



HAL
open science

VO2 nanophotonics

Sébastien Cueff, Jimmy John, Zhen Zhang, Jorge Parra, Jianing Sun, Régis Orobtcchouk, Shriram Ramanathan, Pablo Sanchis

► **To cite this version:**

Sébastien Cueff, Jimmy John, Zhen Zhang, Jorge Parra, Jianing Sun, et al.. VO2 nanophotonics. APL Photonics, 2020, 5 (11), pp.110901. 10.1063/5.0028093 . hal-03006753

HAL Id: hal-03006753

<https://hal.science/hal-03006753>

Submitted on 17 Nov 2020

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

VO₂ Nanophotonics

Sébastien Cueff,¹ Jimmy John,¹ Zhen Zhang,² Jorge Parra,³ Jianing Sun,⁴ Régis Orobtcouk,¹ Shriram Ramanathan,² and Pablo Sanchis³

¹*Institut des Nanotechnologies de Lyon, Ecole Centrale de Lyon, 69134 Ecully, France^{a)}*

²*School of Materials Engineering, Purdue University, West Lafayette, Indiana 47907, USA*

³*Nanophotonics Technology Center, Camino de Vera s/n, Universitat Politècnica de València, 46022 Spain*

⁴*J. A. Woollam Co. Inc., Lincoln, NE, 68526, USA*

The intriguing physics of vanadium dioxide (VO₂) make it not only a fascinating object of study for fundamental research on solid-state physics but also an attractive means to actively modify the properties of integrated devices. In particular, the exceptionally large complex refractive index variation produced by the insulator-to-metal transition of this material opens up interesting opportunities to dynamically tune optical systems. This perspective paper reviews some of the exciting works on VO₂ for nanophotonics of the last decade and suggests promising directions to explore for this burgeoning field.

I. INTRODUCTION

Nanophotonics has enabled tremendous advances in the understanding of light-matter interaction and opened up new ways to control light at the nanoscale¹⁻³. Through the arrangement of designed nanostructured materials, a myriad of possibilities has recently emerged for applications in datacom, quantum optics, displays, bio-sensing or wavefront shaping. In particular, recent progress in dielectric metasurfaces made possible the design and fabrication of flat optics devices that hold promise to replace conventional bulk optics^{4,5}. At optical frequencies ranging from the UV to the mid-infrared regions, nanophotonic systems are patterned at the micro-nano-scale, resulting in building blocks whose geometries and arrangement are definitively set after fabrication. Such low-dimensional devices are static, what makes the dynamic variation of their physical properties not exempt of challenges. Most (if not all) nanophotonic applications would strongly benefit from tunable and reconfigurable properties and finding solutions to overcome such challenges is currently an intense field of research in which many different strategies are explored. A non-exhaustive list would include electro-mechanical systems, liquid crystals, thermal modulations, non-linear optics, and piezoelectric effects. More recently, phase-change materials (PCMs) have become a popular method of optical tunability without any moving parts. These materials are indeed very promising to enable dynamic modification to the physical properties of devices at the micro- and nano-scale. PCMs are a class of materials with unique physical properties: their structural arrangement can be controllably modified back and forth on a fast time-scale using a thermal, electrical or optical excitation^{6,7}. For some of these materials, the crystallographic re-arrangement translates into a large refractive index modification ($\Delta n \geq 1$). Such a large and fast refractive index modulation is a long-sought effect for photonics: an enabling technology to control and tune in real-time the optical properties of devices at the nanoscale.

Among them, vanadium dioxide (VO₂) is a prototypical example of functional materials showing large modifications in their physical properties upon specific external excitation. The complex physics of VO₂ – that we will briefly describe later in this review – ignited discussions among researchers on whether this material should belong to the class of PCMs. Regardless of this debate, we feel the term PCM is particularly well-suited to group together materials whose optical properties can be dynamically modified via a change in their atomic structure, a definition that works for both the tunable chalcogenide materials (e.g. GST, GeTe) and oxides (VO₂).

The insulator-to-metal transition (IMT) partly governing the drastic physical changes in VO₂ was first discovered by Morin in 1959 during his investigations on the temperature dependence of electrical conductivity in several transition-metal oxides⁸. A few years later, another team of researchers from the same laboratory (Bell labs) published a complete experimental data set of the optical properties of VO₂ above and below the transition temperature, which were found largely tunable both for bulk crystals and thin films^{9,10}.

Decades later, in the late 2000s, after years of intense fundamental research on this fascinating material, VO₂ started to be integrated in photonic devices, driven by the need to dynamically modify the response of optical metamaterials¹¹⁻¹³. Since then, VO₂ thin films have been exploited in a plethora of devices, concepts and systems in the field of nanophotonics. In this perspective review, our motivation is therefore to summarize this decade of research exploiting the IMT of VO₂ to dynamically modulate nanophotonic devices and systems.

A few reviews on the physics and properties of VO₂ can already be found in the literature¹⁴⁻¹⁶. They are, in most cases, either dedicated to understanding the complex physics of this material or to review their multiple possible applications. The objective of the present review is not to add another layer of extensive review of all researches conducted on VO₂. Our goal here is to give an in-depth and specific look at the recent results obtained on VO₂ applied to nanophotonics in wavelength ranges from the UV to the mid-infrared. We will see that this topic alone has already garnered so much interest that a review paper appears now necessary. From there, we want the reader to get an overall panorama of current researches on

^{a)}Electronic mail: sebastien.cueff@ec-lyon.fr

the field and on future promising perspectives.

We first briefly describe the intriguing physics of VO₂ and explain the origin of the transition observed in this material. We then describe in more detail the optical properties of VO₂ and give recommendations to study and analyze them. The fourth part is dedicated to review recent works on VO₂ for integrated guided wave optics and the fifth is focused on VO₂ based metasurfaces for free-space photonics. In a sixth and last part, we describe a few interesting directions to explore that we find particularly promising for future works on the field, namely the electrical control, the absorption mitigation, the spatial addressability combined with memory effect and the ultrafast switching of VO₂.

II. THE PHYSICS OF VO₂

VO₂ is a particularly complex material with extremely rich physics that have been actively investigated in the past decades. It is a prototypical example of correlated oxides and Mott insulators. Describing the physics of Mott insulators is outside the scope of this review and we refer the readers to several excellent reviews on the topic^{17,18}. But the basic physics behind this phenomenon can be understood as follows. Band theory has proven to be a very powerful tool to predict and understand the physical properties of materials with known elements arranged in a specific crystallographic structure. This has helped classify materials in “boxes” according to their physical properties, linked to their band diagram. For example, insulators have their Fermi level in a band gap while metals have their Fermi level inside a partially-filled band. The distribution of energy and carriers, as calculated through the energy band diagrams therefore dictates the physics of the materials. However, for some materials this band theory fails, at least partially. Indeed, the physical properties of some transition-metal oxides do not follow band theory. VO₂ is one of the most well-known example of such materials: it has a partially filled d-electron band and, according to band theory, should therefore behave as a metal but surprisingly shows insulating properties at room-temperature. This unexpected effect is due to the strong electron-electron coulombic repulsion that exists in this material and that are not taken into account in band theory. Such a correlation between electrons “freeze” them in their sites and prevent electrical conduction: this is the basic principle of Mott insulators and strongly correlated materials such as VO₂. The IMT is a natural consequence of this strong electron-electron correlation: an appropriate stimulus will break this equilibrium (e.g. heat, above 70°C) and electrons will start to behave as free carriers, similarly as in a regular metal.

Simultaneously to this IMT behavior, VO₂ undergoes a structural change from a monoclinic phase M1 with space group of $P2_1/c$ at room temperature to a rutile phase with space group of $P4_2/mnm$ at temperatures above 70°C⁸. Under strain¹⁹ or doping with e.g. Al or Cr^{20–22}, another type of monoclinic phase M2 with space group of $C2/m$ and triclinic phase T with space group of $P1$ were stabilized. These monoclinic phases M1 and M2 and triclinic phases possess

different patterns in V-V bond length and volume change with respect to the rutile phase. A thermodynamic phase diagram has been established experimentally by Park et al. which illustrates the stability of these monoclinic phases in various strain-temperature windows¹⁹.

This crystallographic transition does not simplify the analysis and understanding of the overall physical properties. Peierls suggested that the structural change induces a lattice deformation, which modifies the periodic ionic potential in the material, resulting in a band structure change²³. Consequently, we have at least two possible explanations as to what could be the driving force and the physical mechanism behind the transition of VO₂. On one hand, we have the evidence of strong electron-electron correlation, suggesting a Mott-Hubbard scenario with strong Coulomb interaction between electrons, triggering the insulator-metal transition²⁴. While on the other hand, a structural transformation with strong dimerization, as predicted by Peierls, is also possible, wherein modification of band structure is caused by the lattice distortion. Critical theoretical works in the last couple of decades by Eyert^{25,26}, Biermann et al.²⁷, He and Millis²⁸, van Veenendaal²⁹, among others have described the electronic structure as well as the magnetic ground state of vanadium dioxides¹⁴. Using first principles studies, Eyert reproduced the basic features of rutile and monoclinic phases with density functional theory and local-density approximations²⁵. With hybrid functionals corrections, better agreement with experiments such as bandgaps and antiferromagnetic ordering has been obtained²⁶. Based on cluster dynamical mean field theory within density functionals, Biermann et al.²⁷ suggested that correlation assisted dynamical V-V singlet pairs plays a critical role in the transition of VO₂. Recent theoretical studies on the transition and band structure suggested an intermediate theory combining both Peierls and Mott, described in terms of Mott-assisted Peierls transition³⁰. This theory seems also consistent with the recent reports of a photoinduced transient ‘metal-like’ state of VO₂ that is produced without modifying the monoclinic phase, hence indicating a purely electronic transition.^{31,32} The ultrafast THz phase diagram reported by Cocker et al.³³ shows that with increasing the incident laser fluence, rutile phase can be nucleated and stabilized. Below 180 K, a transient metallic monoclinic state emerges prior to the nucleation of rutile phase. This photoinduced ultrafast dynamics of VO₂ is consistent with theoretical models proposed by He et al.²⁸ and van Veenendaal²⁹ in critical fluency, coherent structural motion, and metastable M1 metal phases. The difficulty in experimentally untangling the structural and electronic aspects of this transition^{34,35} due to the transient nature poses further challenges to this field. Further technique developments to probe local dynamics of phase evolution under external fields will be important as this class of materials becomes more widely studied and is implemented in device technologies.

Regardless of the intricate physical mechanisms governing the change of phase in VO₂, one of its main advantages is its multifunctional and multistimuli character. Indeed, the IMT of VO₂ can be induced using thermal heating (beyond $T_c \sim 70^\circ\text{C}$ at standard pressure), applied electric fields ($E =$

10^5 V.cm^{-1})^{36,37}, injected carrier densities ($n_e=10^{18} \text{ cm}^{-3}$)³⁸, optical^{39,40} and terahertz pulses⁴¹. Conversely, this material and its IMT can then be used as an electrical switch, a thermal modulator, a thermochromic window, an optical attenuator, to name just a few. For more details on all the possible applications of VO₂, we refer the readers to recent reviews^{14,16}.

III. OPTICAL PROPERTIES OF VO₂ AND RECOMMENDED METHODS TO STUDY THEM

The optical properties of VO₂ thin films can be affected by a variety of factors, including deposition conditions, post treatments, lattice matching to substrates, microstructure and impurity concentrations. Understanding the optical behaviors of each specific sample at insulator and metallic phases and their optical evolution through the IMT process is therefore not only crucial for further utilization but will empower the optimization of VO₂ performance as a tunable medium in various optical and optoelectronic devices. Describing the various growth methods and related fabrication strategies is outside the scope of the present review and we redirect interested readers to a recent review focused on that aspect.¹⁵

In this section, we describe the optical properties of VO₂, their relation to the thin film's structure and pinpoint some of the difficulties in analyzing these complex and tunable optical properties.

A. How to break correlations between thickness and complex refractive index

Spectroscopic ellipsometry has been the standard method to investigate the optical properties of thin VO₂ films. Their refractive indices across the characteristic IMT evolution have been reported from ultra-violet to infrared frequency ranges⁴²⁻⁴⁵. The most pronounced changes occur at infrared wavelengths (Figure 1), where the refractive index value strongly decreases (Fig. 1 (a)), and there is simultaneously a large increase in the extinction coefficient (Fig. 1 (b)). Around the transition temperature and above, Drude losses associated with the metallic delocalized electrons take the place of Lorentzian absorptions in the imaginary dielectric function ϵ_2 , and the permittivity ϵ_1 gradually goes to negative, indicating the transition from insulator to metallic phases (see Fig. 1 (c)). VO₂ therefore presents broad absorptions even in its insulator phase. Such a lack of transparency is known to pose great challenges to optically determine film thickness due to the commonly observed correlations between thickness and refractive index for absorbing materials. The simplest method to prevent these correlations is to measure film's thickness via alternative methods such as profilometry, AFM or X-Ray diffraction, hence avoiding the need to let the thickness be a free fit parameter. However, these methods are not necessarily relevant for very thin films having a non-negligible surface roughness. Another common strategy to break such correlations is to simultaneously analyze ellipsometric data acquired from films with different thicknesses. Assuming the set of

films have identical dielectric functions⁴⁶, thickness was regarded as the sole cause in varying ellipsometric data, thus enabling the independent determination of thickness and refractive index. The main concern associated with this multi-sample approach is the assumption validity when applied to thin VO₂ films. Many findings contradicted this assumption, reporting optical properties that depend on thickness^{44,47}, due to several factors such as surface roughness, vertical gradients in microstructures and in stoichiometry.

