM) simulations are shown in Fig. 3. The simulations were done using Ansoft HFSS software which included the measured optical constants of VO₂ at 20 and 70 °C. The reflected phase spectrum calculated by HFSS is shown in Fig. 3(b). The reflection minima and maxima seen in Fig. 3(a) are the result of the interaction between the a-Si microcavity and the electromagnetic resonance of the VO₂ patch elements. When VO₂ is in its semiconducting phase at temperatures below 45 °C these patch elements have less impact on the resonant frequency, and the resonance is largely due to the microcavity. After the structural transition, and VO₂ is in its metallic phase, there is a stronger interaction between the patch elements and the microcavity resulting in a red shift in the resonance frequency. Structures with electromagnetically resonant elements on transparent microcavities are further discussed in Ref. 21.

The resonating reflection minima shift from 9.2 μm to 11.2 μm covering a significant portion of the atmospheric transmission window in the thermal IR band. This is the first demonstration of a tunable metamaterial in the thermal IR and covers a larger portion of the spectrum than previous VO₂ metamaterial results in the near IR [11]. Although the bandwidth of the resonance increases at 70°C due to the increased extinction coefficient, the reflectarray remains in a resonating state in both VO₂ phases. This may be seen by the reflection maxima that occur at 5 μm for both VO₂ phases. There is also a bandwidth increase after the structural transition at 70 C. Previous VO₂ metamaterial results in the THz band [10] which used a continuous layer of VO₂ had a transition from a resonating to a spectrally flat state in which the device does not appear to be in a resonating mode.

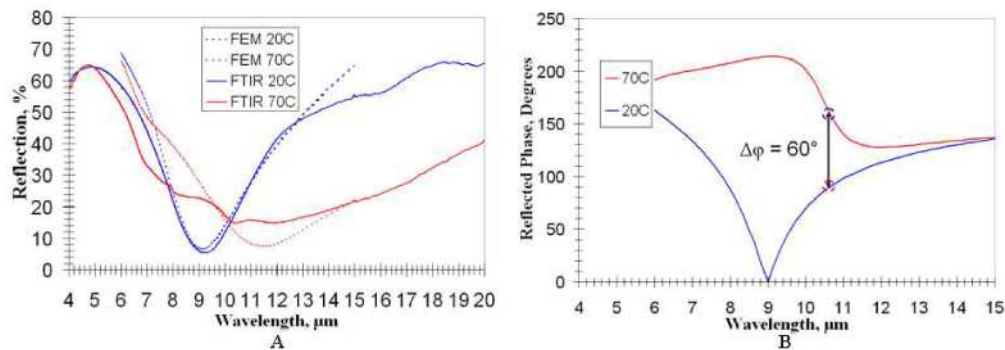


Fig. 3. (Color online) Reflected power and phase spectrum, A: Measured by FTIR compared to FEM simulation, B: Reflected phase spectrum simulated by FEM.

Based on the HFSS simulations, the largest change in reflected phase occurs at the 20°C resonance at 9 μm where the reflected phase changes by more than 180 degrees. At 10.6 μm the reflected phase changes by 60 degrees in the HFSS simulation. This will be confirmed by the interferometer measurement.

4. Experimental demonstration of tunable phase

A schematic of the Twyman-green interferometer used to measure the reflected phase is shown in Fig. 4.

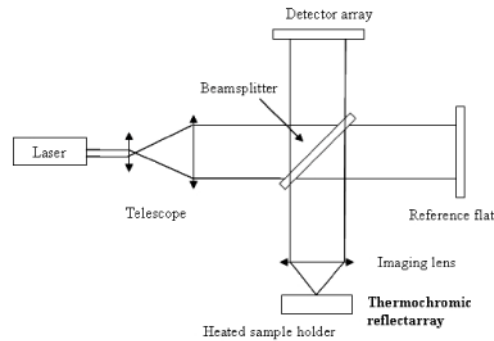


Fig. 4. Twyman-Green interferometer using 10.6 μm CO_2 laser used to measure reflected phase.

Interferograms are produced using a 10.6 μm CO_2 laser. The signal beam of the interferometer is incident on the thermo-chromic reflectarray sample that is oriented as shown in Fig. 1. A hot plate is used as the sample holder with a thermocouple in contact with the wafer to measure the temperature. The detector array is a Spiricon camera used to take interferogram images. Figure 5 shows interferograms measured at 20°C and 70°C. The step between successive fringes is equivalent to a 180 degree relative phase difference. White lines in Fig. 5 highlight one of the interference fringes across the wafer. The step in the fringes indicated by the white line occurs when a fringe falls across the patterned region. The reflected phase of the reflectarray is measured by the size in fringes indicated by the white line.

