#### **Research article**

Luca Bergamini\*, Bigeng Chen, Daniel Traviss, Yudong Wang, Cornelis H. de Groot, Jeffrey M. Gaskell, David W. Sheel, Nerea Zabala, Javier Aizpurua and Otto L. Muskens

# Single-nanoantenna driven nanoscale control of the VO<sub>2</sub> insulator to metal transition

https://doi.org/10.1515/nanoph-2021-0250 Received May 19, 2021; accepted July 20, 2021; published online August 9, 2021

Abstract: The ultrafast concentration of electromagnetic energy in nanoscale volumes is one of the key features of optical nanoantennas illuminated at their surface plasmon resonances. Here, we drive the insulator to metal phase transition in vanadium dioxide (VO<sub>2</sub>) using a laser-induced pumping effect obtained by positioning a single gold nanoantenna in proximity to a VO<sub>2</sub> thermochromic material. We explore how the geometry of the single nanoantenna affects the size and permittivity of the nanometer-scale VO<sub>2</sub> regions featuring phase transition under different pumping conditions. The results reveal that a higher  $VO_2$  phase transition effect is obtained for pumping of the longitudinal or transversal localized surface plasmon depending on the antenna length. This characterization is of paramount importance since the single nanoantennas are the building blocks of many plasmonic nanosystems. Finally, we demonstrate the picosecond dynamics of the VO<sub>2</sub> phase transition characterizing this system, useful for the realization of fast nano-switches. Our work shows that it is possible

to miniaturize the hybrid plasmonic-VO<sub>2</sub> system down to the single-antenna level, still maintaining a controllable behavior, fast picosecond dynamics, and the features characterizing its optical and thermal response.

Keywords: active metasurface; insulator-metal phase transition; photonic nanoswitch; picosecond dynamics; single plasmonic nanoantenna; VO<sub>2</sub>.

# 1 Introduction

Plasmonic single nanoantennas have intrigued vast interest due to their various exceptional properties [1-5]. Excited by an electromagnetic wave that falls within the antenna's plasmonic resonance band, the electrons around the surface of this nanostructure are driven collectively to form localized charge density oscillations, so-called surface plasmons, with the same frequency of the electromagnetic wave [6, 7]. During this process, the energy of the excitation wave is efficiently transferred into these localized surface plasmons (LSP), thus leading to a strong field enhancement at the surface of the nanoantenna, which is usually subwavelength sized [8-10]. Therefore, plasmonic nanoantennas allow light-matter interaction strengthening [3, 11, 12] and efficient photon manipulation at the nano-scale [1, 13-15], as well as boosting nonlinear response [2, 16–18].

The remarkable properties of nanoantennas have stimulated research combining local electromagnetic enhancement with phase change materials. Materials undergoing structural and/or electronic phase transitions as a response to an external stimulus [19-23], such as, for instance, a magnetic field, a light pulse, or direct heating, show a considerable change in their dielectric properties. This feature has been very successfully exploited to achieve large optical modulation contrast in nanophotonic switching devices [24-26]. Among those materials, vanadium dioxide  $(VO_2)$  is characterized by a reversible insulator-to-metal transition (IMT), taking place around a critical temperature of 68 °C [27] and making it a promising

<sup>\*</sup>Corresponding author: Luca Bergamini, Department of Electricity and Electronics, FCT-ZTF, UPV-EHU, 48080 Bilbao, Spain; and Materials Physics Center, CSIC-UPV/EHU and DIPC, 20018 San Sebastian Spain, E-mail: luca.bergamini@ehu.eus. https://orcid. org/0000-0001-7786-1499

Bigeng Chen, Physics and Astronomy, Faculty of Physical Sciences and Engineering, University of Southampton, Highfield, SO17 1BJ Southampton UK; and Research Center for Optical Fiber Sensing, Zhejiang Lab, Hangzhou 311100, China

Daniel Traviss, Yudong Wang, Cornelis H. de Groot and Otto L. Muskens, Physics and Astronomy, Faculty of Physical Sciences and Engineering, University of Southampton, Highfield, SO17 1BJ Southampton UK

Jeffrey M. Gaskell and David W. Sheel, Materials and Physics Research Centre, University of Salford, Manchester, M5 4WT, UK Nerea Zabala, Department of Electricity and Electronics, FCT-ZTF, UPV-EHU, Bilbao, 48080, Spain; and Materials Physics Center, CSIC-UPV/EHU and DIPC, San Sebastian 20018, Spain Javier Aizpurua, Materials Physics Center, CSIC-UPV/EHU and DIPC, San Sebastian 20018, Spain

<sup>🖯</sup> Open Access. © 2021 Luca Bergamini et al., published by De Gruyter. 🔯 By 🔤 This work is licensed under the Creative Commons Attribution 4.0 International License.

building block for high-performance optical devices [28–31], potentially with fast recovery times in the picosecond range [32, 33]. In a previous study, we showed that the combination of arrays of gold antennas on top of a  $VO_2$ substrate can lead to a significant optical switching effect on the  $VO_2$  permittivity, with the advantage of much lower switching energy requirement and faster recovery time as compared to a bare  $VO_2$  film without antennas [34].

Here, we demonstrate all-optical modulation on picosecond timescales of individual gold antennas fabricated onto a VO<sub>2</sub> substrate. The gold nanoantenna-VO<sub>2</sub> (AuNA-VO<sub>2</sub>) system is considered as a single entity with a hybrid response, where the nanoantenna drives the surrounding VO<sub>2</sub> phase transition through nanoscale electromagnetic confinement and the resulting changes in the VO<sub>2</sub> are amplified by the antenna's high sensitivity to changes in its local dielectric environment. Compared to more complex nanostructured antenna geometries loaded with small VO<sub>2</sub> patches [35], the antenna-on-film geometry is simple in design and operates on the selection of a nanoscale active volume through the resonant pumping arrangement. Through a combined numerical and experimental approach, we explore how different parameters, such as the antenna geometry and illumination conditions within a pump-probe scheme, modify the antenna-induced VO<sub>2</sub> phase change. We first characterize the effect on the  $VO_2$  phase change produced by a single gold nanoantenna when both the longitudinal and transversal LSP are selectively excited. Then, we show how this effect changes if the size of the nanoantenna changes, namely if the LSP resonances are tuned. We also study the radiation pattern of the antenna and the effect of using a detector with a finite numerical aperture (N.A.) to explain the features of the obtained optical spectra. Finally, we demonstrate the picosecond dynamics of the AuNA-VO<sub>2</sub> phase change, which makes this system wellsuited for the realization of optical switching technology at up to MHz speed. In this respect, the configuration of an isolated single antenna is of interest as it represents the smallest possible switching unit for applications. Hence, the determination of the key parameters underlying the antenna capability of inducing the phase change and producing a hybrid nonlinear optical response is desirable for designing optimized nanostructures, such as nanoscale nonlinear optical devices and switches.

# 2 Experimental setup

In our study, we consider individual gold nanoantennas fabricated on top of a  $VO_2$  film. As shown in Figure 1(A), the

first step of the fabrication process was the deposition of 50-nm-thick VO<sub>2</sub> films on boroaluminosilicate glasses coated with a 30 nm thin layer of fluorine-doped tin oxide (FTO), which allowed for the production of VO<sub>2</sub> films with low surface roughness and a suitable thermochromic transition temperature. The procedure followed for the realization of these high-quality films is detailed in reference [36]. In short, VO<sub>2</sub> was deposited using atmospheric pressure chemical vapor deposition at a temperature of 375 °C using vanadium (IV) chloride and water as the precursors for growth. High-quality films of <10 nm roughness were obtained using this method, with a thickness of around 50 nm achieved for a growth time of 90 s. Gold nanoantennas of 45 nm thickness, were fabricated on top of the VO<sub>2</sub> film by using e-beam lithography and liftoff. A Ti layer of 5 nm thickness was used to improve the adhesion of the gold to the  $VO_2$ .