Another approach that we find particularly suited to break the correlations is through analyzing IMT dynamic data using a tunable optical model while maintaining a constant thickness to fit all states of VO₂⁴³. This method focuses on the dominant effect on ellipsometric data originated from the evolution of dielectric functions during the phase transition process. By constraining the absorption resonance energies and linking oscillator parameters during the IMT, the correct thickness is expected to fit the large amount of spectroscopic data at various temperatures and different phases. The thickness can be confirmed through uniqueness test – i.e. only one thickness value can fit all states of VO₂ – and better sensitivity in the metallic phase was reported. Note that this method is only possible because the IMT of VO₂ does not modify the thin film thickness.

Other new methods could be found to break such correlations but we emphasize that these correlations between thicknesses and complex refractive index are very important, as one can easily (and unfortunately) overlook them during the analysis and thus obtain incorrect dispersion data sets.

B. Oscillator models and optical dispersion throughout the IMT

The imaginary dielectric functions, ϵ_2 , of the insulating films deposited on silicon or silicon oxide substrates typically consist of two broad absorptions in visible and near infrared spectrum, with center energy varying around 3.5 eV and 1 eV, respectively. As film grows thicker (> 100 nm), a peak at ~ 2.1 eV becomes more distinguishable. For films on sapphire substrate, the presence of this absorption peak has been unearthed at much lower thickness (~ 30 nm), which could be related to the improved film quality on sapphire even at the early stage of deposition. Each of these absorption peaks find their physical roots in inter-band transitions between different orbitals (for more details, see e.g.¹⁴). Lorentz oscillators have been commonly applied to describe these absorptions^{10,43,49}. The energies, amplitude and broadening of these oscillators are adjusted across the IMT to accommodate optical changes induced by the phase transition. The Lorentz peak at ~ 1 eV red-shifts noticeably as temperature rises. It is eventually replaced by a Drude oscillator during the IMT to better represent the metallic absorptions. By monitoring these oscillator parameters through the thermal ramping process, Kakiuchida et al.⁴² provided insights on band structural transition and band gap changes through the IMT process.

During the IMT of VO₂, interesting regimes of intermediate phases occur, in which the medium becomes a combination of

metallic and insulator regions, as shown via scanning near-field infrared microscopy (Fig. 1 (e))⁴⁸. Effective medium theory (EMT) has been applied to derive the optical properties of thin VO₂ films at these intermediate states when dynamic monitoring is not feasible. The method assumes the coexistence of insulating and metallic domains and approximates the refractive index based on the domain volume fractions at intermediate temperatures during the IMT process. EMT method is potentially valuable in describing gradual transitions, such as VO₂ films on silicon substrate⁴⁴. However, sharp optical changes of thin films with better quality or long-range order will greatly challenge the effectiveness of EMT models. The demand of drastic changes in oscillator parameters near the percolation transition could lead to failure in regression analysis or non-physical outcomes.

C. Influence of the substrate and microstructure on the optical properties and hysteresis.

Film microstructure has been shown to strongly impact the optical properties of VO₂ films. Important information on the dynamic structural transformation of VO₂ can be obtained via various techniques such as X-ray diffraction, transmission electron microscopy, pump-probe electron diffraction and Raman spectroscopy^{31,50–53}. Using in situ real time spectroscopic ellipsometry (RTSE), Motyka et al.⁴⁷ reported changing optical properties and structural variation during film growth and post deposition treatments. A two-layer model revealed a more disordered/amorphous film with Lorentzian characteristics at early stage of the deposition or at film surface. As film grows thicker, the bulk materials become better ordered with larger grain sizes and stronger metallic conduction, leading to the potential shift of bandgap to lower energy.

This structural and compositional dependence can also be found in the transition profile, that can vary from abrupt to gradual and in the phase transition temperature that can be shifted depending on film quality, grain size, stoichiometry, degree of strain, and impurity concentrations^{43,45}. Figure 1 (f) shows the phase transition hysteresis loop and the transition temperatures optically determined from the reversible IMT thermal process on two different VO₂ samples. Thin films deposited on sapphire substrate present a sharp and narrow transition in comparison to the large hysteresis measured on films prepared on silicon⁴³. These findings are consistent with resistivity measurement^{44,47}, both resulting from better film quality on c-plane sapphire likely due to smaller lattice mismatch at the film/substrate interface.

Wan et al.⁴⁴ studied the optical properties of VO₂ at both insulator and metallic phases from the visible to the far-infrared regions, as shown in Figs. 1 (g)-(h). The complex refractive indices from 2 to 11 μm were reported to be less sensitive to deposition process and film thickness. In that region, Drude losses dominate the metallic phase while Lorentzian absorptions tapered down towards longer wavelengths at room temperature. Their measurements therefore indicate a spectral window (2-11 μm) in which there are no, or negligible, microstructure- or substrate-dependence in the optical proper-

ties of VO₂ (see Figs.1 (g)-(h)). On the other hand, they also noticed a couple of substrate-dependent features: in the metallic state, Lorentz absorption around $\sim 10 \mu\text{m}$ was found to be absent for films on sapphire, but present for films on silicon, likely due to other polymorphs of vanadium oxide. Moreover, strong vibrational resonances with distinctive substrate-dependence were observed between 17 to 25 μm at insulating states. Further investigation in the cause of these absorptions is necessary in tuning VO₂ films for infrared applications.

The crystalline structures of VO₂ theoretically present anisotropy. In most cases though, an isotropic optical model will sufficiently describe VO₂ thin films due to the following reasons. First, the films may consist of polycrystalline domains of different orientations, or different polymorphs of VO_x, resulting in isotropic behaviors on a macroscopic scale. This is typically what happens when VO₂ is deposited on Si or SiO₂^{15,54,55}. Secondly, the optical measurement is generally less sensitive to out-of-plane refractive index for a very thin absorbing film. Lastly, as we have seen before, in some cases correlations between thickness and refractive index for thin absorbing films add uncertainties to the accuracy and reliability of anisotropic analysis. More confidence in identifying anisotropy would be expected in thicker films with better crystallinity and stoichiometry control.

As we have seen in this part, the optical properties of VO₂ as well as the transition behavior, are affected by a number of factors mainly linked to the substrate, thickness and microstructure of the VO₂ thin film. It is therefore crucial to carefully analyze the optical properties of each VO₂ sample to adapt future devices design to its specificity.

IV. VO₂ FOR GUIDED INTEGRATED PHOTONICS

Photonic integrated circuits (PICs) allow to exploit the benefits of light for communication and data processing at the micro and nano scale. To this end, silicon has become the mainstream technology for novel developments in a wide variety of applications (datacom, telecom, sensing, high-performance computing ...)^{56–58}. However, silicon is not a particularly well-suited material for enabling active functionalities due to its intrinsic properties. In particular, the control of the optical phase and/or optical amplitude based on the plasma dispersion effect usually yields to tradeoffs among speed of operation, energy consumption, insertion losses or footprint⁵⁶. Silicon active devices with switching speeds in the picosecond time scale are possible. However, active lengths in the millimeter range are typically required for enabling large extinction ratios. More compact devices can be achieved by using resonant structures but at the expense of significantly narrowing the optical operation bandwidth. In this context, the hybrid integration of silicon with CMOS-compatible materials featuring unique properties has opened a path to achieve ultra-compact, broadband, and highly-efficient guided photonic devices⁵⁹. Such type of devices are desired to develop advanced PICs with complex functionalities and large-scale integration. As it has been mentioned, VO₂ stands out for the ultra-large change in its complex refractive index between

the insulating and metallic states. This feature enables hybrid tunable VO₂ waveguides with lengths down to hundreds of nanometers. Moreover, this kind of waveguides are broadband since the changes in the VO₂ refractive index span a spectral range from the visible to the mid-infrared wavelength regions. The scheme of a commonly reported hybrid VO₂/Si waveguide is depicted on Fig. 2. This latter comprises a silicon waveguide with a patch of VO₂ atop. By triggering the IMT of VO₂ with an external excitation (heat, electric field, or light), the guided mode could experience a change in both optical phase and amplitude.

In the following, we describe recently reported results, organized in three different parts, according on the methods used to trigger the IMT of VO₂.

A. Thermally-modulated VO₂/Si waveguides.

Several hybrid VO₂/Si waveguide devices, intended to function as amplitude modulators, have been proposed and demonstrated by thermally triggering the IMT^{54,60–65,70,71}. To this end, the temperature of the chip can be controlled using a Peltier device or local heat into the VO₂ patch can be applied using Joule heating with metallic microheaters. One of the first hybrid VO₂/Si devices (Fig. 3a) was demonstrated by Briggs et al.⁵⁴. They showed the capabilities of VO₂ for optical switching at telecom wavelengths with an active length of only 2 μm but with a moderate extinction ratio of 6.5 dB and insertion losses of around 2 dB. For a given polarization of the guided optical mode, there is always a trade-off between the extinction ratio and the insertion losses and most groups try to enhance the former and reduce the latter by engineering the hybrid VO₂/Si waveguide. Extinction ratios of 16 dB and insertion losses of 3.8 dB have been demonstrated with an optimized 3-μm-long hybrid VO₂/Si waveguide (Fig. 3b)⁶⁰. Ultra-short hybrid VO₂/Si waveguides with a length of only 500 nm have also been demonstrated by embedding the VO₂ within the waveguide instead of placing it on top (Fig. 3c)⁶¹. In this configuration, extinction ratios with almost 10 dB but relatively high insertion losses of around 6.5 dB have been reported. The optical switching performance can be further improved by engineering the morphology of the VO₂ layer (Fig. 3d). In such a way, insertion losses below 1 dB and extinction ratios above 20 dB with switching times in the microsecond range were demonstrated for a 20-μm-long hybrid VO₂/Si waveguide⁶². On the other hand, the integration of VO₂ on silicon waveguide could also open alternative applications such as tunable polarizers. Sanchez et al. demonstrated a 20-μm-long transverse-electric (TE) pass polarizer exploiting the polarization dependence loss of the hybrid waveguide (Fig. 3e)⁶³. They showed a rejection of 19 dB for the transverse-magnetic (TM) polarization in the active state together with switching times of few microseconds for a wavelength range between 1540 and 1570 nm. The integration of hybrid VO₂/Si waveguides in add-drop ring resonators could also enable 2x2 optical switches⁶⁵. Recent experimental results have shown an extinction ratio up to 25 dB with insertion loss of 1.4 dB by placing a

19-μm-long VO₂/Si waveguide in a ring resonator (Fig. 3f)⁶⁴.

B. Electrically-controlled VO₂/Si waveguides.

The control of the VO₂ phase transition in hybrid waveguide has also been demonstrated by applying an electric field between two separated metallic contacts, as seen in Fig. 3(g)-(h). Such a scheme is more interesting for future integrated applications and should enable faster switching times compared to the purely thermal heating counterpart, as the applied heat can be localized and confined only to the VO₂ patch on the chip. Markov et al. used this approach to investigate the electro-optical switching dynamics in an ultra-short VO₂/Si hybrid waveguide (Fig. 3g)⁶⁶. They showed switching times of less than 2 ns for the IMT. The relaxation time to fall back to the insulating state involved a thermal dissipation process making the recovery slower. Nonetheless, by limiting the current to reduce Joule heating, they predicted switching time of the relaxation down to 3 ns but at the expense of a lower extinction ratio. Joushaghani et al. demonstrated a similar electro-optic VO₂/Si switch (Fig. 3h)⁶⁷. They achieved a high extinction ratio of 12 dB in a 1-μm-long device with insertion losses of 5 dB. Moreover, by biasing VO₂ near the onset of the IMT, they tested the capabilities of the device as a photodetector and achieved a responsivity in excess of 10 A/W with optical powers lower than 1 μW. Several proposals have also been made for developing hybrid plasmonic modulators^{72–76}. However, the experimental demonstration of such devices is still lacking. On the other hand, the overall power consumption highly depends on the VO₂ patch size and the external resistance required to limit the maximum current flowing in the metallic state and avoid damaging the electrical contacts⁷⁷. Thus, the only reasonable solution to reduce the power consumption of electrically-controlled devices with respect to thermal heating approaches is to design very short hybrid waveguide, what could restrict the maximum achievable extinction ratio.