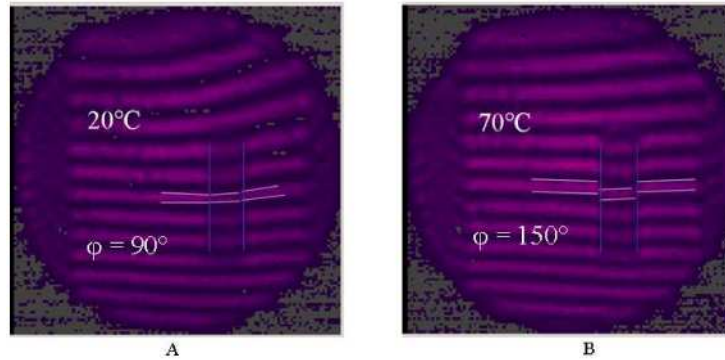


Fig. 5. (Color online) Interferograms of thermo-chromic reflectarray at A: 20°C and B: 70°C. White lines added to emphasize fringe contrast. The patterned reflectarray region is highlighted by a blue line.

By comparing the step in the white line to the distance between successive fringes (180 degrees) the reflected phase may be calculated. At 20°C, when the VO_2 elements are in the monoclinic phase, the reflected phase of the reflectarray is nearly equal to the a-Si cavity and thus there is only a small step in the fringes across the patterned region. At 70°C, after the transition to the rutile phase, the reflected phase has shifted 60 degrees as evidenced by the step in the fringes across the elements. This is in agreement with the FEM prediction, and thus the ellipsometry measurement of the optical constants, the FTIR measurement of the

spectral reflectance, and the interferometric measurement of the reflected phase are all in agreement. Figure 6 shows the measured reflected phase as a function of temperature.

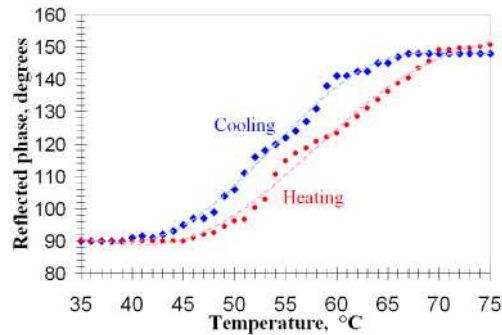


Fig. 6. (Color online) Measured reflected phase as a function of temperature during heating and cooling. Data points from interferogram analysis with polynomial fit indicated by the broken line.

The data in Fig. 6 was taken by analyzing the interferograms measured versus temperature while the sample was heated from 35 through 75 °C and then cooled through the same temperature range. From 65 through 45 °C, there is a linear decrease in the reflected phase corresponding to the exponential change in the resistivity of the VO₂ elements which is seen in Fig. 2(a). Following the hysteresis in the electrical resistivity upon heating and cooling, there is hysteresis in the reflected phase.

5. Discussion

The change in both reflected phase and resistivity in the VO₂ elements occurs between 45 and 65 °C for the films used in this article prepared by thermal oxidation. In pure single phase VO₂ deposited by pulsed laser deposition as in Ref. 18 the transition is more abrupt occurring between 65 and 75 °C. The hysteresis is also more significant in pure single phase VO₂ resulting in thermochromic transitions occurring over different temperature ranges for heating and cooling [18]. Lower transition temperature and wider thermal range with less hysteresis is seen in VO₂ doped with about 1 at.% W [18], or can be caused by characteristics of the microstructure such as small grains [22] or surface roughness and voids [23]. The optimal microstructure of VO₂ for thermochromic metamaterials will be the subject of future work, but the current VO₂ elements result in a linear change in reflected phase that may be easily controlled over a 20°C range. This is useful for beam-steering applications as the phase may be scanned continuously instead of being limited to binary states. The reflected phase may also be changed quickly by resistive heating in a thin film beneath the groundplane as in Ref. 10 or by optical heating [24].

6. Conclusion

Using thermochromic VO₂ square-patch elements, the reflection minima was shown to be scanned spectrally from 9.2 to 11.2 μm by FTIR measurements. This was in agreement with finite-element HFSS simulations that used the optical constants of VO₂ measured by IR ellipsometry. The same HFSS simulation calculated a 60 degree phase change at 10.6 μm, and this was confirmed by interferometer measurements. Interferometric measurements also showed a linear change in phase that occurred as the sample temperature changed from 45 through 65 °C.

Acknowledgements

This research was supported by grants from Northrop Grumman Space Technologies, and the Florida High Tech Corridor Council.