Single-antenna spectroscopy was performed using a spatial modulation microscopy (SMM) technique [14, 15, 37-39]. A schematic of the SMM setup is displayed in Figure 1(B). A picosecond probe light filtered from a broadband supercontinuum light source (Fianium, 450-2500 nm) was focused on the nanoantennas via a reflective Cassegrain objective (Edmund Optics, 0.5 N.A.). Individual spectral components from 1.1 to 1.9 µm wavelength, with a bandwidth of 2% of the center wavelength, were selected from the supercontinuum source using a subtractive mode double prism monochromator. The polarization was fixed along the length direction of the nanoantennas. A piezoactuated flexure mirror driven at 200 Hz produced onedimensional periodic displacement of the focused light spot with respect to the nanoantennas, i.e., spatial modulation (SM) of the spot. Another objective (Mitutoyo 100×, 0.5 N.A.) was placed after the transparent sample to collect transmitted light for subsequent lock-in amplification detection. We also applied an optical chopper to modulate the probe beam for better signal recovery using the dualchannel demodulation option of the lock-in amplifier (Ametek Model 7270).

The SMM technique allows single antenna spectroscopy by means of a position-modulated probe beam and provides the normalized SM transmission of the single antenna  $(\Delta T/T)_{\text{SMM}}$ , defined as the difference in transmission between the surface with antenna and without antenna, normalized to the average transmission [37–39]. For an antenna that is absorbing or scattering light away from the forward beam, the transmission with the antenna is reduced and hence  $(\Delta T/T)_{\text{SMM}}$  is negative. In general, the inverse quantity  $(-\Delta T/T)_{\text{SMM}}$  is considered to be connected to the particle extinction cross-section minus the fraction of forward scattering captured by the detection system [39].



**Figure 1:** (A) Sketch of the single Au antenna-VO<sub>2</sub> system. (B) Schematic of the combined SMM and pump-probe experimental setup. (C) Sketch of the laser pump-probe illumination conditions. The black, blue, and red arrows represent the incident electric field of the probe, of the || polarized pump, and of the  $\perp$  polarized pump, respectively. The green arrow indicates the incident wavevector in the three cases.

This relationship becomes considerably more complicated for the geometry under study where the particle is positioned on a partially reflecting and absorbing substrate which itself varies under different optical pumping conditions. It is therefore difficult to separate the antenna cross-section from the VO<sub>2</sub> background and in this study, we, therefore, compare the value  $(-\Delta T/T)_{\text{SMM}}$  itself to the theoretical response.

In order to optically induce the IMT of the VO<sub>2</sub> films, we used optical pump pulses of 11 ps time duration which were focused onto the sample using the same optical system as the supercontinuum probe, as sketched in Figure 1(B). The pump was generated from the fiber oscillator operating at 1060 nm (Fianium) through a second amplifier stage parallel to the supercontinuum source. The polarization of this beam was selectively taken along either the length (|| polarization) or the width ( $\perp$  polarization) of the nanoantenna, as illustrated schematically in Figure 1(C). The initial temperature of the sample was kept at 45 °C and the repetition rate was set to 1 MHz in all studies apart from those explicitly investigating the temperature or repetition rate dependence of the effects.

The supercontinuum output was used as a probe to record the SM signal of the sample at a variable time delay from the pump pulse. Both pump and probe beams are reflected by the piezo flexure mirror placed directly behind the objective, and therefore the position of the pump focus is also modulated in the same way as the probe. As a result, the SM signal compares the pumped AuNA-VO<sub>2</sub> hybrid to the pumped bare VO<sub>2</sub> substrate and produces the change of

probe transmission  $(-\Delta T/T)_{\text{SMM}}$  at the same optical pump power. By comparing the antenna-VO<sub>2</sub> hybrid  $(-\Delta T/T)_{\text{SMM}}$ obtained under pumping conditions with the one obtained with pump off, we can monitor the antenna-mediated pump effect on the VO<sub>2</sub> IMT.

# 3 Modeling method

The strong pump-dependent optical response of the AuNA-VO<sub>2</sub> hybrid includes a combination of direct heating and antenna-mediated heating of the VO<sub>2</sub> by the pump laser. At the same time, the presence of the antenna with its LSP excitation, occurring in the concerned wavelength range, enhances the detection of changes in the VO<sub>2</sub>. We performed detailed numerical simulations of the thermoplasmonic response in order to interpret the experimental results and realize a qualitative comparison of the antenna-mediated effects on the VO<sub>2</sub> IMT.

To mimic the pump-probe experiments performed on the single  $AuNA-VO_2$  samples, three-step simulations were carried out using the commercial COMSOL Multiphysics software [40]. The system was modeled according to the geometry and conditions of the experiments described in the previous section.

The first and second simulation steps reproduce the pumping process, through the combination of an electrodynamical calculation followed by a heat-dynamical calculation. This combination allows determining how the complex  $VO_2$  permittivity spatially changes around the AuNAs (see Section 2.2 of Supplementary Information). In the first step of our multiphysics simulation, we calculated the overall direct and antenna-mediated absorption of the pump light by the VO<sub>2</sub>, considered here as an insulator layer. In the second step, the outcome of the optical modeling was used to obtain, through the thermal diffusion simulation, a temperature spatial map of the system at a time of 40 ps after the pump was switched off. In our simulations, we considered an initial sample base temperature of 45 °C. Results obtained after optical absorption and thermal diffusion simulations (steps 1 and 2 of the model) are shown in Figure 2(A) and (B) for a 312 nm long gold nanorod (matching the actual nanorod in the experiment), for polarizations respectively parallel and perpendicular to the antenna, as sketched in Figure 1(C). The temperature profile is then mapped onto a permittivity, following a temperature and wavelength-dependent model, as shown in Figure 2(C) and (D). In order to more accurately model the observed behavior, we have included in our mapping procedure a gradual temperaturedependent behavior of the permittivity. Here, the complex VO<sub>2</sub> permittivity changes according to a smoothed Heaviside temperature step function centered around 68 °C and varying between the values characterizing the insulator (below 58 °C) and metal (above 78 °C) behavior (see

also Section 2.1 of Supplementary Material for more details). Therefore, the VO<sub>2</sub> permittivity becomes a function of two variables, i.e., the incident probe light wavelength and the VO<sub>2</sub> temperature. This behavior is in line with detailed experimental studies of the temperature-dependent permittivity [5], but it constitutes a new approach going beyond the approximation made in previous studies [34] where the IMT was modeled as a discrete step in the optical response at the threshold temperature. Finally, the temperature profile is converted into a profile of the dielectric function by mapping the IMT transition onto this temperature profile, as displayed in Figure 2(E) and (F). As a side note, latent heat at the critical temperature [41] was also included in the model by increasing the heat capacity in a 2 °C temperature window around the critical temperature (see Section 2.2 of Supplementary Information).