C. Optically switched VO₂/Si waveguides.

Finally, all-optical switching schemes to control the state of hybrid VO₂/Si waveguides could be the most promising approach. Notably, the switching timescale of the IMT, triggered by optical excitation, has been demonstrated down to the femtosecond and would allow ultra-fast speed with a seamless integration in PICs^{39,55,68,69,78–80}. However, such type of ultra-fast device has not been demonstrated yet. Ryckman et al. demonstrated the first all-optical hybrid VO₂/Si device⁶⁸. They integrated the hybrid VO₂/Si waveguide in a small ring resonator and induced the IMT of VO₂ by pumping the patch out-of-plane with a pulsed laser in the visible. No switching times were reported and the optical switching of the VO₂ was attributed to be photothermal. Afterwards, the same authors investigated the timescale of such devices (Fig. 3i)⁵⁵. In this case, they used a pulsed laser of a few nanoseconds to

excite the IMT of VO₂. On one hand, a fluence dependence in the completion of the IMT was observed. For this device, pump fluences above 12.7 pJ/μm² drove the VO₂ to its metallic state. On the other hand, the measured IMT timescale was found similar to the pump-pulse width (~25 ns) with a minimum influence of the fluence. However, the switching time from metallic to insulating (relaxation time) showed a high dependence with the fluence and the VO₂ patch size. By increasing the pump fluence above the threshold up to ~70 pJ/μm² and enlarging the VO₂ patch from 500 nm to 1 μm, the relaxation time increased from ~30 ns to ~3 μs. Thus, the relaxation dynamics were found to depend on the thermal diffusion of the monoclinic phase. Haglund et al. investigated the timescale of hybrid VO₂/Si waveguides (Fig. 3c) by illuminating out-of-plane the device with femtosecond laser acting as a pump and using an in-plane probe laser to record any change in the VO₂⁸⁰. For this case, a 900-nm-long VO₂ patch reportedly showed switching speeds lower than 2 ps for fluences between 50 and 100 pJ/μm². More recently, sub-ps switching times have also been demonstrated by optimizing the fluences and reducing the volume of VO₂ in the hybrid waveguide.⁸¹

While these are promising results, the out-of-plane excitation is not the best approach for integration in PICs and future all-optical schemes with both the pump and the probe guided within the waveguide would be more desirable. All-optical switching with hybrid waveguides using an in-plane approach has been recently demonstrated⁶⁹. In this case, SiN instead of Si was used for the hybrid waveguide, to handle the pump between 700 and 1000 nm and the probe at 1550 nm (Fig. 3j). An extinction ratio of 10 dB was achieved for a 5-μm-long hybrid waveguide with a switching energy as low as 6.4 pJ. However, the timescale was not reported. Parra et al. have recently addressed this question using a similar in-plane pump-probe technique in the telecom wavelength region with a hybrid VO₂/Si waveguide⁸². Their temporal results suggest a thermal dynamics in which the phase change of VO₂ is thermally triggered and therefore limited to the nano/microsecond range.

In view of this recent work, a remaining important challenge is to find ways to reach the femtosecond time-scale of the IMT in integrated devices. Promising directions could be to precisely adjust the excitation source to only trigger the electronic IMT of the VO₂ without introducing parasitic heat generation^{83–85}. In addition, engineering the thermal environment of devices to efficiently dissipate heat would also greatly help minimizing the relaxation time of the IMT.

V. VO₂ METASURFACES AND METAMATERIALS FOR FREE-SPACE NANOPHOTONICS

Recent years have seen the emergence of optical metasurfaces^{4,5}. In these devices, an abrupt phase/amplitude shift is printed on a surface through engineered nano-elements. By spatially arranging such meta-atoms on a substrate one can design metasurfaces tailored for specific optical functionalities such as lenses, polarizers, retroreflectors, holo-

grams, perfect absorbers, among many others.^{4,86–88}. Some of these flat optics devices already surpass the performances of conventional diffractive optics components. As this field is becoming mature with conventional 'passive' materials, researchers are now actively seeking means to dynamically modify the properties of these nano-elements to demonstrate actively reconfigurable metasurfaces, what may revolutionize the field of integrated optics. We review in the following, different approaches and concepts that exploit VO₂ to dynamically tune the free-space optical response of flat optics devices.

A. Tunable metasurfaces based on un-patterned thin films

A thin-film material with adjustable complex permittivity can be considered as the simplest form of a metasurface, especially if it presents spatial variations of permittivity. As described in section III, across the transition of VO₂, intermediate states are produced, in which coexist metal and insulator phases at the nanoscale. These different mixed states can be regarded as naturally disordered metamaterials with tunable optical properties, and in the following we analyze the different features of these appealing states.

Perfect absorption was achieved by Kats et al. in a system where a thin layer of VO₂ (~180 nm) was grown on sapphire substrate [Fig.4(a)]⁸⁹. At temperatures close to the IMT [Fig. 4(b), ~343 K], the absorption losses equal the radiative losses and the so-called critical coupling conditions are reached, producing an absorption of 99.75% at a wavelength of λ=11.6 μm. Given this near-perfect absorption is dynamically tunable, over the transition temperature range, the reflectivity at λ=11.6 μm can be largely modulated from 80% to 0.25%. Similarly, Butakov et al. reported broadband tunable reflection and transmission in a tri-layer system (Ge/VO₂/Al₂O₃) in the mid-infrared and demonstrated electrical tuning of such a system⁹⁰. Furthermore, Rensberg et al. reported that the suppression of reflection can be engineered by depositing an ultrathin layer of VO₂ on epsilon-near-zero substrates such as aluminum-doped zinc oxide (AZO), SiO₂, and ZnO [Figs. 4(e)-(g)] and tuned by temperature⁹¹. A minimum of reflectance is found close to the plasma resonance of AZO [Fig. 4(e)] and the reststrahlen band of SiO₂ while local minimum is absent on ZnO. Conversely, as stated by Kirchhoff's law, the absorption is closely related to the thermal emission, implying that a good absorber should also be a good thermal emitter. As Figs. 4(c) and 4(d) show, a 150-nm-thick VO₂ film deposited on sapphire exhibits "perfect" blackbody-like emissivity ~1 in the vicinity of the IMT (~74.5°C) over a wavelength range of 40 cm⁻¹⁹².

Utilizing the distinct optical feature of VO₂ between metallic and insulating phase, several optical and radiative thermal devices have been proposed based on un-patterned thin films. A limiting optical diode, in which the phase transition of VO₂ is triggered asymmetrically depending on the direction of incident light [Figs. 4(h) and (i)], was designed using a stack comprising a semi-transparent metallic layer, a VO₂

layer, and a transparent substrate⁹³. In such a device, a backward illumination triggers the IMT, hence resulting in a reduced transmission in that direction, while a forward illumination with the same intensity leaves the VO₂ state in its insulating phase leading to high transmission. A similar yet different concept imagined exploiting VO₂ to control heat fluxes: a radiative thermal transistor, capable of modulating and amplifying radiative heat transfer in the far-field has been proposed by Joulain et al. [Figs. 4(j) and (k)]⁹⁴. By placing VO₂ between two blackbodies having different temperatures, a radiative flux amplification factor α larger than 1 can be achieved in the transition region from 341 to 345 K driven by the emissivity variations of VO₂ at metallic and dielectric phases.

Engineering the geometric structure or spatial variation of phase through controlling growth condition and introducing defects are other interesting methods to fabricate metasurfaces without relying on etching processes. Using ion irradiation through masks [Figs. 4(l) and (m)], Rensberg et al. introduced defects into designated regions of VO₂ and locally changed the transition temperature^{95,98}. As a result, a metasurface composed of metallic and insulating phase of VO₂ forms upon heating ($T \sim 60^\circ\text{C}$) and vanishes at temperatures away from it (e.g. 30°C and 80°C), the reflectance of which then shows engineered switchability and polarization dependence across the transition region between 25 and 90°C . Another original method of creating "natural" metasurfaces can be obtained by controlling the structure and texture of VO₂ through specific strain governed by the substrate nature and orientation, as shown in Figs. 4(o) and (p). By using a-cut (11 $\bar{2}$ 0) sapphire substrate which supports anisotropy growth, Ligmajer et al. grew a layer of self-structured VO₂ nanobeams having widths in the range 100-200 nm and lengths of 1000-2000 nm⁹⁶. The measured extinction spectra exhibited a broadband strong polarization dependence in both the metallic and insulating phases. Such devices may be used for large-scale modulators with polarization control.

Given the large modifications in the local dielectric environment during the IMT, VO₂ can also be utilized as a tunable substrate to form heterostructure via direct contact with other photonic materials. Folland et al. reported a tunable hyperbolic metasurface device [Fig. 4(q)] by transferring a natural hyperbolic material medium – an isotopically enriched hexagonal boron nitride (*h*BN) – on top of a VO₂ crystal⁹⁷. In their study, they demonstrated that the insulating and metallic domains of VO₂ can reflect, transmit, and launch hyperbolic phonon polaritons (HPhPs) at domain boundaries and showed a reconfigurable control of in-plane HPhPs propagation. Modulation on the wavelength of HPhPs by factor of 1.6 was achieved across these domains.

B. Dynamic modulation of spontaneous light emission

Since the pioneering work of Purcell, we know that the physics of spontaneous light emission is both governed by

the quantum mechanical electronic transitions of the emitter and by the optical environment, also known as Local Density of Optical States (LDOS). As VO₂ presents very large modulations of its complex permittivity upon the IMT, it can be exploited to dynamically modify the LDOS and therefore open up interesting means to control the spontaneous emission of quantum emitters in integrated devices. This has been experimentally demonstrated in a multilayer stack comprising thin films of quantum emitters (Er³⁺:Y₂O₃) and VO₂ (see Fig.5(a))⁹⁹. The device, comprising a quarter-wavelength phase-change layer located between an emitter layer and a metal mirror, was specifically designed such that the VO₂ IMT can be externally switched optically while also having optimized influence on the LDOS of the emitter layer. Upon switching the VO₂ layer, there is a π phase shift in the effective optical path length, which maximizes the influence of the change of refractive index on the surrounding LDOS. Using this device, combined with the symmetry difference in the polarization of electric dipole (ED) and magnetic dipole (MD) transitions of erbium ions, one can dynamically switch between spectrally distinct ED-dominant and MD-dominant emission by simply changing the state of VO₂. With this concept, a broadband all-optical direct modulation of 1.5 μm emission from erbium ions was shown. Interestingly, such a dynamic optical modulation scheme was experimentally demonstrated to be more than three orders of magnitude faster than the excited state lifetime of the erbium emitters (see Fig.5(b)), hence enabling fast direct modulation even for long lifetime quantum emitters⁹⁹.

Other interesting ideas were further developed from that concept, as described in the following. Very recently, Jha et al. have used a similar configuration, in which VO₂ thin films are integrated in proximity to quantum emitters (*h*BN) as a means to modulate their LDOS. This tunable LDOS is then used to modulate the emission rate of quantum emitters, which in turns enabled resolving both the spatial position of the quantum emitter and its three-dimensional dipole orientation¹⁰³. Szilard et al. have calculated the enhancement of spontaneous rate of ED and MD emitters in the vicinity of VO₂ layers (see Fig.5(f) and (g)). They have shown that both ED and MD transition rates can be strongly enhanced, especially in the mid-infrared range at some specific stages of the IMT (in between insulating and metal). They further suggest that the IMT hysteresis could be used as another degree of freedom for dynamic control of the spontaneous emission with a memory effect. Other works studied the potential of VO₂ nanolayers associated with plasmonic antennas (see Fig.5(c-e)¹⁰⁰) or hyperbolic metamaterials (Fig.5(h-i)¹⁰²) for further controlling the enhancement of spontaneous emission of quantum emitters.

C. Actively reconfigurable plasmonic antennas

In the last decade, VO₂ has been largely used as a means to control the resonance of plasmonic scatterers. These devices are usually based on plasmonic resonances in metallic nanostructures, fabricated on top of a VO₂ thin film^{13,41,89,104,105}.

Some of the earlier works were based on split-ring resonators devices patterned above VO₂ thin films^{13,106}. By changing the phase of VO₂, the authors were able to controllably modify the resonant wavelength of the system in the MIR and visible range. Many different shapes and sizes of metallic nano-antennas were reported and experimentally demonstrated to be tuned by the underlying VO₂ thin film, spanning all wavelength regions from the visible to the terahertz range (see some selected examples in Fig. 6 (a-h)). For the vast majority of these works, the VO₂ layer produces an amplitude modulation of the free-space reflection or transmission through the devices. Later on, numerous studies made use of similar plasmonic nano-elements (disks, antennas, slot) arrays for alternative applications such as optically-triggered memory devices¹⁰⁷, tunable color generation^{108,109}, optical phase-array¹¹⁰, switchable polarization rotation¹¹¹, active directional switching of surface plasmon polaritons¹¹². As VO₂ transitions from insulator to metal, Butakov et al. exploited this IMT to demonstrate switchable dielectric-plasmonic resonators using directly patterned VO₂ scatterers¹¹³. Alternatively, Muskens et al. have exploited the spatially-confined hotspots of antennas to locally switch VO₂ thin films. This method helped both reducing the energy consumption and the recovery time of VO₂, hence enabling reversible switching at over two million cycles per second, i.e. much faster than for a VO₂ thin film alone¹¹⁴.

VI. SUMMARY AND FUTURE PROMISING PERSPECTIVES

We have seen that VO₂ thin films have been used for about a decade to dynamically tune and switch nanophotonic devices, both for integrated guided wave optics and for free-space optics. In the majority of cases, VO₂ was used as a simple on-off optical switch, driven thermally or optically. Given its natural functionalities, we feel this material is so far underused and its salient features such as the electrical control, the large hysteresis and the multilevel intermediate states deserve to be better exploited in nanophotonic devices. We review in this section different works that explore these directions that we find particularly promising.

A. Electrically-controlled tunable VO₂-based metasurfaces

As described previously, the IMT of VO₂ can be triggered via a large number of different stimuli. Among them, the electrical control of the state of VO₂ appears as the most useful implementation towards real world applications. Indeed, although it is much easier to demonstrate proof-of-concepts in a lab using controlled hot plates or lasers, many of the future applications should not rely on such external means of switching. Electrical control, on the other hand, is a widespread technique that is ubiquitous in modern devices. It is however challenging to create functional electrically-controlled VO₂ devices for different reasons. The first one is mainly technological: it is not straightforward to integrate electrodes with

VO₂, or even to nanopattern it altogether, because this material combines a high-reactivity with most of the wet-etchant chemicals used to process metals, and a good resistance to common dry-etching process. This implies that the technological processes for this platform are not yet mature and require internal developments for each laboratory. The second and most important reason is that a design that would work for thermal or optical switching has little chance to be readily adapted for electrical excitation. This is due to the presence of electrodes that obviously introduce differences in the overall optical properties. The architectures of devices should therefore be completely redesigned to properly take into account the influence of electrodes. This should not be seen as a limitation of the technology but rather as another layer of complexity that has to be thought through. In this section, we therefore review interesting advances in experimental demonstrations of electrically-controlled VO₂-based devices for nanophotonics.

One of the first works on the electrical control of VO₂ to tune an integrated optical device was reported by Driscoll et al. in 2009¹². In this study, the authors demonstrated a frequency-agile metamaterial based on split-ring resonators operating in the THz range (see Fig. 7 (a)). Using a simple planar electrode architecture at controlled temperatures, they were able to electrically-induce persistent tuning of the metamaterial's resonance. Following this pioneering demonstration, a few works reported the electrical control of VO₂ state on top of silicon-based waveguides, as described in section IV^{66,67}. More recently, three different groups reported experimental demonstrations of electrically-controlled VO₂-based nanophotonic devices. Liu et al. proposed in 2016¹¹⁶, a metal-insulator-metal configuration comprising a VO₂ thin film sandwiched in between metal antennas and a dielectric spacer, as displayed in Fig. 7 (d)-(e). Interestingly, the top metallic cross-shaped antennas are patterned as an array that connects them together, hence enabling electrical current to flow through the structure and to electrically trigger the IMT of VO₂. The authors then demonstrated a very large optical reflectance modulation, from ~0% to ~80% at a wavelength of 3 μm upon electrically-switching VO₂. This report was followed a year later (2017) by the experimental demonstration of a near-infrared spectral tuning of metadevices comprising VO₂ nano-elements placed at the feed gap of bow-tie antennas suspended membranes¹¹⁷. As shown in Figure 7 (b), VO₂ unit cells, about ~30nm-wide, are all electrically-connected via metallic lines and the spectral tuning is driven by Joule heating. This configuration presents the advantage of minimizing the volume of VO₂, hence reducing thermal mass, energy consumption and switching times (reported to be in the millisecond range). These two demonstrations were followed in 2019 by the report of an electrically-triggered modulation of optical phase in a one-dimensional metasurface array of metal-VO₂-insulator-metal waveguides (see Figs. 7 (f) and (g))¹¹⁸. In this study, the electrical control follows the same principle as in the two previous works: the patterned metallic structures on top of VO₂ serve as a means to distribute the flowing current throughout the device and trigger the VO₂ IMT via Joule heating. Using this reflectarray architecture, the

authors report a phase modulation as high as 250° , accompanied by spectral tuning and intensity modulation. This is the first demonstration of electrically tunable continuous optical phase modulation using VO_2 .

This "trilogy" of reports therefore nicely demonstrate that VO_2 -based devices enable the active modulation of intensity, spectrum and phase of reflected fields via electrical means. These works therefore pave the way for future efficient electrical control of different aspects of light fields for free space optics using metasurface-inspired configurations. Future works in that field may focus on improving metrics such as the optical efficiency and the switching time that are so far relatively modest. Designing devices that can separately modulate the amplitude and phase of reflected/transmitted optical fields via electrical control appear to be very challenging but if successful would unlock such technologies for multifunctional integrated tunable devices.

B. Tackling the absorption issue

The non-negligible optical absorption in both states of VO_2 could be a major drawback for most applications as a tunable optical medium. We review here two main strategies to mitigate this issue.

One straightforward solution is to design devices in spectral regions where the absorption is the lowest. As seen in Fig. 1 (g), the extinction coefficient of VO_2 in its insulating state slowly decreases at longer wavelengths and reaches a minimum down to $k \sim 0.07$ in the range 2-10 μm . When excited to the metallic state, the extinction coefficient dramatically increases, up to values of $k \sim 5-10$. In that range of the mid-IR, there is therefore room to design devices with low losses in one state and large absorption in the other state. This combination of features would be particularly useful for compact on-off modulators with large extinction ratio, both for guided and free-space optics. Looking back at the previously described results, we can indeed observe that the devices showing the highest modulation amplitudes are those designed for this wavelength range (see e.g. Fig. 4 (a-b) and Fig. 7 (e)) in which reflection modulation from $\sim 0\%$ to $\sim 80\%$ were demonstrated at wavelengths of 11 μm and 3 μm , respectively. This wavelength region appears also very promising for integrated guided wave optics, as many recent works demonstrated low-loss mid-IR waveguides on various platforms such as silicon, germanium or SiGe ¹¹⁹. We foresee that the use of VO_2 for mid-IR guided optics should lead to very interesting devices and large-scale tunable systems in the near future.

Another solution to avoid the large losses in VO_2 is to appropriately engineer the hybrid photonic structure of VO_2 . One way of doing so is to use ultra-thin films of VO_2 . Indeed, the complex refractive index modulation of VO_2 is so large that a few nanometers of material suffices for active tuning in many devices. As previously mentioned, VO_2 is a complex material whose growing conditions are far from being straightforward. Fabricating ultra-thin films of VO_2 is therefore challenging in itself. Quackenbush et al. have reported the successful growth of ultra-thin films (7.5-1 nm) of VO_2

by molecular beam epitaxy^{120,121}. Interestingly, they demonstrated that the IMT of VO_2 remains unchanged even for 1-nm-thick layers (i.e. about two unit cells). This important result holds promise for the future use of ultra-thin functional layers of VO_2 . Recently, Guo et al. reported the use of atomic layer deposition (ALD) to grow ultrathin layers of VO_2 ¹²². With this method, they demonstrated the conformal coating of VO_2 layers on ITO nanorods array (see Fig. 8 (a) & (b)). Such a growing technique appears promising to seamlessly implement ultrathin layers of VO_2 on nanostructures with an overall low optical absorption.

It was recently shown that VO_2 nanocrystals (VO_2 -NCs) implanted in SiO_2 as spherical nano-inclusions (see Fig. 8 (c)) provide an alternative path towards low-loss tunable media¹²³. By exploiting the VO_2 IMT, it is possible to tune and switch multipolar modes supported by VO_2 -NCs in the visible and to gradually produce a plasmonic mode in the NIR whose intensity is directly controlled by the VO_2 state. It was shown that the complex refractive index of such an effective medium (a slab of VO_2 nanospheres embedded in SiO_2) presents distinct optical tunability compared to unpatterned VO_2 . By adjusting the VO_2 -NCs size, the effective medium can be designed to have a large refractive-index tunability without inducing modulation of the extinction coefficient at specific wavelengths. This zero-induced-extinction refractive index tuning opens up new regimes of record large figure of merit ($\Delta n/\Delta k$) and designer optical tunability, unattainable with conventional unpatterned PCMs layers (see Fig. 8 (c)) and suggests a new practical direction to produce low-loss tunable optical metamaterials.

C. Spatially-addressed optical control of IMT and memory effects

As mentioned in previous sections, VO_2 is known to present a broad hysteresis in its IMT cycle. This hysteresis can be, to some extent, engineered via strain or doping^{14,15,35,124}. However, there is a surprisingly low number of papers that actually use this functionality in devices. Such an effect can be used to demonstrate memory effects. Lei et al. exploited this hysteresis to demonstrate an all-optical memory effect in hybrid plasmonic nanostructures¹⁰⁷. More recently, Fan et al. reported an optoelectronic memory device with electrical writing and optical reading using epitaxial VO_2 thin films grown on GaN ¹²⁵. The memory effect has also been demonstrated in hybrid VO_2/Si waveguides⁶². Such memory devices are still volatile in nature though, as their 'memory' state only lasts several microseconds at best. However, we describe in the following three recent works that elegantly exploited the hysteretic behavior of VO_2 to demonstrate all-optical non-volatile nanophotonic devices.

The VO_2 -NCs described in the previous section typically present an ultra-broad hysteresis behavior, with an IMT occurring at $\sim 80^\circ\text{C}$ upon heating and a relaxation to the insulating state at $\sim 25^\circ\text{C}$ upon cooling (see Fig. 9 (a)). Hence, by keeping the sample at temperatures as low as $\sim 30^\circ\text{C}$, it is possible to maintain a persistent metallic state in VO_2 -NCs. Jostmeier

et al. have exploited that functionality to optically imprint photonic elements onto an unpatterned VO₂-NCs platform¹²⁶. Using a visible laser scanned at specific points of the sample, they locally switch VO₂-NCs and define patterns such as gratings and zone plates, as shown in Fig. 9 (a). This original technique appears as a very flexible way to fabricate reconfigurable photonic devices, as they can later be erased by simply cooling down the sample to room temperature.

Two recent works used the same principle to optically imprint arbitrary reconfigurable patterns via locally switching the state of VO₂ thin films. The first one, reported in 2018 by Dong et al.¹²⁷, demonstrated the dynamic writing and erasing of arbitrary patterns and reconfigurable photonic devices such as beam-steerers, linear polarizers and concentric-ring gratings at a wavelength of 10.6 μm (see Fig. 9 (b)). Each of the patterns, fabricated using a 532 nm laser, can be 'stored' in the VO₂ film by maintaining the sample at temperatures around the IMT i.e. $\sim 60^\circ\text{C}$. They coined this reconfigurable VO₂-based platform a 'programmable metacanvas'. A following recent work used the same method to demonstrate a spatially-resolved control of thermal emission in a large wavelength range of the mid-IR (8-14 μm)¹²⁸. As shown in Fig. 9 (c) they exploited both the hysteresis and the intermediate phases of VO₂ to write non-volatile multilevel states in the thin layer, each producing a different level of thermal emission.

These three works demonstrate very promising ways to fully exploit VO₂ for its hysteretic behavior and large optical modulation in the mid-IR. One may find the need for temperature control to be inconvenient but this could alternatively be seen as a very practical method to easily erase and reconfigure devices. These demonstrations may pave the way for future real-time adaptive optical systems.

D. Ultra-fast switching time in integrated devices

Finally, a very important open question remains on whether ultra-fast switching times, below the picosecond range, are actually feasible in practical hybrid integrated devices. All-optical switching seems to be the most promising approach based on previous experiments on non-integrated devices (see for instance³⁹). However, ultra-fast all-optical switching on hybrid integrated devices has only been very recently demonstrated⁸¹. The key point is to control and minimize the thermal component associated with the VO₂ phase transition without penalizing the optical switching performance.

All in all, progresses towards a better use and integration of VO₂ in nanophotonic devices will go in parallel with progresses in better understanding and control of the complex physics of VO₂.

ACKNOWLEDGMENTS

SC acknowledges funding from the French National Research Agency (ANR) under the project SNAPSHOT (ANR-16-CE24-0004). ZZ and SR acknowledge AFOSR

FA9550-18-1-0250 for support. PS acknowledges Generalitat Valenciana (PROMETEO/2019/123) and Ministerio de Economía y Competitividad (MINECO/FEDER, UE) (TEC2016-76849). JP acknowledges the Ministerio de Ciencia, Innovación y Universidades for his FPU17/04224 grant. We thank M. Kats for fruitful discussions and feedback on the manuscript.

Data availability

Data sharing is not applicable to this article as no new data were created or analyzed in this study.