The third step involves calculating the transmission of the system by incorporating the new spatial distribution of the VO<sub>2</sub> permittivity value. The three steps are done for the AuNA-VO<sub>2</sub> hybrid, as well as for a bare VO<sub>2</sub> substrate without antenna under identical pumping conditions. From the calculated transmissions with and without the nanoantenna under the same pump conditions, the theoretical normalized differential transmission  $(-\Delta T/T)_{\text{th}}$ values are obtained similar to the experimental case. Note



**Figure 2:** Colormaps of the antenna-mediated pump induced temperature ( $\theta$ ) for (A) || pump and (B)  $\perp$  pump. These colormaps refer to an antenna with dimensions 312 × 103 × 50 nm ( $L \times W \times H$ ) and a pump wavelength of 1060 nm. The 3D plot of the (C) real and (D) imaginary part of the VO<sub>2</sub> permittivity used in the simulations of the system, as a function of both wavelength and temperature. The green line indicates the VO<sub>2</sub> permittivity values for the 1450 nm cut wavelength used for the plots in (E) and (F). Colormaps of the antenna-mediated pump-induced real part of the VO<sub>2</sub> permittivity ( $\varepsilon_R$ ) for (E) || pump and (F)  $\perp$  pump. These colormaps refer to a probe wavelength of 1450 nm. Cuts along half either (A, E) the width or (B, F) the length of the antenna are performed to better display the hot spots due to || pump and  $\perp$  pump, respectively. All simulations are characterized by an initial sample temperature of about 45 °C.

that the resulting  $(-\Delta T/T)_{\text{th}}$  depends on the size of the simulated area and thus the quantitative matching with the experiment requires matching the area under consideration to the diffraction-limited spot size, which scales effectively with the square of the wavelength (see Section 2.3 of Supplementary Material for a detailed discussion). In our simulations, the areas are smaller, and instead of artificially scaling the transmission, we have chosen to directly plot the theoretical values from the modeling and perform a qualitative comparison.

In addition to the above model analysis, it was found that the phenomenology of the observed effects depends critically on the selected numerical aperture of the detection system. The reason for this is the effect of forward scattering, which adds an important contribution in the differential transmission toward longer wavelength, effectively reducing the  $(-\Delta T/T)_{\text{SMM}}$  values and even completely flipping its sign. These effects follow previous single-particle studies in the regime where extinction and forward scattering are of comparable magnitude [4] and will be discussed further below.

#### **4 Results**

#### 4.1 VO<sub>2</sub> phase transition produced by single-antenna-mediated optical pump

In our experiment, we first considered a single gold nanoantenna with a size of  $312 \times 103 \times 50$  nm ( $L \times W \times H$ ), for which the scanning electron microscopy (SEM) image is shown in the inset of Figure 3(A). The normalized SM transmission signal ( $-\Delta T/T$ )<sub>SMM</sub> of the antenna without a pump is presented in Figure 3(A) (black line) and shows a peak at around a wavelength of 1550 nm due to the LSP excitation in the antenna. The value of ( $-\Delta T/T$ )<sub>SMM</sub> of around  $10^{-2}$  signifies that the presence of the antenna results in a reduction of the transmission of 1% compared to the bare VO<sub>2</sub> film. For an isolated antenna and assuming negligible forward scattering, given the diffraction-limited spot size of around 7 µm<sup>2</sup>, the value of ( $-\Delta T/T$ )<sub>SMM</sub> translates to an extinction cross-section of 1.3 × 10<sup>-13</sup> m<sup>2</sup>.

The blue and red lines in Figure 3(A) show the  $(-\Delta T/T)_{\text{SMM}}$  response for the same antenna under picosecond optical pumping of the AuNA-VO<sub>2</sub> hybrid at a pulse energy of 0.6 nJ, for parallel and perpendicular orientation of the pump electric field, respectively. Optical pumping results in a blue shift and a reduction of the peak height of the plasmon mode for both polarizations. Additionally, the  $(-\Delta T/T)_{\text{SMM}}$  spectrum develops a negative overshoot, in

the presence of pumping, at longer probe wavelengths beyond the LSP position. The negative overshoot becomes more pronounced upon further increasing the pump power, as illustrated in Figure S9 of the Supplementary Material. The sign reversal in the  $(-\Delta T/T)_{\text{SMM}}$  signal indicates that rather than scattering light away from the transmission, the antenna increases forward transmission compared to the bare substrate under similar pumping conditions. In our numerical modeling, we found that this effect depends critically on the numerical aperture of the detection optics, as is explained in more detail in Section 4.3 further below.

To shed light on the precise pumping conditions in this nJ range of optical powers for both the VO<sub>2</sub> substrate and the AuNA-VO<sub>2</sub> hybrid, we performed a sweep of the optical pump power from 0.25 to 1 nJ, covering the range over which the IMT switching condition takes place. Figure 3(B) and (C) respectively show results for the overall transmission T, normalized to the transmission with pump off  $(T_0)$ , and for the SM signal  $(-\Delta T/T)_{SMM}$  corresponding to the AuNA-VO<sub>2</sub> hybrid, both at a fixed wavelength of 1450 nm. This wavelength is chosen as it most clearly shows the strongest pump-polarization-dependent effect for intermediate pump energy. A full set of  $(-\Delta T/T)_{\text{SMM}}$  data for three different wavelengths around the antenna resonance peak is presented in Supplementary Material Section 2.8, Figure S12. Given the small effect of the antennas of around 1%, the  $T/T_0$  response is governed by the bare VO<sub>2</sub> film without the AuNA. Over the full range of pump powers, we find that the illumination induces a reduction of around 10% of the global transmission T, of which 5% is reached at an energy of 0.6 nJ (indicated by the magenta vertical line). In comparison, Figure 3(C) shows that the  $(-\Delta T/T)_{SMM}$ signal for the AuNA-VO<sub>2</sub> hybrid at an energy of 0.6 nJ has a reduction of around 50% for the perpendicular pump polarization (red curve). Therefore, the main contribution to the  $(-\Delta T/T)_{\text{SMM}}$  spectra results from a change in  $\Delta T$ (i.e., antenna effect), rather than changes in T itself (i.e., pumping of the VO<sub>2</sub> substrate).

The antenna-mediated response in Figure 3(C) is articulated by the large difference between pumping parallel and perpendicular to the antenna length, as given by the blue and red curves in Figure 3(C), respectively. In fact, for the bare VO<sub>2</sub> substrate in Figure 3(B), changing the polarization of the pump laser does not have any effect and the two curves are overlapping. This polarization-dependent response for the pump is only seen for pump energies in the range 0.4–1 nJ. Indeed, the results for the AuNA-VO<sub>2</sub> hybrid of Figure 3(C) show that the pump energy range can be roughly divided into three parts, depending on the effect that the two pump polarizations produce on the optical



**Figure 3:** (A–C) Measured and (D–E) simulated response of AuNA-VO<sub>2</sub> system, for the single antenna with dimensions of  $312 \times 103 \times 50$  nm ( $L \times W \times H$ ). The inset of (A) shows the SEM image of this antenna. (A) Normalized spatial modulation transmission  $(-\Delta T/T)_{SMM}$  and (D) normalized differential transmission  $(-\Delta T/T)_{th}$  as a function of the probing wavelength for the 312 nm long antenna for conditions of no pump (black curve), parallel pump (||, blue curve) and perpendicular pump ( $\perp$ , red curve). The pump laser is characterized by a wavelength of 1060 nm and an energy of 0.6 nJ. The green line indicates the 1450 nm probing light wavelength considered in plots (B, C, E, and F). (B) Measured and (E) simulated  $T/T_0$  as a function of the pump energy at 1450 nm probe wavelength and for || pump (blue curve) and  $\perp$  pump (red curve). Blue and red curves perfectly overlap in (E), as the simulated response of a flat infinite stack does not depend on the polarization of the normally impinging light. (C) Measured  $(-\Delta T/T)_{SMM}$  and (F) simulated  $(-\Delta T/T)_{th}$  as a function of the pump energy of 0.6 nJ considered in (A, D). All measurements and simulations are performed at a repetition rate of 1 MHz and sample base temperature of 45 °C.

response of the system. For low pump energies (<0.4 nJ), the two pump polarizations originate a similar variation of the system optical response, which is close to the value without the pump. This is due to the almost negligible antenna effect produced by the relatively low pump energies. At intermediate pump energies between 0.4 and 1 nJ, different behavior of the  $(-\Delta T/T)_{\text{SMM}}$  response is found due to the different antenna-mediated effects for the two polarizations. Finally, the two polarizations produce again similar  $(-\Delta T/T)_{\text{SMM}}$  for energies as high as 1 nJ. For the higher energy regime, the pump energy is sufficient to

induce direct heating of the  $VO_2$  film while the resonant antenna-mediated effects are no longer uniquely driving the local IMT, therefore the response becomes again independent of pump polarization.