- ¹L. Novotny and B. Hecht, *Principles of nano-optics* (Cambridge university press, 2012).
- ²A. F. Koenderink, A. Alu, and A. Polman, "Nanophotonics: shrinking light-based technology," *Science* **348**, 516–521 (2015).
- ³J. W. Haus, *Fundamentals and applications of nanophotonics* (Woodhead Publishing, 2016).
- ⁴N. Yu and F. Capasso, "Flat optics with designer metasurfaces," *Nat. Mater.* **13**, 139–150 (2014).
- ⁵S. M. Kamali, E. Arbabi, A. Arbabi, and A. Faraon, "A review of dielectric optical metasurfaces for wavefront control," *Nanophotonics* **7**, 1041–1068 (2018).
- ⁶Z. Yang, C. Ko, and S. Ramanathan, "Oxide electronics utilizing ultrafast metal-insulator transitions," *Annual Review of Materials Research* **41**, 337–367 (2011).
- ⁷S. Raoux, "Phase change materials," *Annual Review of Materials Research* **39**, 25–48 (2009).
- ⁸F. Morin, "Oxides which show a metal-to-insulator transition at the neel temperature," *Physical review letters* **3**, 34 (1959).
- ⁹A. Barker Jr, H. Verleur, and H. Guggenheim, "Infrared optical properties of vanadium dioxide above and below the transition temperature," *Physical Review Letters* **17**, 1286 (1966).
- ¹⁰H. W. Verleur, A. Barker Jr, and C. Berglund, "Optical properties of $v o 2$ between 0.25 and 5 ev," *Physical Review* **172**, 788 (1968).
- ¹¹T. Driscoll, S. Palit, M. M. Qazilbash, M. Brehm, F. Keilmann, B.-G. Chae, S.-J. Yun, H.-T. Kim, S. Cho, N. M. Jokerst, *et al.*, "Dynamic tuning of an infrared hybrid-metamaterial resonance using vanadium dioxide," *Applied Physics Letters* **93**, 024101 (2008).
- ¹²T. 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**, 1518–1521 (2009).
- ¹³M. 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 $v o 2$ phase transition," *Optics express* **17**, 18330–18339 (2009).
- ¹⁴K. Liu, S. Lee, S. Yang, O. Delaire, and J. Wu, "Recent progresses on physics and applications of vanadium dioxide," *Materials Today* **21**, 875–896 (2018).
- ¹⁵R. Shi, N. Shen, J. Wang, W. Wang, A. Amini, N. Wang, and C. Cheng, "Recent advances in fabrication strategies, phase transition modulation, and advanced applications of vanadium dioxide," *Appl. Phys. Rev.* **6**, 011312 (2019).
- ¹⁶Y. Ke, S. Wang, G. Liu, M. Li, T. J. White, and Y. Long, "Vanadium dioxide: The multistimuli responsive material and its applications," *Small* **14**, 1802025 (2018).
- ¹⁷N. Mott, *Metal-insulator transitions* (CRC Press, 2004).
- ¹⁸M. Imada, A. Fujimori, and Y. Tokura, "Metal-insulator transitions," *Rev. Mod. Phys.* **70**, 1039 (1998).
- ¹⁹J. H. Park, J. M. Coy, T. S. Kasirga, C. Huang, Z. Fei, S. Hunter, and D. H. Cobden, "Measurement of a solid-state triple point at the metal-insulator transition in $v o 2$," *Nature* **500**, 431–434 (2013).
- ²⁰J. Pouget and H. Launois, "Metal-insulator phase transition in $v o 2$," *Le Journal de Physique Colloques* **37**, C4–49 (1976).
- ²¹M. Marezio, D. B. McWhan, J. Remeika, and P. Dernier, "Structural aspects of the metal-insulator transitions in cr-doped $v o 2$," *Physical Review B* **5**, 2541 (1972).
- ²²M. Ghedira, H. Vincent, M. Marezio, and J. Launay, "Structural aspects

- of the metal-insulator transitions in VO_2 ,” *Journal of Solid State Chemistry* **22**, 423–438 (1977).
- ²³R. E. Peierls, *Quantum theory of solids* (Clarendon Press, 1996).
- ²⁴T. Huffman, C. Hendriks, E. Walter, J. Yoon, H. Ju, R. Smith, G. Carr, H. Krakauer, and M. Qazilbash, “Insulating phases of vanadium dioxide are mott-hubbard insulators,” *Physical Review B* **95**, 075125 (2017).
- ²⁵V. Eyert, “The metal-insulator transitions of VO_2 : A band theoretical approach,” *Annalen der Physik* **11**, 650–704 (2002).
- ²⁶V. Eyert, “ VO_2 : a novel view from band theory,” *Physical Review Letters* **107**, 016401 (2011).
- ²⁷S. Biermann, A. Poteryaev, A. Lichtenstein, and A. Georges, “Dynamical singlets and correlation-assisted peierls transition in VO_2 ,” *Physical review letters* **94**, 026404 (2005).
- ²⁸Z. He and A. J. Millis, “Photoinduced phase transitions in narrow-gap mott insulators: The case of VO_2 ,” *Physical Review B* **93**, 115126 (2016).
- ²⁹M. van Veenendaal, “Ultrafast photoinduced insulator-to-metal transitions in vanadium dioxide,” *Physical Review B* **87**, 235118 (2013).
- ³⁰F. Grandi, A. Amaricci, and M. Fabrizio, “Unraveling the mott-peierls intrigue in vanadium dioxide,” *Physical Review Research* **2**, 013298 (2020).
- ³¹V. R. Morrison, R. P. Chatelain, K. L. Tiwari, A. Hendaoui, A. Bruhács, M. Chaker, and B. J. Siwick, “A photoinduced metal-like phase of monoclinic VO_2 revealed by ultrafast electron diffraction,” *Science* **346**, 445–448 (2014).
- ³²R. Yoshida, T. Yamamoto, Y. Ishida, H. Nagao, T. Otsuka, K. Saeki, Y. Mu-raoka, R. Eguchi, K. Ishizaka, T. Kiss, *et al.*, “Ultrafast photoinduced transition of an insulating VO_2 thin film into a nonrutile metallic state,” *Physical Review B* **89**, 205114 (2014).
- ³³T. Cocker, L. Titova, S. Fourmaux, G. Holloway, H.-C. Bandulet, D. Bras-sard, J.-C. Kieffer, M. El Khakani, and F. Hegmann, “Phase diagram of the ultrafast photoinduced insulator-metal transition in vanadium dioxide,” *Physical Review B* **85**, 155120 (2012).
- ³⁴Z. Tao, T.-R. T. Han, S. D. Mahanti, P. M. Duxbury, F. Yuan, C.-Y. Ruan, K. Wang, and J. Wu, “Decoupling of structural and electronic phase transitions in VO_2 ,” *Physical review letters* **109**, 166406 (2012).
- ³⁵J. Laverock, S. Kittiwatanakul, A. Zakharov, Y. Niu, B. Chen, S. Wolf, J. Lu, and K. Smith, “Direct observation of decoupled structural and elec-tronic transitions and an ambient pressure monocliniclike metallic phase of VO_2 ,” *Physical review letters* **113**, 216402 (2014).
- ³⁶C. Ko and S. Ramanathan, “Observation of electric field-assisted phase transition in thin film vanadium oxide in a metal-oxide-semiconductor de-vice geometry,” *Applied Physics Letters* **93**, 252101 (2008).
- ³⁷Y. Zhou, X. Chen, C. Ko, Z. Yang, C. Mouli, and S. Ra-manathan, “Voltage-triggered ultrafast phase transition in vanadium diox-ide switches,” *IEEE Electron Device Letters* **34**, 220–222 (2013).
- ³⁸G. Stefanovich, A. Pergament, and D. Stefanovich, “Electrical switching and mott transition in VO_2 ,” *Journal of Physics: Condensed Matter* **12**, 8837 (2000).
- ³⁹A. Cavalleri, C. Tóth, C. W. Siders, J. Squier, F. Ráksi, P. Forget, and J. Kieffer, “Femtosecond structural dynamics in VO_2 during an ultrafast solid-solid phase transition,” *Physical review letters* **87**, 237401 (2001).
- ⁴⁰A. Cavalleri, T. Dekorsy, H. H. Chong, J.-C. Kieffer, and R. W. Schoen-lein, “Evidence for a structurally-driven insulator-to-metal transition in VO_2 : A view from the ultrafast timescale,” *Physical Review B* **70**, 161102 (2004).
- ⁴¹M. Liu, H. Y. Hwang, H. Tao, A. C. Strikwerda, K. Fan, G. R. Keiser, A. J. Sternbach, K. G. West, S. Kittiwatanakul, J. Lu, *et al.*, “Terahertz-field-induced insulator-to-metal transition in vanadium dioxide metamaterial,” *Nature* **487**, 345–348 (2012).
- ⁴²H. Kakiuchida, P. Jin, S. Nakao, and M. Tazawa, “Optical properties of vanadium dioxide film during semiconductive–metallic phase transition,” *Japanese Journal of Applied Physics* **46**, L113 (2007).
- ⁴³J. Sun and G. K. Pribil, “Analyzing optical properties of thin vanadium oxide films through semiconductor-to-metal phase transition using spec-troscopic ellipsometry,” *Appl. Surf. Sci.* **421**, 819–823 (2017).
- ⁴⁴C. Wan, Z. Zhang, D. Woolf, C. M. Hessel, J. Rensberg, J. M. Hensley, Y. Xiao, A. Shahsafi, J. Salman, S. Richter, *et al.*, “On the optical prop-erties of thin-film vanadium dioxide from the visible to the far infrared,” *Annalen der Physik* **531**, 1900188 (2019).
- ⁴⁵S. Amador-Alvarado, J. Flores-Camacho, A. Solís-Zamudio, R. Castro-García, J. Pérez-Huerta, E. Antúnez-Cerón, J. Ortega-Gallegos, J. Madrigal-Melchor, V. Agarwal, and D. Ariza-Flores, “Temperature-dependent infrared ellipsometry of VO_2 thin films across the insulator to metal transition,” *Scientific Reports* **10**, 1–11 (2020).
- ⁴⁶J. Swann and D. De Smet, “Ellipsometric investigation of vanadium diox-ide films,” *Journal of applied physics* **58**, 1335–1338 (1985).
- ⁴⁷M. Motyka, B. Gauntt, M. W. Horn, E. Dickey, and N. Podraza, “Mi-crostructural evolution of thin film vanadium oxide prepared by pulsed-direct current magnetron sputtering,” *Journal of Applied Physics* **112**, 093504 (2012).
- ⁴⁸M. M. Qazilbash, M. Brehm, B.-G. Chae, P.-C. Ho, G. O. Andreev, B.-J. Kim, S. J. Yun, A. Balatsky, M. Maple, F. Keilmann, *et al.*, “Mott transition in VO_2 revealed by infrared spectroscopy and nano-imaging,” *Science* **318**, 1750–1753 (2007).
- ⁴⁹A. Perucchi, L. Baldassarre, P. Postorino, and S. Lupi, “Optical properties across the insulator to metal transitions in vanadium oxide compounds,” *Journal of Physics: Condensed Matter* **21**, 323202 (2009).
- ⁵⁰J. Wei, H. Ji, W. Guo, A. H. Nevidomskyy, and D. Natelson, “Hydrogen stabilization of metallic vanadium dioxide in single-crystal nanobeams,” *Nature nanotechnology* **7**, 357–362 (2012).
- ⁵¹H. Yoon, M. Choi, T.-W. Lim, H. Kwon, K. Ihm, J. K. Kim, S.-Y. Choi, and J. Son, “Reversible phase modulation and hydrogen storage in multi-valent VO_2 epitaxial thin films,” *Nature materials* **15**, 1113–1119 (2016).
- ⁵²S. A. Donges, O. Khatib, B. T. O’Callahan, J. M. Atkin, J. H. Park, D. Cob-den, and M. B. Raschke, “Ultrafast nanoimaging of the photoinduced phase transition dynamics in VO_2 ,” *Nano letters* **16**, 3029–3035 (2016).
- ⁵³S. Zhang, J. Y. Chou, and L. J. Lauhon, “Direct correlation of structural domain formation with the metal insulator transition in a VO_2 nanobeam,” *Nano letters* **9**, 4527–4532 (2009).
- ⁵⁴R. M. Briggs, I. M. Pryce, and H. A. Atwater, “Compact silicon pho-tonic waveguide modulator based on the vanadium dioxide metal-insulator phase transition,” *Optics express* **18**, 11192–11201 (2010).
- ⁵⁵J. D. Ryckman, K. A. Hallman, R. E. Marvel, R. F. Haglund, and S. M. Weiss, “Ultra-compact silicon photonic devices reconfigured by an optically induced semiconductor-to-metal transition,” *Optics express* **21**, 10753–10763 (2013).
- ⁵⁶G. T. Reed, G. Mashanovich, F. Y. Gardes, and D. Thomson, “Silicon optical modulators,” *Nature photonics* **4**, 518–526 (2010).
- ⁵⁷K. Yao, R. Unni, and Y. Zheng, “Intelligent nanophotonics: merging pho-tonics and artificial intelligence at the nanoscale,” *Nanophotonics* **8**, 339–366 (2019).
- ⁵⁸J. Wang, F. Sciarrino, A. Laing, and M. G. Thompson, “Integrated pho-tonic quantum technologies,” *Nature Photonics* **1**, 1–12 (2019).
- ⁵⁹K. Liu, C. R. Ye, S. Khan, and V. J. Sorger, “Review and perspective on ultrafast wavelength-size electro-optic modulators,” *Laser & Photonics Reviews* **9**, 172–194 (2015).
- ⁶⁰K. Shibuya, Y. Atsumi, T. Yoshida, Y. Sakakibara, M. Mori, and A. Sawa, “Silicon waveguide optical modulator driven by metal–insulator transi-tion of vanadium dioxide cladding layer,” *Optics express* **27**, 4147–4156 (2019).
- ⁶¹K. J. Miller, K. A. Hallman, R. F. Haglund, and S. M. Weiss, “Silicon waveguide optical switch with embedded phase change material,” *Optics express* **25**, 26527–26536 (2017).
- ⁶²I. Olivares, L. Sánchez, J. Parra, R. Larrea, A. Griol, M. Menghini, P. Homm, L.-W. Jang, B. van Bilzen, J. W. Seo, *et al.*, “Optical switching in hybrid VO_2/Si waveguides thermally triggered by lateral microheaters,” *Optics express* **26**, 12387–12395 (2018).
- ⁶³L. D. Sánchez, I. Olivares, J. Parra, M. Menghini, P. Homm, J.-P. Locquet, and P. Sanchis, “Experimental demonstration of a tunable transverse elec-tric pass polarizer based on hybrid VO_2/Si technology,” *Optics letters* **43**, 3650–3653 (2018).
- ⁶⁴V. Jeyaselvan, A. Pal, P. A. Kumar, and S. K. Selvaraja, “Thermally-induced optical modulation in a vanadium dioxide-on-silicon waveguide,” *OSA Continuum* **3**, 132–142 (2020).
- ⁶⁵L. Sanchez, S. Lechago, A. Gutierrez, and P. Sanchis, “Analysis and de-sign optimization of a hybrid VO_2/Si microring switch,” *IEEE Photonics Journal* **8**, 1–9 (2016).
- ⁶⁶P. Markov, R. E. Marvel, H. J. Conley, K. J. Miller, R. F. Haglund Jr, and S. M. Weiss, “Optically monitored electrical switching in VO_2 ,” *Acs Photonics* **2**, 1175–1182 (2015).
- ⁶⁷A. Joushaghani, J. Jeong, S. Paradis, D. Alain, J. S. Aitchison, and J. K.

- Poon, "Wavelength-size hybrid si-vo 2 waveguide electroabsorption optical switches and photodetectors," *Optics express* **23**, 3657–3668 (2015).
- ⁶⁸J. D. Ryckman, V. Diez-Blanco, J. Nag, R. E. Marvel, B. Choi, R. F. Haglund, and S. M. Weiss, "Photothermal optical modulation of ultra-compact hybrid si-vo 2 ring resonators," *Optics express* **20**, 13215–13225 (2012).
- ⁶⁹H. M. Wong, Z. Yan, K. A. Hallman, R. E. Marvel, R. P. Prasankumar, R. F. Haglund Jr, and A. S. Helmy, "Broadband, integrated, micron-scale, all-optical si₃n₄/vo₂ modulators with pj switching energy," *ACS Photonics* **6**, 2734–2740 (2019).
- ⁷⁰B. Janjan, M. Miri, D. Fathi, M. Heidari, and D. Abbott, "Hybrid si₃n₄ modulator thermally triggered by a graphene microheater," *IEEE Journal of Selected Topics in Quantum Electronics* (2020).
- ⁷¹M. Sadeghi, B. Janjan, M. Heidari, and D. Abbott, "Mid-infrared hybrid si/vo 2 modulator electrically driven by graphene electrodes," *Optics Express* **28**, 9198–9207 (2020).
- ⁷²J. T. Kim, "Cmos-compatible hybrid plasmonic modulator based on vanadium dioxide insulator-metal phase transition," *Optics letters* **39**, 3997–4000 (2014).
- ⁷³P. Markov, K. Appavoo, R. F. Haglund, and S. M. Weiss, "Hybrid si-vo 2-au optical modulator based on near-field plasmonic coupling," *Optics Express* **23**, 6878–6887 (2015).
- ⁷⁴J.-H. Choe and J. T. Kim, "Design of vanadium dioxide-based plasmonic modulator for both te and tm modes," *IEEE Photonics Technology Letters* **27**, 514–517 (2014).
- ⁷⁵S. Mohammadi-Pouyan, M. Miri, and M. H. Sheikhi, "Design of a vanadium dioxide-based dual-polarization optical pam4 modulator," *JOSA B* **35**, 3094–3103 (2018).
- ⁷⁶B. Younis, A. Heikal, M. Hussein, S. Obayya, and M. F. O. Hameed, "Hybrid si-vo 2 modulator with ultra-high extinction ratio based on slot tm mode," *Optics Express* **27**, 37454–37468 (2019).
- ⁷⁷L. Sánchez, A. Rosa, A. Griol, A. Gutierrez, P. Himm, B. Van Bilzen, M. Menghini, J.-P. Locquet, and P. Sanchis, "Impact of the external resistance on the switching power consumption in vo₂ nano gap junctions," *Applied Physics Letters* **111**, 031904 (2017).
- ⁷⁸L. Chen, H. Ye, Y. Liu, D. Wu, R. Ma, and Z. Yu, "Numerical investigations of an optical switch based on a silicon stripe waveguide embedded with vanadium dioxide layers," *Photonics Research* **5**, 335–339 (2017).
- ⁷⁹J. K. Clark, Y.-L. Ho, H. Matsui, and J.-J. Delaunay, "Optically pumped hybrid plasmonic-photon waveguide modulator using the vo₂ metal-insulator phase transition," *IEEE Photonics Journal* **10**, 1–9 (2017).
- ⁸⁰R. F. Haglund, K. A. Hallman, K. J. Miller, and S. M. Weiss, "Picosecond optical switching in silicon photonics using phase-changing vanadium dioxide," in *CLEO: Science and Innovations* (Optical Society of America, 2019) pp. STh4H–1.
- ⁸¹K. A. Hallman, K. J. Miller, A. Baydin, S. M. Weiss, and R. F. Haglund, "Sub-picosecond all-optical switching in a hybrid vo₂: silicon waveguide at 1550 nm," *arXiv preprint arXiv:2007.02812* (2020).
- ⁸²J. Parra, T. Ivanova, M. Menghini, P. Himm, J.-P. Locquet, and P. Sanchis, "Temporal dynamics of all-optical switching in hybrid vo₂/si waveguides," *arXiv preprint arXiv:2007.11452* (2020).
- ⁸³D. Wegkamp, M. Herzog, L. Xian, M. Gatti, P. Cudazzo, C. L. McGahan, R. E. Marvel, R. F. Haglund Jr, A. Rubio, M. Wolf, *et al.*, "Instantaneous band gap collapse in photoexcited monoclinic vo₂ due to photocarrier doping," *Physical review letters* **113**, 216401 (2014).
- ⁸⁴Z. Tao, F. Zhou, T.-R. T. Han, D. Torres, T. Wang, N. Sepulveda, K. Chang, M. Young, R. R. Lunt, and C.-Y. Ruan, "The nature of photoinduced phase transition and metastable states in vanadium dioxide," *Scientific reports* **6**, 1–10 (2016).
- ⁸⁵M. R. Otto, L. P. R. de Cotret, D. A. Valverde-Chavez, K. L. Tiwari, N. Émond, M. Chaker, D. G. Cooke, and B. J. Siwick, "How optical excitation controls the structure and properties of vanadium dioxide," *Proceedings of the National Academy of Sciences* **116**, 450–455 (2019).
- ⁸⁶S. Wang, P. C. Wu, V.-C. Su, Y.-C. Lai, M.-K. Chen, H. Y. Kuo, B. H. Chen, Y. H. Chen, T.-T. Huang, J.-H. Wang, *et al.*, "A broadband achromatic metalens in the visible," *Nature nanotechnology* **13**, 227–232 (2018).
- ⁸⁷A. Arbabi, E. Arbabi, Y. Horie, S. M. Kamali, and A. Faraon, "Planar metasurface retroreflector," *Nature Photonics* **11**, 415 (2017).
- ⁸⁸G. Zheng, H. Mühlenbernd, M. Kenney, G. Li, T. Zentgraf, and S. Zhang, "Metasurface holograms reaching 80% efficiency," *Nature nanotechnology* **10**, 308–312 (2015).
- ⁸⁹M. A. Kats, D. Sharma, J. Lin, P. Genevet, R. Blanchard, Z. Yang, M. M. Qazilbash, D. Basov, S. Ramanathan, and F. Capasso, "Ultra-thin perfect absorber employing a tunable phase change material," *Applied Physics Letters* **101**, 221101 (2012).
- ⁹⁰N. A. Butakov, M. W. Knight, T. Lewi, P. P. Iyer, D. Higgs, H. T. Chorsi, J. Trastoy, J. Del Valle Granda, I. Valmianski, C. Urban, *et al.*, "Broadband electrically tunable dielectric resonators using metal-insulator transitions," *Acs Photonics* **5**, 4056–4060 (2018).
- ⁹¹J. Rensberg, Y. Zhou, S. Richter, C. Wan, S. Zhang, P. Schöppe, R. Schmidt-Grund, S. Ramanathan, F. Capasso, M. A. Kats, *et al.*, "Epsilon-near-zero substrate engineering for ultrathin-film perfect absorbers," *Physical Review Applied* **8**, 014009 (2017).
- ⁹²M. A. Kats, R. Blanchard, S. Zhang, P. Genevet, C. Ko, S. Ramanathan, and F. Capasso, "Vanadium dioxide as a natural disordered metamaterial: perfect thermal emission and large broadband negative differential thermal emittance," *Physical Review X* **3**, 041004 (2013).
- ⁹³C. Wan, E. H. Horak, J. King, J. Salman, Z. Zhang, Y. Zhou, P. Roney, B. Gundlach, S. Ramanathan, R. H. Goldsmith, *et al.*, "Limiting optical diodes enabled by the phase transition of vanadium dioxide," *ACS Photonics* **5**, 2688–2692 (2018).
- ⁹⁴K. Joulain, Y. Ezzahri, J. Drevillon, and P. Ben-Abdallah, "Modulation and amplification of radiative far field heat transfer: Towards a simple radiative thermal transistor," *Applied Physics Letters* **106**, 133505 (2015).
- ⁹⁵J. Rensberg, S. Zhang, Y. Zhou, A. S. McLeod, C. Schwarz, M. Goldflam, M. Liu, J. Kerbusch, R. Nawrodt, S. Ramanathan, *et al.*, "Active optical metasurfaces based on defect-engineered phase-transition materials," *Nano letters* **16**, 1050–1055 (2016).
- ⁹⁶F. Ligmajer, L. Kejik, U. Tiwari, M. Qiu, J. Nag, M. Konecny, T. Sikola, W. Jin, R. F. Haglund Jr, K. Appavoo, *et al.*, "Epitaxial vo₂ nanostructures: A route to large-scale, switchable dielectric metasurfaces," *ACS photonics* **5**, 2561–2567 (2018).
- ⁹⁷T. G. Folland, A. Fali, S. T. White, J. R. Matson, S. Liu, N. A. Aghamiri, J. H. Edgar, R. F. Haglund, Y. Abate, and J. D. Caldwell, "Reconfigurable infrared hyperbolic metasurfaces using phase change materials," *Nature communications* **9**, 1–7 (2018).
- ⁹⁸Z. Zhang, F. Zuo, C. Wan, A. Dutta, J. Kim, J. Rensberg, R. Nawrodt, H. H. Park, T. J. Larrabee, X. Guan, *et al.*, "Evolution of metallicity in vanadium dioxide by creation of oxygen vacancies," *Physical Review Applied* **7**, 034008 (2017).
- ⁹⁹S. Cuffe, D. Li, Y. Zhou, F. J. Wong, J. A. Kurvits, S. Ramanathan, and R. Zia, "Dynamic control of light emission faster than the lifetime limit using vo₂ phase-change," *Nat. Commun.* **6**, 8636 (2015).
- ¹⁰⁰E. Petronijevic, M. Centini, T. Cesca, G. Mattei, F. A. Bovino, and C. Sibilia, "Control of an nanoantenna emission enhancement of magnetic dipolar emitters by means of vo₂ phase change layers," *Optics express* **27**, 24260–24273 (2019).
- ¹⁰¹D. Szilard, W. Kort-Kamp, F. Rosa, F. Pinheiro, and C. Farina, "Hysteresis in the spontaneous emission induced by vo₂ phase change," *JOSA B* **36**, C46–C51 (2019).
- ¹⁰²S. K. Chamoli, M. ElKabbash, J. Zhang, and C. Guo, "Dynamic control of spontaneous emission rate using tunable hyperbolic metamaterials," *Optics Letters* **45**, 1671–1674 (2020).
- ¹⁰³P. K. Jha, H. Akbari, Y. Kim, and H. A. Atwater, "Nanoscale axial position and orientation measurement of hexagonal boron nitride quantum emitters using a tunable nanophotonic environment," *arXiv preprint arXiv:2007.07811* (2020).
- ¹⁰⁴S. K. Earl, T. D. James, T. J. Davis, J. C. McCallum, R. E. Marvel, R. F. Haglund, and A. Roberts, "Tunable optical antennas enabled by the phase transition in vanadium dioxide," *Optics express* **21**, 27503–27508 (2013).
- ¹⁰⁵M. Seo, J. Kyoung, H. Park, S. Koo, H.-s. Kim, H. Bernien, B. J. Kim, J. H. Choe, Y. H. Ahn, H.-T. Kim, *et al.*, "Active terahertz nanoantennas based on vo₂ phase transition," *Nano letters* **10**, 2064–2068 (2010).
- ¹⁰⁶K. Appavoo and R. F. Haglund Jr, "Detecting nanoscale size dependence in vo₂ phase transition using a split-ring resonator metamaterial," *Nano letters* **11**, 1025–1031 (2011).
- ¹⁰⁷D. Y. Lei, K. Appavoo, F. Ligmajer, Y. Sonnefraud, R. F. Haglund Jr, and S. A. Maier, "Optically-triggered nanoscale memory effect in a hybrid plasmonic-phase changing nanostructure," *ACS photonics* **2**, 1306–1313 (2015).

This is the author's peer reviewed, accepted manuscript. However, the online version of record will be different from this version once it has been copyedited and typeset.