The results of the modeling of the L = 312 nm antenna under the same experimental pump and probe conditions are shown in Figure 3(D)–(F), obtained using the detailed calculations presented in Figure 2 for the same system. The calculated normalized differential transmission spectra under laser pumping at different polarizations are indicated by the blue and red curves in Figure 3(D). The simulations are qualitatively in agreement with the experiments, where the difference in vertical scale corresponds to a difference in the illumination area used for the calculations compared to the experiment. Under pumping conditions corresponding to 0.6 nJ pulse energy, we see a similar trend where the mode blue shifts and the peak amplitude decreases. The difference in LSP amplitude and profile between the two pump polarizations can be traced back to the formation of  $VO_2$  hot spots as seen in Figure 2(E) and (F), which are regions around the antenna where the VO<sub>2</sub> permittivity transitions from the dielectric to the metallic state, depending on the final temperature. The presence of these hot spots is inferred by both the temperature maps (Figure 2(A) and (B)), due to pump at a wavelength of 1060 nm, and the after-pump VO<sub>2</sub> permittivity maps, obtained at a wavelength of 1450 nm (Figure 2(E) and (F)). As revealed by the temperature maps, none of the two pump polarizations produces a temperature equal to or greater than 78 °C, which is the requirement for achieving a complete IMT transition, with a 0.6 nJ pump energy and an initial sample temperature of 45 °C. Indeed, the main effect is to decrease the real part of the permittivity: as a consequence, the pumped antenna feels a surrounding medium with a smaller permittivity with respect to the unperturbed antenna in absence of pumping, resulting in a blue shift of the plasmon peak.

The greater peak blue shift and decrease for the polarization perpendicular to the antenna ( $\perp$ , red curve) with respect to the parallel polarization (||, blue curve) can be explained in terms of the interplay between two effects: the value of the VO<sub>2</sub> permittivity assumed in the VO<sub>2</sub> hot spots and the size of these hot spots. The former depends on the VO<sub>2</sub> absorption, through either the longitudinal or transversal AuNA plasmon mode, i.e., the electronic excitation along the long or short axis of the antenna, respectively, at the pump wavelength of 1060 nm. This electronic excitation, in turn, depends on both the relative position of the plasmon peaks with respect to this pump wavelength and their absolute intensity and width.

As revealed by the simulations, the longitudinal mode peak is located at a wavelength of around 1550 nm, while the transversal mode peak occurs around 1000 nm (see Section 2.4 of Supplementary Material). The transversal mode is therefore located closer to the 1060 nm pump than the longitudinal mode. Even if the longitudinal plasmon peak is more intense and broader, the transversal plasmon is more efficiently excited. The overall effect is a similar intensity of the antenna excitation for the two polarizations at the pump wavelength of 1060 nm, which in turn leads to similar maximum temperature values (see Figure 2(A) and (B)) and minimum values of the VO<sub>2</sub> permittivity inside the  $VO_2$  hot spots (see Figure 2(E) and (F)). However, the shape of the antenna and the more efficient excitation of the antenna transversal mode produce a total volume of the  $VO_2$ hot spots larger in the case of perpendicular pump polarization than under parallel pump polarization. These two effects, namely a similar minimum value of the  $VO_2$  real permittivity and larger overall volume of the  $VO_2$  hot spots, lead to a greater blue shift and resonance damping for the perpendicular pump.

Simulations were also performed to retrieve the  $T/T_0$ and  $(-\Delta T/T)_{th}$  theoretical curves as a function of the pump energy, as displayed in Figure 3(E) and (F), respectively. A qualitative agreement of the simulations with the experiments of Figure 3(B) and (C) is found. The theoretical results confirm that the main contribution to the phase transition of the VO<sub>2</sub> in the hot spots is due to the presence of the pumped antenna and not to the direct effect of the pump on the VO<sub>2</sub> film. Moreover, the increase of temperature in specific sites of the VO<sub>2</sub> close to the nanoantenna can be attributed to significant absorption of the electromagnetic field in these sites and not to a conductive flux of heat from the pumped antenna itself. In fact, the conductive heat flux contribution is negligible, consistent with previous studies on antenna arrays [34] and confirmed by the obtained temperature maps (see Figure 2(A) and (B)), which do not reveal a uniform distribution of the temperature in the VO<sub>2</sub> around the antenna as expected under a conductive heat flux.

Finally, for a comprehensive description of the analyzed spectra, we note the flattening tail of both the  $(-\Delta T/T)_{\text{SMM}}$  and  $(-\Delta T/T)_{\text{th}}$  curves in the red part of the optical range, namely for wavelengths greater than 1800 nm. Further investigations reveal that these flattening tails are due to the onset of the intraband transitions in the FTO layer, which slightly affects also the plasmon peak position and intensity (see Section 2.5 of Supplementary Material for a detailed discussion).

#### 4.2 Effect of antenna size/resonance tuning on the VO<sub>2</sub> hot spots

The strong redshift of the longitudinal plasmon resonance of the L = 312 nm antenna positioned on the high-index VO<sub>2</sub> substrate means that the optical wavelength distance between the LSP and the pump wavelength at 1060 nm is significant. A situation where the antenna resonance is located closer to the pump wavelength is obtained for a shorter antenna. The realized antenna, shown in the SEM image in the inset of Figure 4(A), is characterized by a size of 222 × 110 × 50 nm ( $L \times W \times H$ ). This example allows



**Figure 4:** (A) Measured normalized spatial modulation transmission  $(-\Delta T/T)_{SMM}$  and (C) simulated normalized differential transmission  $(-\Delta T/T)_{th}$  as a function of probe wavelength for a 222 nm long antenna (antenna size  $L \times W \times H = 222 \times 110 \times 50$  nm). The behavior under (black curve) no pump is compared to the ones under (blue curve) || pump and (red curve)  $\perp$  pump. The pump laser is characterized by a wavelength of 1060 nm and an energy of 0.6 nJ. The green lines indicate the 1350 nm cut probing light wavelength used for plots (B, D). The inset of (A) shows the SEM image of the realized 222 nm long single antenna. (B) Measured and (D) simulated  $T/T_0$  as a function of the pump energy. The curves refer to a probe wavelength of 1350 nm and to both (blue curve) || pump and (red curve)  $\perp$  pump. The magenta lines indicate the cut pump energy of 0.6 nJ considered in plots (A, C). Blue and red curves perfectly overlap in (D) as the simulated response of a flat infinite stack does not depend on the polarization of the normally impinging light. Colormaps of the antenna-mediated pump induced temperature for (E) || pump and (F)  $\perp$  pump. These colormaps refer to a pump wavelength of 1060 nm. Cut along half either (E) the width or (F) the length of the antenna is performed to better display the hot spots due to || pump and  $\perp$  pump, respectively. All the presented measurements and simulations are performed with a repetition rate of 1 MHz and an initial sample temperature of about 45 °C.

studying the influence of the antenna geometry on the creation of VO<sub>2</sub> hot spots. The obtained experimental and simulated curves, showing the plasmon peak evolution under conditions of laser pumping at 0.6 nJ for both polarizations, are displayed in Figure 4(A) and (C), respectively. The curves exhibit qualitatively good agreement, especially in the red part of the spectrum. In the blue part of the spectrum the simulations reveal a crossing between the curves, with the  $(-\Delta T/T)_{\rm th}$  referred to the perpendicular pump higher than the one of the parallel pump, while the experimental curves approach each other but do not cross. These small differences may be attributed to variations in the modeled and experimental configurations, possibly related to local variations in the granular VO<sub>2</sub> film itself and in the antenna morphology, which are challenging to address precisely using modeling. We note that these shorter antenna dimensions are very close to the limits of our experimental capabilities both in nanofabrication and single-antenna spectroscopy.

Nevertheless, the overall qualitative agreement between experiments and simulations confirms the validity of the temperature- and wavelength-dependent  $VO_2$ permittivity used in our modeling. The simulations reveal that, for this shorter antenna, the pumping with polarization parallel to the antenna produces a greater suppression with respect to the perpendicular pump. In this antenna, the longitudinal mode is located at around 1350 nm wavelength, while the transversal mode is at around 1000 nm (see Section 2.4 of Supplementary Material), so that the optical distance to the pump wavelength of 1060 nm is still larger for the longitudinal mode than for the transversal mode. However, the longitudinal mode is more intense and broader than the transversal one, and this leads to a slightly higher excitation at the pump wavelength of 1060 nm for parallel pumping. At the same time, the maximum temperature value, and in turn the maximum VO<sub>2</sub> permittivity change, is almost the same for the two pump polarizations, as revealed by the temperature maps of Figure 4(E) and (F). What makes the peak decrease larger for the parallel pump polarization as compared to the perpendicular pump polarization is the overall volume of the created hot spots, which is bigger for the former than for the latter.

The  $T/T_0$  curves as a function of the pump laser energy are also reported in Figure 4(B) and (D). A  $T/T_0$  reduction of around 2.5% is found at the wavelength of 1350 nm for pump energy of 0.6 nJ, which is again much smaller than the approximately 50% reduction observed for the  $(-\Delta T/T)_{\text{SMM}}$  and  $(-\Delta T/T)_{\text{th}}$  curves under the same conditions. This result corroborates that the direct effect of the pump laser on the phase change of the VO<sub>2</sub> layer is not predominant with respect to the antenna-mediated one produced by the pumping of the LSP resonance.

The results reported in Figures 3 and 4 are part of a larger series of spectra taken at pump energies between 0 and 1 nJ. These results are summarized in Supplementary Material Section 2.6. Additionally, similar results for a number of single nanoantennas and dimer antennas are presented in Supplementary Material Section 2.10, showing that the observed responses are general for all AuNAs investigated.

# 4.3 Effect of the finite numerical aperture of the detector on the single-antenna response

In the attempt of improving the qualitative agreement between the experimental and calculated curves, we performed a further theoretical investigation of the effects of

the finite detection aperture on the optical response. We found that the finite numerical aperture of the detector strongly affects the recorded response of the single nanoantenna, especially at longer wavelengths far from the plasmon resonance. If we collect the light in the farfield inside a cone determined by the numerical aperture of the detector (0.5 N.A., which corresponds to a total angle of the cone of  $60^{\circ}$  in air and  $40^{\circ}$  in the SiO<sub>2</sub>), under conditions of no pumping we obtain the results shown by the black solid curves in Figure 5(A) and (B) for the 222 nm long and 312 nm long antenna, respectively. When compared to the results by a detector with a large aperture (black dashed curves in Figure 5(A) and (B)), it is clear that the finite numerical aperture of the detector results in an overall reduction of the apparent extinction as well as a change in the overall shape of the resonance profile. Reducing the N.A. affects mainly the shape in the red part of the spectra, leading to a decrease of the optical response  $(-\Delta T/T)_{\rm th}$ . For the 222 nm long antenna, the  $(-\Delta T/T)_{\rm th}$ curve becomes negative, similar to the trend observed in experiments (Figure 4(A)).

The change from positive values to negative values of  $(-\Delta T/T)$  can be explained by the wavelength-dependent radiation pattern of the antenna which is projected onto the



**Figure 5:** (A, B) Simulated normalized differential transmission  $(-\Delta T/T)_{th}$  as a function of probe wavelength for a (A) 222 nm long antenna and a (B) 312 nm long antenna (width and height equal to the antennas studied before). The response obtained with a detector characterized by a large N.A. (black dashed curve) and a 0.5 N.A. (black solid curve) is compared. In the latter case, the detector is placed 4 um away from the antenna, as shown by the green line of panels C and D. The behavior under a 0.6 nJ parallel pump is also reported for a detector with collecting 0.5 N.A. (blue curve). (C, D) The calculated difference between the *z*-component of the Poynting vector of the system with (*P<sub>z</sub>*) and without the antenna (*P<sub>z,sub</sub>*) at a wavelength of (C) 1300 nm and (D) 2000 nm, which correspond to the point 1 and 2 marked in panel A. The difference is normalized to the intensity of the probing laser (*I*<sub>0</sub>). The *XZ* plane refers to a plane parallel to the antenna length and passing through the middle of the antenna width. The maps are obtained under no pump, namely when the VO<sub>2</sub> film is completely dielectric. The green line and the white rectangle mark off the edge of the 0.5 N.A. detector and of the antenna, respectively. The displayed colorscale

The green line and the white rectangle mark off the edge of the 0.5 N.A. detector and of the antenna, respectively. The displayed colorscale holds for both maps.

limited aperture of the detector. In Figure 5(C) and (D), we plot the colormaps of the normalized difference between the z-component of the Poynting vector for the system  $(P_z)$  with and  $(P_{z,sub})$  without the antenna, namely  $-(P_z - P_{z,sub})/I_0$ , with  $I_0$  being the intensity of the probing laser. Since the integral over the detector surface (green line in Figure 5(C) and (D)) of  $P_z$  and  $P_{z,sub}$  gives the measured transmission of the system with and without the antenna, respectively,  $-(P_z - P_{z,sub})$  is directly linked to the  $(-\Delta T)_{th}$ . The maps refer to a 222 nm long antenna under no pump and are taken on a plane parallel to the antenna length and cutting through its center. The radiation pattern on resonance, i.e., for a wavelength of 1300 nm (Figure 5(C), corresponding to point 1 of Figure 5(A), is significantly different from the one of the antenna out of resonance. i.e., for a wavelength of 2000 nm (Figure 5(D), corresponding to point 2 of Figure 5(A)). In the former case, the reduction of light in the near-forward direction due to the strong antenna resonance leads to almost all positive  $-(P_z - P_{z,sub})$  contributions entering the detector area. In contrast, at the longer wavelength, more light is scattered in the near forward direction and the positive extinction lobes appear at larger angles (see also Section 2.9 of Supplementary Material). In this case, the antenna increases the overall near-forward transmission compared to the bare VO<sub>2</sub> film.

To make the study more comprehensive, we repeated the simulations with a 0.5 N.A. detector but under the application of a 0.6 nJ pumping. The results are given by the blue curves in Figure 5(A) and (B). The overall antenna-mediated effect on the VO<sub>2</sub> film leads to further damping of the resonant optical response, with respect to the unpumped system, in agreement with what is observed in the experiments of Figure 4. Additionally, we see for the 222 nm long antenna that the spectral range in which negative  $(-\Delta T/T)$  is obtained is further extended toward shorter wavelengths. The negative cross-over is not seen for the larger 312 nm antenna. Clearly, the exact balance of these subtle effects depends strongly on the ratio of scattering to absorption of the antenna, as well as the exact value of the  $VO_2$  background response  $P_{z.sub}$ . For example, a modest reduction of the pump power for the bare VO<sub>2</sub> substrate immediately results in improvements as  $P_{z,sub}$  is increased, and hence the  $(-\Delta T/T)$  response becomes more negative toward longer wavelengths. However, such further fine tuning of the precise simulation conditions, while potentially leading to the improved agreement, does not give us fundamental insights into the underlying physics. Therefore, we do not report any further optimizations in our study beyond the basic model simulation.