PLEASE CITE THIS ARTICLE AS DOI:10.1063/1.50028093

- ¹⁰⁸S. Song, X. Ma, M. Pu, X. Li, Y. Guo, P. Gao, and X. Luo, "Tailoring active color rendering and multiband photodetection in a vanadium-dioxide-based metamaterial absorber," *Photonics Research* **6**, 492–497 (2018).
- ¹⁰⁹F.-Z. Shu, F.-F. Yu, R.-W. Peng, Y.-Y. Zhu, B. Xiong, R.-H. Fan, Z.-H. Wang, Y. Liu, and M. Wang, "Dynamic plasmonic color generation based on phase transition of vanadium dioxide," *Advanced Optical Materials* **6**, 1700939 (2018).
- ¹¹⁰G. Kaplan, K. Aydin, and J. Scheuer, "Dynamically controlled plasmonic nano-antenna phased array utilizing vanadium dioxide," *Optical Materials Express* **5**, 2513–2524 (2015).
- ¹¹¹S. K. Earl, T. D. James, D. E. Gómez, R. E. Marvel, R. F. Haglund Jr, and A. Roberts, "Switchable polarization rotation of visible light using a plasmonic metasurface," *Apl Photonics* **2**, 016103 (2017).
- ¹¹²S.-J. Kim, H. Yun, K. Park, J. Hong, J.-G. Yun, K. Lee, J. Kim, S. J. Jeong, S.-E. Mun, J. Sung, *et al.*, "Active directional switching of surface plasmon polaritons using a phase transition material," *Scientific reports* **7**, 43723 (2017).
- ¹¹³N. A. Butakov, I. Valmianski, T. Lewi, C. Urban, Z. Ren, A. A. Mikhailovsky, S. D. Wilson, I. K. Schuller, and J. A. Schuller, "Switchable plasmonic–dielectric resonators with metal–insulator transitions," *Acs Photonics* **5**, 371–377 (2018).
- ¹¹⁴O. L. Muskens, L. Bergamini, Y. Wang, J. M. Gaskell, N. Zabala, C. De Groot, D. W. Sheel, and J. Aizpurua, "Antenna-assisted picosecond control of nanoscale phase transition in vanadium dioxide," *Light: Science & Applications* **5**, e16173–e16173 (2016).
- ¹¹⁵M. A. Kats, R. Blanchard, P. Genevet, Z. Yang, M. M. Qazilbash, D. Basov, S. Ramanathan, and F. Capasso, "Thermal tuning of mid-infrared plasmonic antenna arrays using a phase change material," *Optics letters* **38**, 368–370 (2013).
- ¹¹⁶L. Liu, L. Kang, T. S. Mayer, and D. H. Werner, "Hybrid metamaterials for electrically triggered multifunctional control," *Nature communications* **7**, 1–8 (2016).
- ¹¹⁷Z. Zhu, P. G. Evans, R. F. Haglund Jr, and J. G. Valentine, "Dynamically reconfigurable metadevice employing nanostructured phase-change materials," *Nano Lett.* **17**, 4881–4885 (2017).
- ¹¹⁸Y. Kim, P. C. Wu, R. Sokhoyan, K. Mauser, R. Glauddell, G. Kafaie Shirmanesh, and H. A. Atwater, "Phase modulation with electrically tunable vanadium dioxide phase-change metasurfaces," *Nano letters* **19**, 3961–3968 (2019).
- ¹¹⁹R. Soref, "Mid-infrared photonics in silicon and germanium," *Nature photonics* **4**, 495–497 (2010).
- ¹²⁰N. Quackenbush, J. W. Tashman, J. A. Mundy, S. Sallis, H. Paik, R. Misra, J. A. Moyer, J.-H. Guo, D. A. Fischer, J. C. Woicik, *et al.*, "Nature of the metal insulator transition in ultrathin epitaxial vanadium dioxide," *Nano letters* **13**, 4857–4861 (2013).
- ¹²¹N. F. Quackenbush, H. Paik, J. C. Woicik, D. A. Arena, D. G. Schlom, and L. F. Piper, "X-ray spectroscopy of ultra-thin oxide/oxide heteroepitaxial films: a case study of single-nanometer vo₂/tio₂," *Materials* **8**, 5452–5466 (2015).
- ¹²²P. Guo, M. S. Weimer, J. D. Emery, B. T. Diroll, X. Chen, A. S. Hock, R. P. Chang, A. B. Martinson, and R. D. Schaller, "Conformal coating of a phase change material on ordered plasmonic nanorod arrays for broadband all-optical switching," *ACS nano* **11**, 693–701 (2017).
- ¹²³J. John, Y. Gutierrez, Z. Zhang, H. Karl, S. Ramanathan, R. Orobtcouk, F. Moreno, and S. Cueff, "Multipolar resonances with designer tunability using vo₂ phase-change materials," *Physical Review Applied* **13**, 044053 (2020).
- ¹²⁴X. Tan, T. Yao, R. Long, Z. Sun, Y. Feng, H. Cheng, X. Yuan, W. Zhang, Q. Liu, C. Wu, *et al.*, "Unraveling metal-insulator transition mechanism of vo₂ triggered by tungsten doping," *Scientific reports* **2**, 1–6 (2012).
- ¹²⁵L. Fan, Y. Chen, Q. Liu, S. Chen, L. Zhu, Q. Meng, B. Wang, Q. Zhang, H. Ren, and C. Zou, "Infrared response and optoelectronic memory device fabrication based on epitaxial vo₂ film," *ACS applied materials & interfaces* **8**, 32971–32977 (2016).
- ¹²⁶T. Jostmeier, J. Zimmer, H. Karl, H. J. Krenner, and k. Betz, "Optically imprinted reconfigurable photonic elements in a vo₂ nanocomposite," *Appl. Phys. Lett.* **105**, 071107 (2014).
- ¹²⁷K. Dong, S. Hong, Y. Deng, H. Ma, J. Li, X. Wang, J. Yeo, L. Wang, S. Lou, K. B. Tom, *et al.*, "A lithography-free and field-programmable photonic metacanvas," *Advanced Materials* **30**, 1703878 (2018).
- ¹²⁸Z. Xu, Q. Li, K. Du, S. Long, Y. Yang, X. Cao, H. Luo, H. Zhu, P. Ghosh, W. Shen, *et al.*, "Spatially resolved dynamically reconfigurable multilevel control of thermal emission," *Laser & Photonics Reviews* **14**, 1900162 (2020).

This is the author's peer reviewed, accepted manuscript. However, the online version of record will be different from this version once it has been copyedited and typeset.

PLEASE CITE THIS ARTICLE AS DOI:10.1063/1.50028093

FIG. 1. Optical dispersion of VO₂ thin film on sapphire, as extracted from temperature-controlled spectroscopic ellipsometry measurements. (a) Refractive index n , (b) extinction coefficient k , (c) real part of permittivity ϵ_1 , (d) imaginary part of permittivity ϵ_2 . (e) Scanning near-field infrared microscopy images showing the coexistence of insulating and metallic domains, with the progressive appearance of nanoscale metallic regions (represented as light blue, green and red colors) during the IMT (adapted from⁴⁸). (f) Thermal hysteresis of the extinction coefficient (k) of VO₂ film on Si and sapphire substrates at $\lambda=1350\text{nm}$, showing differences in optical properties and in phase transition behavior (adapted from⁴³). (g) and (h) Optical dispersion of VO₂ deposited on silicon and sapphire in the mid- and far-infrared regions (adapted from⁴⁴).

FIG. 2. Scheme of a hybrid VO₂/Si waveguide and different external excitations to control its properties.

FIG. 3. Hybrid VO₂/Si waveguide devices. The first and most of the proposed devices have been based on thermally triggering the VO₂ phase transition with switching times in the microsecond range. (a) The most common hybrid waveguides are developed by depositing the VO₂ on top of (a) rib⁵⁴ or (b) strip silicon waveguides⁶⁰. (c) Shorter devices have been achieved by embedding the VO₂ within the silicon waveguide⁶¹. (d) Enhanced optical switching performance has also been demonstrated by engineering the morphology of the VO₂ layer⁶². In addition, novel applications have also arisen such as (e) polarizers⁶³ and (f) switches based on add-drop ring resonators^{64,65}. Electrically-controlled devices have been investigated for enabling faster switching times. In this case, an electric field is applied between two separated metallic contacts on top of the hybrid VO₂/Si waveguide as seen in (g)⁶⁶ and (h)⁶⁷. All-optical switching schemes could also be a promising route towards ultra-fast speed. (i) Most of the works have been based on pumping the VO₂ with out-of-plane approaches such^{55,68}. (j) Recently, all-optical switching using an in-plane approach has also been demonstrated in a hybrid SiN waveguide⁶⁹. However, the timescale of the device was not reported. The feasibility of ultra-fast time-scales modulation still remains an open question.

FIG. 4. Tunable metasurface using unpatterned VO₂ thin films. (a) and (b) Perfect absorber based on VO₂⁸⁹. (a) Experimental setup on measuring the reflectivity of a 180 nm VO₂ grown on sapphire substrate. (b) Temperature dependent reflectivity spectrum of devices where the reflectivity at 343 K approaches zero at $\lambda=11.6 \mu\text{m}$. (c) and (d) Perfect thermal emission of 150 nm VO₂ deposited on sapphire substrate observed during heating from 35 to 74.5°C (c) and from 74.5 – 100°C (d)⁹². (e)-(g) Substrate engineering of VO₂ based thin film absorbers where the temperature dependent mid IR reflectance of VO₂ on AZO showing tunable plasmonic (e), VO₂ on SiO₂ with tunable phononic (f) and VO₂ on ZnO with tunable transparency (g)⁹¹. (h) and (i) Limiting optical diode made from VO₂, where the intense backward illumination triggers the onset of metallic phase leading to reduced transmission, while VO₂ maintains insulating when illuminating from forward direction resulting in high transmission⁹³. (j) and (k) Radiative thermal transistor based on metal insulator transition of VO₂⁹⁴. The transistor geometry is shown in (j) where a phase changing material e.g. VO₂ is placed between two blackbodies. In the transition regime of VO₂, the amplification factor α of such thermal transistor is larger than 1 (k). (l)-(n) Active metasurface built on defect engineered VO₂⁹⁵. (i) Ion beam irradiation through a mask creates defective region with lower transition temperature. The pattern created by the pristine and irradiated VO₂ during heating (n) gives rise to a tunable polarization-dependent reflectance at $\lambda=11 \mu\text{m}$ (m). (o) and (p) Tunable metasurface based on nanostructured VO₂ thin films⁹⁶. (o) Metasurface with VO₂ nanobeams grown epitaxially on a-cut sapphire substrate. (p) The extinction spectra of such device at room temperature and 80°C with incident light polarized perpendicular or parallel with the nanobeams, where the broad extinction peak at 1480 nm in metallic state with perpendicular light is attributed to a localized surface plasmon resonance. (q) Tunable infrared hyperbolic metasurface using VO₂, where hBN is transferred on a VO₂ film and polaritons are imaged by the s-SNOM⁹⁷. The changing in local dielectric environment from metallic and dielectric domains in VO₂ enables reconfigurable control of in-plane hyperbolic phonon polariton propagation.

FIG. 5. Various methods and architectures for dynamically tuning the spontaneous emission using VO₂ thin films. (a) a multilayer stack comprising a metallic mirror, a VO₂ layer and an Erbium thin film emitter⁹⁹ enables an all-optical direct modulation of spontaneous emission. (b) This modulation was experimentally demonstrated to be more than three orders of magnitude faster than the excited state lifetime of the erbium emitters (c)-(e) A similar multilayer platform with an additional plasmonic antenna on top to further enhance the spontaneous emission rates¹⁰⁰. (f)-(g) Enhancement of the ED and MD emission rates of quantum emitters in the vicinity of a VO₂ thin film during the IMT¹⁰¹. (h)-(i) A multilayer stack whose optical dispersion is modulated from elliptical to hyperbolic via the change of phase of VO₂. The spontaneous emission rate of emitters is calculated to be affected by this dispersion modification¹⁰²

FIG. 6. Selected examples of tunable plasmonic antennas using VO₂. In (a)-(h), different architectures and geometries of metallic antennas on top of VO₂. The four selected examples have the same functionalities: a dynamic modulation of transmission/reflection through the IMT of VO₂. By appropriately designing antennas, different spectral regions can be targeted from visible to teraHertz ranges. (a)-(b) Visible range using Silver nanorod¹⁰⁴; (c)-(d) NIR with split-ring resonator¹³; (e)-(f) MIR with Y-shaped antennas¹¹⁵; (g)-(h) TeraHertz with gold slot antennas array¹⁰⁵. Other functionalities were demonstrated such as (i) switchable polarization rotation¹¹¹ or (j) active directional switching of surface plasmon polaritons¹¹²