#### 4.4 Effect of repetition rate and initial sample temperature on the singleantenna response

To investigate the time evolution of the response of the AuNA-VO<sub>2</sub> hybrid to optical pumping, we use the pumpprobe technique to study the fast switching dynamics of the  $(-\Delta T/T)_{\text{SMM}}$  optical modulation in the system as a function of repetition rate and base temperature. Results are shown in Figure 6(A)–(F). The antenna under study was the 312 nm long single antenna studied in Section 4.1 and shown in Figure 3.

To explore the accumulated thermal effect in the AuNA-VO<sub>2</sub> hybrid induced by the pump pulse train, we increased the repetition rate of the pump and probe light from 1 to 5 MHz, while keeping the pulse energy fixed at 0.6 nJ. The measured temporal dynamics of  $(-\Delta T/T)_{\text{SMM}}$  are shown in Figure 6(A). It can be seen in the blue and red curves that single-cycle fast switching of the  $(-\Delta T/T)_{SMM}$ signal takes place on the time scale of picoseconds, while a slow accumulated thermal effect also exists [34]. These include the overall response of the AuNA-VO<sub>2</sub> hybrid to the optical pumping and the nanoscale volume of the induced thermal effect. The amplitude of the picosecond modulation effect is shown in Figure 6(E), where we plot the quantity  $\Delta(-\Delta T/T)_{\text{SMM}}$  defined as the difference of  $(-\Delta T/T)_{\text{SMM}}$  at +100 and -100 ps, respectively. At the higher repetition rates, the fast picosecond modulation effect was almost completely suppressed, as shown by the orange and cyan curves in Figure 6(A). In the case of a high repetition rate pulse train, the AuNA-VO<sub>2</sub> compound cannot sufficiently cool down before the next pulse arrives, which results in a stationary state of the system above the phase transition temperature. A different way of explaining this is that the average pump power increases with the number of pulses for fixed pulse energy, which is therefore heating up the sample locally more at higher repetition rates. As the system is not at any time relaxed back to its unperturbed state, picosecond excitation pulses in this case do not produce an ultrafast effect on top of the stationary switching state. Even at 5 MHz, however, there is still polarization dependence, indicating that the energy is coupled into the sample through the anisotropic antenna-VO<sub>2</sub> system. At low repetition rates, the signal also drops slightly, indicating that the background temperature of 45 °C is slightly below the ideal working point at low repetition; the stationary heating associated with a 1 MHz pulse train brings this up to result in a higher modulation amplitude.

The corresponding temporal dynamics of the global  $VO_2$  film transmission also exhibit a similar trend with a



**Figure 6:** Time-dependence of (A)  $(-\Delta T/T)_{SMM}$  and of (B) transmission  $T/T_0$  for repetition rates of 1 and 5 MHz. The measurements are performed for both || and  $\perp$  pumping conditions, at a probe wavelength of 1450 nm, and a pump energy of 0.6 nJ. The base sample temperature is set to 45 °C using a heater stage. Time-dependence of (C)  $(-\Delta T/T)_{SMM}$  and of (D) bare VO<sub>2</sub> transmission  $T/T_0$  for different initial base temperatures of the sample set by the heater stage. The measurements are performed for the  $\perp$  pump at a wavelength of 1450 nm, with a pump energy of 0.6 nJ and a repetition rate of 1 MHz. The negative and positive times in the plots refer to times when the pump is enabled and off, respectively. The time zero is taken as the time of switch-off of the pump. The legend in (A) and (C) also holds for (B) and (D), respectively. (E, F) Difference  $\Delta(-\Delta T/T)_{SMM}$  taken between values at -100 and +100 ps, for different repetition rates from 0.2 to 5 MHz for both pump polarizations (E) and for different base temperatures (F).

transition from ultrafast to stationary IMT between 1 and 5 MHz repetition rate, as shown in Figure 6(B), which indicates a similar thermal effect in the modulation of the bare VO<sub>2</sub> itself. It is clearly seen that the stationary state at the higher repetition rate has a much lower transmission of around 78% of the unperturbed state, indicating that the increase in repetition rate results in full switching of the VO<sub>2</sub> substrate at 0.6 nJ pulse energy, compared to only partial switching at 1 MHz. These results establish an upper limit to the switching speed of a few MHz for picosecond reversible modulation of the AuNA-VO<sub>2</sub> hybrid.

Finally, we investigated the switching dynamics for different base temperatures by varying the overall VO<sub>2</sub> film temperature from 21.7 to 60 °C. The results are shown in Figure 6(C) for perpendicularly polarized pumping at 1 MHz modulation rate and 0.6 nJ pulse energy, with the fast response signal  $\Delta(-\Delta T/T)_{SMM}$  shown in Figure 6(F). The fast modulation depth was gradually reduced at temperatures above 40 °C, which is caused by the limited cooling of the AuNA-VO<sub>2</sub> hybrid to the substrate at elevated base temperature. The corresponding bare substrate transmissions with increased base temperature are shown in Figure 6(D). At 60 °C we cannot observe the picosecond switching by the pump anymore, as full IMT switching is obtained at the stationary state induced by combined base temperature and optical pumping. Notably, full switching of the VO<sub>2</sub> substrate results in a strong reduction of the

transmission, much more than observed for the singleantenna studies shown in Figures 3 and 4.

# **5** Discussion

The experimental single-particle optical studies of optically pumped AuNAs on a  $VO_2$  film using the SMM technique allow us to separate antenna-mediated and bulk optical excitation pathways through the dependence of the antenna resonances on polarization and pulse energy of the pump. The interpretation of these effects required the detailed modeling of the thermo-optical behavior of the system. In addition, it was found that part of the phenomenology is attributed to the finite numerical aperture of the experimental system, which is sensitive to changes in the antenna radiation pattern as the film is switched.

The detection of antenna response using the SMM technique poses additional challenges in interpretation. In the SMM approach, the pumped antenna- $VO_2$  hybrid is referenced to the pumped  $VO_2$  substrate under identical conditions. The direct influence of the antenna on the local  $VO_2$  configuration means that it is not possible to separate the effects of the bare antenna and local  $VO_2$  contributions in the combined response. Therefore, it makes sense to consider the local AuNA- $VO_2$  hybrid system as a single

nano-object with respect to the homogeneous  $VO_2$  background. In this framework, local switching of the  $VO_2$ surrounding the AuNA produces a new object with a modified scattering response, which sensitively depends on the size and position of the switched domains. The complication in this framework is that the pumped  $VO_2$  film is not a constant either, and hence all measurements are referenced against a varying background.

Upon switching off the bulk VO<sub>2</sub> film, the VO<sub>2</sub> dielectric function, as shown in Figure 2(C) and (D), transitions from a high permittivity semiconductor to permittivity close to zero in the spectral range between 1100-1800 nm, resulting in a blueshift and suppression in resonant cross-section at high pump energies. Reduction of the film permittivity also changes the radiation pattern into the substrate, where the high-angle lobes are reduced and emission at lower angles is increased. The higher amount of forward scattering at long wavelengths results in a sign reversal of the SM response, as effectively the AuNA-VO<sub>2</sub> hybrid results in an increased near-forward transmission compared to the bare VO<sub>2</sub> film. While ideally these complications in interpretation could be avoided by using higher-N.A. detection optics, we emphasize that the numerical aperture in the infrared spectral range is strongly limited by the availability of suitable achromatic optics and the aperture used in these experiments is among the highest available.

Moreover, since the IMT transition implies a change from positive to negative of the real part of the complex permittivity, these systems are good candidates for obtaining (near-) zero permittivity materials actively controllable. The epsilon-near-zero materials are of interest for addressing phenomena like enhanced nonlinearities, emission tailoring, and light tunneling [42–44].

# 6 Conclusions

In conclusion, we studied systems composed of single gold nanoantennas on top of a  $VO_2$  film. We demonstrated that a pump laser can be used to induce a single-antennamediated permittivity change in the film, strongly dependent on the properties of the antenna. Apart from the geometrical nanoscale control and the fast switching (up to 1 MHz) of the  $VO_2$  hot spots, we demonstrated that the geometry of the antenna can be used as a means to control the distribution of the induced electromagnetic fields around the antenna itself, thus obtaining different effects on the  $VO_2$  for the used perpendicular and parallel pump polarizations. Detailed understanding of the experimental behavior required capturing numerically the local optical and thermal dynamics around the antennas and on the bare  $VO_2$  film, as well as taking into account effects of the finite numerical aperture of the detection system.

The presented experimental results and their theoretical interpretations reveal a smooth transition of the VO<sub>2</sub> permittivity from dielectric-like to metallic-like values around the reported critical temperature of 68 °C. This temperature dependence of the VO<sub>2</sub> permittivity establishes a novel insight into the evolution of the optical response of the hybrid system, and helps to clarify: 1) the predominant contribution on the VO<sub>2</sub> phase transition due to the pumped-single-antenna with respect to the one due to film direct pumping; 2) the nature of the transformation of the VO<sub>2</sub> optical response, which roots in the properties of the electromagnetic fields originated by the pumped antenna inside the VO<sub>2</sub> film, and not in the transfer of heat from the pumped antenna into it.

The combination of a smooth transition in temperature and a pump-power-dependent size of the single-antenna mediated switching volume enables a continuous tuning of the AuNA-VO<sub>2</sub> response over a wide range of optical pumping powers. Even though, from the experiments and simulation results, we can conclude that the NA-VO<sub>2</sub> hybrids in our study are not able to induce a complete IMT transition in the VO<sub>2</sub> layer, but the diverse mechanisms of modification of the AuNA-VO<sub>2</sub> response open up the possibility of using this kind of systems for finely tuning the permittivity of the medium surrounding the antenna both in time and in space. Overall, single nanoantennas are of interest for fast, nanoscale control of IMT materials, like VO<sub>2</sub>, and could potentially be used for the realization of ultracompact photonic nanoswitches that are actively controllable.

**Author contributions:** All the authors have accepted responsibility for the entire content of this submitted manuscript and approved submission.

**Research funding:** LB, NZ, and JA acknowledge financial support from Spanish MICIN (DOI 10.13039/501100004837) through Project Ref. No. PID2019-107432GB-I00, from the Department of Education of the Basque Government under project IT1164-19 and the Department of Industry of the Basque Government under Elkartek project KK-2018/0000. OM and BC acknowledge support from EPSRC (DOI 10.13039/501100000266) through grants EP/J016918/1 and EP/M009122/1.

**Conflict of interest statement:** The authors declare no conflicts of interest regarding this article.

**Data availability statement:** Data associated with this work are available from https://doi.org/10.5258/SOTON/D1917.

# References

- V. Giannini, A. I. Fernández-Domínguez, S. C. Heck, and S. A. Maier, "Plasmonic nanoantennas: fundamentals and their use in controlling the radiative properties of nanoemitters," *Chem. Rev.*, vol. 111, no. 6, pp. 3888–3912, 2011.
- P. Biagioni, J.-S. Huang, and B. Hecht, "Nanoantennas for visible and infrared radiation," *Rep. Prog. Phys.*, vol. 75, no. 2, p. 024402, Jan.2012.
- [3] P. Bharadwaj, B. Deutsch, and L. Novotny, "Optical antennas," Adv. Opt. Photonics, vol. 1, no. 3, pp. 438–483, Nov.2009, http://aop.osa.org/abstract.cfm?URI=aop-1-3-438.
- [4] K. Lindfors, T. Kalkbrenner, P. Stoller, and V. Sandoghdar, "Detection and spectroscopy of gold nanoparticles using supercontinuum white light confocal microscopy," *Phys. Rev. Lett.*, vol. 93, p. 037401, Jul.2004.
- [5] M. Currie, M. A. Mastro, and V. D. Wheeler, "Characterizing the tunable refractive index of vanadium dioxide," *Opt. Mater. Express*, vol. 7, no. 5, pp. 1697–1707, May.2017, http://www. osapublishing.org/ome/abstract.cfm?URI=ome-7-5-1697.
- [6] J. A. Schuller, E. S. Barnard, W. Cai, Y. Chul Jun, J. S. White, and M. L. Brongersma, "Plasmonics for extreme light concentration and manipulation," *Nat. Mater.*, vol. 9, no. 3, p. 193, 2010.
- M. Pelton, J. Aizpurua, and G. Bryant, "Metal-nanoparticle plasmonics," *Laser Photonics Rev.*, vol. 2, no. 3, pp. 136–159, 2008, https://onlinelibrary.wiley.com/doi/abs/10.1002/lpor. 200810003.
- [8] K. B. Crozier, A. Sundaramurthy, G. S. Kino, and C. F. Quate, "Optical antennas: resonators for local field enhancement," *J. Appl. Phys.*, vol. 94, no. 7, pp. 4632–4642, 2003.
- [9] P. Muehlschlegel, H.-J. Eisler, O. J. F. Martin, B. Hecht, and D. W. Pohl, "Resonant optical antennas," *Science*, vol. 308, no. 5728, pp. 1607–1609, 2005.
- [10] T. B. Hoang, G. M. Akselrod, C. Argyropoulos, J. Huang, D. R. Smith, and M. H. Mikkelsen, "Ultrafast spontaneous emission source using plasmonic nanoantennas," *Nat. Commun.*, vol. 6, p. 7788, 2015.
- [11] P. Zijlstra, P. M.R. Paulo, and M. Orrit, "Optical detection of single non-absorbing molecules using the surface plasmon resonance of a gold nanorod," *Nat. Nanotechnol.*, vol. 7, no. 6, p. 379, 2012.
- [12] F. Peyskens, A. Dhakal, P. Van Dorpe, N. Le Thomas, and R. Baets, "Surface enhanced Raman spectroscopy using a single mode nanophotonic-plasmonic platform," ACS Photonics, vol. 3, no. 1, pp. 102–108, 2015.
- [13] K. Li, M. I. Stockman, and D. J. Bergman, "Self-similar chain of metal nanospheres as an efficient nanolens," *Phys. Rev. Lett.*, vol. 91, p. 227402, Nov.2003, https://link.aps.org/doi/10.1103/ PhysRevLett.91.227402.
- [14] M. Abb, P. Albella, J. Aizpurua, and O. L. Muskens, "All-optical control of a single plasmonic nanoantenna–ito hybrid," *Nano Lett.*, vol. 11, no. 6, pp. 2457–2463, 2011.
- [15] Y. Wang, M. Abb, S. A. Boden, J. Aizpurua, C. H. De Groot, and O. L. Muskens, "Ultrafast nonlinear control of progressively loaded, single plasmonic nanoantennas fabricated using helium ion milling," *Nano Lett.*, vol. 13, no. 11, pp. 5647–5653, 2013.
- [16] T. Schumacher, K. Kratzer, D. Molnar, M. Hentschel, H. Giessen, and M. Lippitz, "Nanoantenna-enhanced ultrafast nonlinear

spectroscopy of a single gold nanoparticle," *Nat. Commun.*, vol. 2, p. 333, 2011.