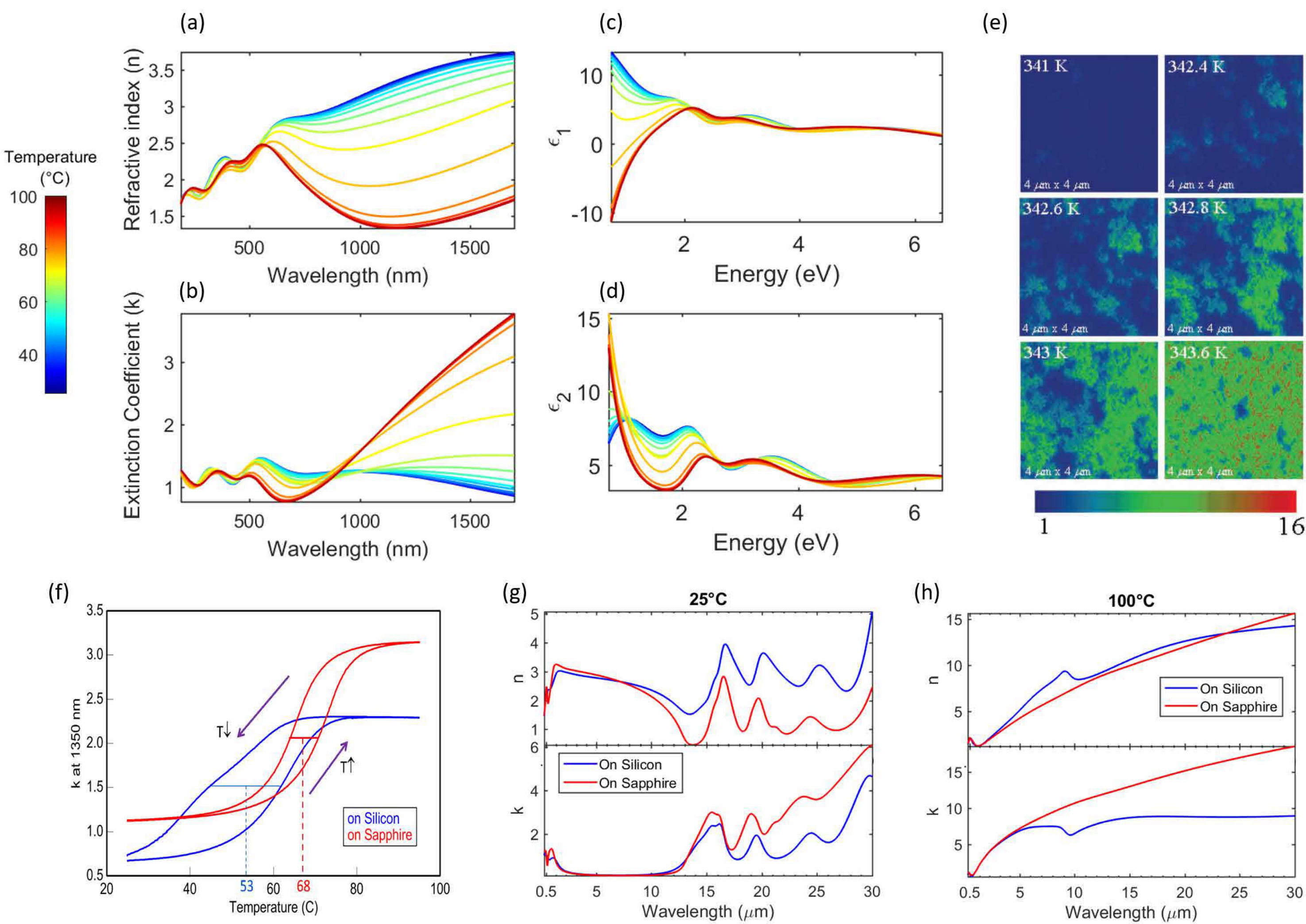
FIG. 7. Electrically-controlled optical modulation using VO₂. (a) Tunable transmission in the terahertz domain using an array of splitting resonators¹². (b)-(c) Array of electrically-connected hybrid metal/VO₂ antennas¹¹⁷. The electrical switching of VO₂ nano-elements enables tuning the optical absorption in the NIR. (d)-(e) A Metal-Insulator-VO₂-Metal stack comprising a connected array of antennas¹¹⁶. The electrical switching of VO₂ produces a large modulation of the transmission through the progressive disappearance of the plasmonic resonances. (f)-(h) A similar Metal-Insulator-VO₂-Metal stack arranged in a one-dimensional photonic crystal array¹¹⁸. The electrically-induced IMT switch is here producing a π phase-shift, hence enabling tunable phase modulation metasurface.

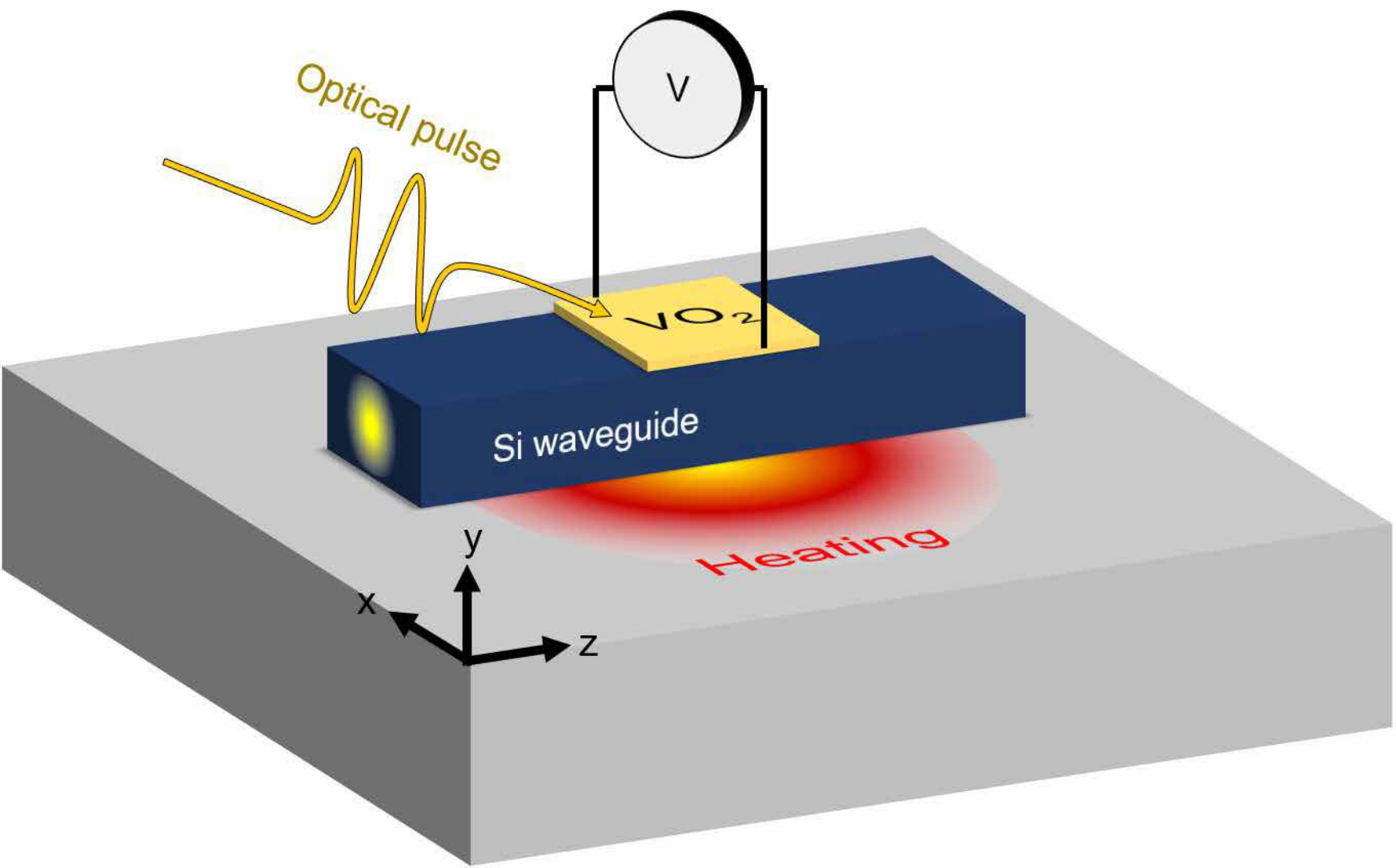
This is the author's peer reviewed, accepted manuscript. However, the online version of record will be different from this version once it has been copyedited and typeset.

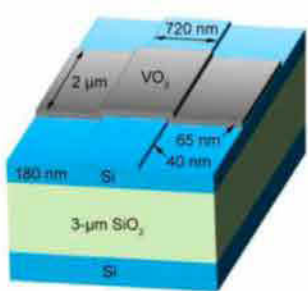
PLEASE CITE THIS ARTICLE AS DOI:10.1063/1.50028093

FIG. 8. Different methods of mitigating the optical absorption of VO₂ by reducing its size (a)-(b) ALD method enables growing ultra-thin and conformal layers of VO₂¹²² (c)-(d) VO₂-nanocrystals helps reducing the overall optical absorption and increase the figure of merit $\Delta n/\Delta k$.¹²³

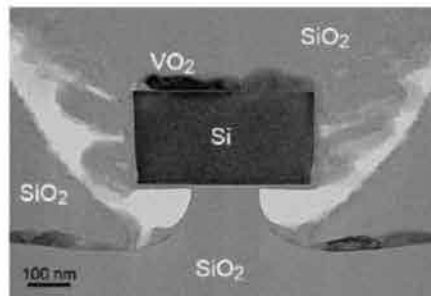
FIG. 9. Examples of spatially-addressed tunable regions and VO₂-based optically-imprinted optical elements with memory effects . (a) The laser imprinting of optical devices was demonstrated on VO₂-NCs platform and the large hysteresis behavior, typical in these VO₂-NCs enables to store such structures as non-volatile devices by maintaining the samples at temperatures close to RT ($\sim 30^\circ\text{C}$)¹²⁶. (b) The so-called 'programmable metacanvas' utilizing both the hysteresis and the local optical switching of VO₂ to fabricate and erase reconfigurable photonic devices¹²⁷. (c) A similar concept of locally-controlled VO₂ state using laser-scanning techniques to fabricate devices. In addition, the authors demonstrated the optical writing of multilevel states and controlled thermal emission¹²⁸.







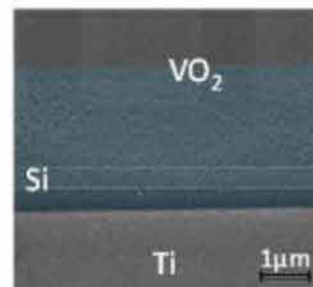
(a)



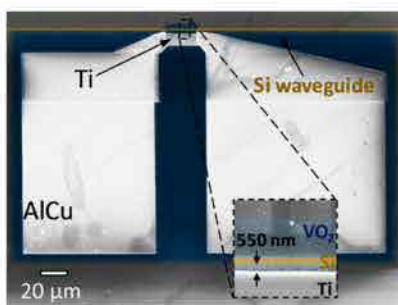
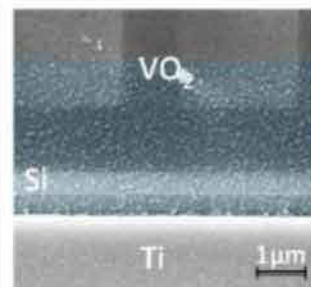
(b)



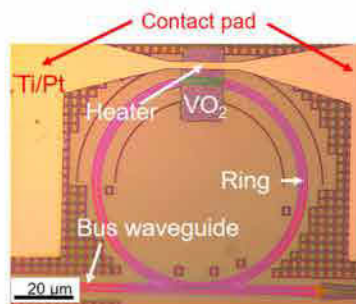
(c)



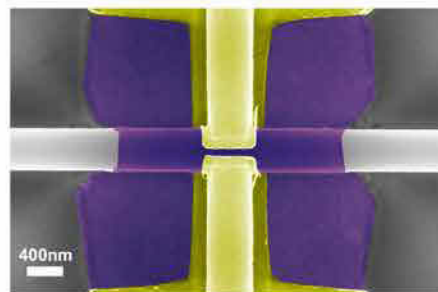
(d)



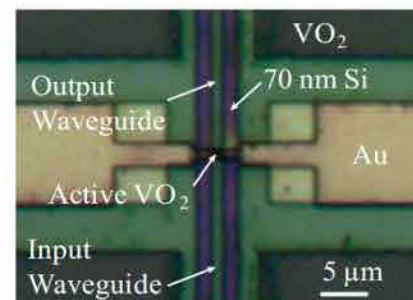
(e)



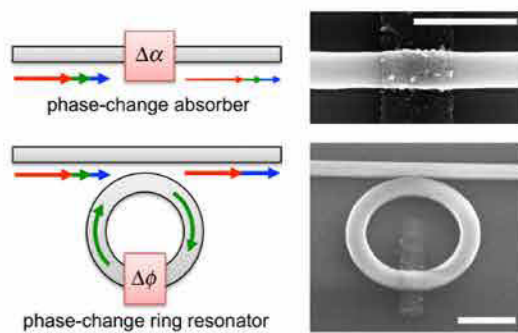
(f)



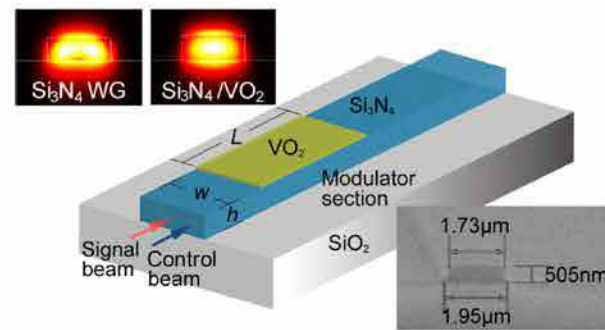
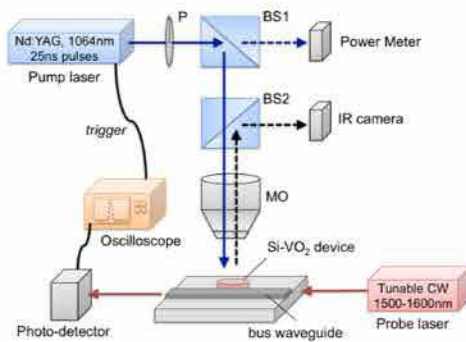
(g)



(h)



(i)



(j)