- [17] M. Celebrano, X. Wu, M. Baselli, et al., "Mode matching in multiresonant plasmonic nanoantennas for enhanced second harmonic generation," *Nat. Nanotechnol.*, vol. 10, no. 5, p. 412, 2015.
- [18] G. Armelles, L. Bergamini, A. Cebollada, N. Zabala, and J. Aizpurua, "A novel vibrational spectroscopy using spintronicplasmonic antennas: magneto-refractive surface-enhanced infrared absorption," *J. Appl. Phys.*, vol. 129, no. 7, p. 073103, 2021.
- [19] K. Shportko, S. Kremers, M. Woda, D. Lencer, J. Robertson, and M. Wuttig, "Resonant bonding in crystalline phase-change materials," *Nat. Mater.*, vol. 7, no. 8, p. 653, 2008.
- [20] Z. Yang, C. Ko, and S. Ramanathan, "Oxide electronics utilizing ultrafast metal-insulator transitions," *Annu. Rev. Mater. Res.*, vol. 41, pp. 337–367, 2011.
- [21] G. Armelles, L. Bergamini, N. Zabala, et al., "Broad band infrared modulation using spintronic-plasmonic metasurfaces," *Nanophotonics*, vol. 8, no. 10, pp. 1847–1854, 2019.
- [22] G. Armelles, L. Bergamini, A. Cebollada, et al., "Magnetic modulation of far- and near-field IR properties in rod-slit complementary spintronic metasurfaces," *Opt. Express*, vol. 28, no. 22, pp. 32584–32600, Oct.2020, http://www.opticsexpress. org/abstract.cfm?URI=0e-28-22-32584.
- [23] N. Maccaferri, L. Bergamini, M. Pancaldi, et al., "Anisotropic nanoantenna-based magnetoplasmonic crystals for highly enhanced and tunable magneto-optical activity," *Nano Lett.*, vol. 16, no. 4, pp. 2533–2542, 2016.
- [24] K. F. MacDonald, Z. L. Sámson, M. I. Stockman, and N. I. Zheludev, "Ultrafast active plasmonics," *Nat. Photonics*, vol. 3, no. 1, pp. 55–58, 2009.
- [25] S. Abdollahramezani, O. Hemmatyar, H. Taghinejad, et al., "Tunable nanophotonics enabled by chalcogenide phasechange materials," *Nanophotonics*, vol. 9, no. 5, pp. 1189–1241, May.2020, https://www.degruyter.com/view/journals/ nanoph/9/5/article-p1189.xml.
- [26] M. Wuttig, H. Bhaskaran, and T. Taubner, "Phase-change materials for non-volatile photonic applications," *Nat. Photonics*, vol. 11, no. 8, pp. 465–476, 2017.
- [27] M. M. Qazilbash, M. Brehm, B. -G. Chae, et al., "Mott transition in VO<sub>2</sub> revealed by infrared spectroscopy and nano-imaging," *Science*, vol. 318, no. 5857, pp. 1750–1753, 2007.
- [28] D. Wegkamp and J. Stähler, "Ultrafast dynamics during the photoinduced phase transition in VO<sub>2</sub>," *Prog. Surf. Sci.*, vol. 90, no. 4, pp. 464–502, 2015.
- [29] D. Y. Lei, K. Appavoo, Y. Sonnefraud, R. F. Haglund Jr., and S. A. Maier, "Single-particle plasmon resonance spectroscopy of phase transition in vanadium dioxide," *Opt. Lett.*, vol. 35, no. 23, pp. 3988–3990, Dec.2010, http://ol.osa.org/abstract.cfm? URI=ol-35-23-3988.
- [30] D. 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*, vol. 2, no. 9, pp. 1306–1313, 2015.
- [31] Y. Abate, R. E. Marvel, J. I. Ziegler, et al., "Control of plasmonic nanoantennas by reversible metal-insulator transition," *Sci. Rep.*, vol. 5, no. 1, pp. 1–8, 2015.

- [32] J. K. Clark, Y.-L. Ho, H. Matsui, B. Vilquin, H. Tabata, and J.-J. Delaunay, "Photoinduced metal-like phase of VO<sub>2</sub> with subns recovery," ACS Photonics, vol. 7, no. 9, pp. 2395–2404, 2020.
- [33] K. A. Hallman, K. J. Miller, A. Baydin, S. M. Weiss, and R. F. Haglund, "Sub-picosecond response time of a hybrid VO<sub>2</sub>: silicon waveguide at 1550 nm," *Adv. Opt. Mater.*, vol. 9, no. 4, p. 2001721, 2021, https://onlinelibrary.wiley.com/doi/abs/10. 1002/adom.202001721.
- [34] O. L. Muskens, L. Bergamini, Y. Wang, et al., "Antenna-assisted picosecond control of nanoscale phase transition in vanadium dioxide," *Light Sci. Appl.*, vol. 5, no. 10, p. e16173, 2016.
- [35] Z. Zhu, P. G. Evans, R. F. Haglund, and J. G. Valentine, "Dynamically reconfigurable metadevice employing nanostructured phase-change materials," *Nano Lett.*, vol. 17, no. 8, pp. 4881–4885, 2017.
- [36] J. M. Gaskell, M. Afzaal, D. W. Sheel, et al., "Optimised atmospheric pressure cvd of monoclinic VO<sub>2</sub> thin films with picosecond phase transition," *Surf. Coating. Technol.*, vol. 287, pp. 160–165, 2016, http://www.sciencedirect.com/science/ article/pii/S0257897215305211.
- [37] A. Arbouet, D. Christofilos, N. Del Fatti, et al., "Direct measurement of the single-metal-cluster optical absorption," *Phys. Rev. Lett.*, vol. 93, no. 12, p. 127401, 2004.
- [38] O. L. Muskens, P. Billaud, M. Broyer, N. Del Fatti, and F. Vallée, "Optical extinction spectrum of a single metal nanoparticle: quantitative characterization of a particle and of its local

environment," *Phys. Rev. B*, vol. 78, no. 20, p. 205410, 2008.

- [39] A. Crut, P. Maioli, N. Del Fatti, and F. Vallée, "Optical absorption and scattering spectroscopies of single nano-objects," *Chem. Soc. Rev.*, vol. 43, no. 11, pp. 3921–3956, 2014.
- [40] COMSOL Inc., "COMSOL multiphysics," Available at: https:// www.comsol.com.
- [41] C. N. Berglund and H. J. Guggenheim, "Electronic properties of VO<sub>2</sub> near the semiconductor-metal transition," *Phys. Rev.*, vol. 185, pp. 1022–1033, Sep.1969, https://link.aps.org/doi/10. 1103/PhysRev.185.1022.
- [42] D. Yoo, F. Vidal-Codina, C. Cirac, et al., "Modeling and observation of mid-infrared nonlocality in effective epsilon-nearzero ultranarrow coaxial apertures," *Nat. Commun.*, vol. 10, no. 1, pp. 1–9, 2019.
- [43] T. Tumkur, Y. Barnakov, S. T. Kee, N. MA, and V. Liberman, "Permittivity evaluation of multilayered hyperbolic metamaterials: ellipsometry vs. reflectometry," *J. Appl. Phys.*, vol. 117, no. 10, p. 103104, 2015.
- [44] R. Liu, Q. Cheng, T. Hand, et al., "Experimental demonstration of electromagnetic tunneling through an epsilon-near-zero metamaterial at microwave frequencies," *Phys. Rev. Lett.*, vol. 100, no. 2, p. 023903, 2008.

**Supplementary Material:** The online version of this article offers supplementary material (https://doi.org/10.1515/nanoph-2021-0250).