Optical limiters are nonlinear devices that feature decreasing transmittance with increasing incident optical intensity, and thus can protect sensitive components from high-intensity illumination. The ideal optical limiter reflects rather than absorbs light in its active (“limiting”) state, minimizing risk of damage to the limiter itself. Previous efforts to realize reflective (rather than absorbing) limiters were based on embedding nonlinear layers into relatively thick multilayer photonic structures, resulting in substantial fabrication complexity, reduced speed and, in some instances, limited working bandwidth. In this paper, these tradeoffs are overcome by using the insulator-to-metal transition (IMT) in vanadium dioxide (VO₂) to achieve intensity-dependent modulation of resonant transmission through aperture antennas. Due to the large change of optical properties across the IMT, low-quality-factor resonators are sufficient to achieve high on–off ratios in the transmittance of the limiter. As a result, our ultrathin reflective limiter (thickness ≈1/100 of the free-space wavelength) is broadband in terms of operating wavelength (>2 µm at 10 µm) and angle of incidence (up to ≈50° away from the normal). Our analysis of the experimental results via opto-thermal simulations provides insight into limiter performance and is a useful guidance for further engineering efforts.

1. Introduction

The growing ubiquity of high-power light sources[1,2] and the increasing sensitivity of photodetectors and cameras[3,4] motivate designs of optical limiters that can protect delicate optical components and human eyes against high-intensity illumination. Optical limiters are designed to be transparent for low-intensity incident light (open state), but should have gradually decreasing transmittance for increasing intensity (limiting state) such that the output intensity remains roughly constant.[5–9]

Existing optical limiters are primarily based on nonlinear absorption,[9,10] with alternatives including nonlinear refraction,[11] nonlinear scattering,[12,13] or nonlinear reflection.[14] However, each of these mechanisms has drawbacks. The use of nonlinear refraction, scattering, or reflection requires additional care to redirect the refracted or scattered light away from any light-sensitive devices, so nonlinear absorption is typically preferred. A variety of nonlinear absorption mechanisms across many material systems have been used, including reverse saturable absorption in organic/organometallic polymers,[8,15] multi-photon absorption in liquid crystals[16] and graphene and its derivatives,[17] free-carrier absorption in semiconductors,[18] nonlinear absorption in correlated materials,[19] and many others.[9,10,20,21] Despite their popularity, absorptive limiters are often not ideal because absorption of intense incident light can damage the limiter itself, for example, via melting, sputtering, delamination, or ablation.

The ideal solution is thus a limiter that transitions from a high-transmittance state at low intensity to a reflecting state at high intensity. Such devices have recently been proposed and demonstrated using multilayer photonic structures with the nonlinear medium comprising gallium arsenide (GaAs)[22] or germanium–antimony–tellurium (GST).[23] In the first demonstration based on GaAs,[22] the spectral and angular bandwidths were small due to the large quality factor of the photonic-crystal resonator, which was needed to enhance the optical nonlinearity. In the recent demonstration based on GST,[23] a complex multilayer structure with multiple embedded GST layers was used to achieve broadband performance, enabled by the large effective nonlinearity due to the phase change of GST. However, the use of multiple active layers within a thick structure can limit the switching speed, and the nonvolatile phase change in GST means that a limiter based
Figure 1. a) Schematic of an optical limiter based on a gold frequency-selective surface (FSS) on top of a thin film of VO$_2$ with GaAs (001) as the transparent substrate. b) Simulated open- and limiting-state transmittance and absorptance of the structure in “a)" with $d_1 = 0.2 \mu$m, $d_2 = 3.1 \mu$m, and $D = 3.5 \mu$m. The thickness of gold and VO$_2$ are 50 nm and 100 nm, respectively. The central wavelength of the open-state transmittance peak is $\lambda = 10.6 \mu$m, with peak transmittance of 0.7. The limiting-state transmittance is smaller than 0.01, accompanied by absorptance of $\approx 0.06$ at all wavelengths. c) Simulated angle-dependent open-state transmittance of the design in “b" for both s- and p-polarizations at $\lambda = 10.6 \mu$m. d) The central wavelength of the open-state resonant transmittance is tunable between 4 and 11 $\mu$m by changing $d_1$ from 1 to 3 $\mu$m. The value of $d_2$ was fixed at 0.2 $\mu$m. The value of $D$ is always set to $(d_1 + 0.4) \mu$m.

2. Design and Simulation

Our design comprises a gold FSS, a thin film of VO$_2$, and a transparent substrate (GaAs), as shown in Figure 1a. The IMT in VO$_2$ can be thermally triggered by heating to $\approx 70^\circ$C,[27,28] and can be a source of photothermal nonlinearity.[26,29] The very large change in optical properties across the IMT is a source of photothermal nonlinearity[26,29] that is orders of magnitude stronger than conventional nonlinearities (e.g., the Kerr effect[25,31]). Indeed, the limiting characteristics of VO$_2$ films based on this material can only switch to the limiting state a single time before it must be manually reset.

In this paper, we demonstrate an ultrathin, volatile, reflective optical limiter that only comprises two subwavelength-thickness functional layers and operates over a broad range of wavelengths and incidence angles. This is achieved using resonant transmission through a metallic frequency-selective surface (FSS) comprising low-quality-factor aperture antennas, made optically responsive using the insulator-to-metal transition (IMT) of vanadium dioxide (VO$_2$), which can be triggered optically via a photothermal process.[24,25] The very large change in optical properties across the IMT enables the strong modulation of resonant transmission through the aperture antennas, resulting in a limiter with broadband transmission in the open state and low transmission and absorption in the limiting state.

VO$_2$ is not nearly as reflective as noble metals (e.g., gold and silver)[26,34] and therefore a nontrivial amount of incident power can still be absorbed by the metal-phase VO$_2$. Our use of an FSS on top of a VO$_2$ film significantly reduces the limiting-state absorption, enabling the use of the IMT in VO$_2$ for reflective limiting. The gold FSS in our design is an array of close-packed cross-slit aperture antennas within a thin film of gold (Figure 1a) that has high resonant transmission when the near VO$_2$ film is in the insulating phase. Once the IMT is triggered by sufficiently intense incident light (e.g., intensity of $\approx 1$–100 kW cm$^{-2}$, as shown in the next section), the resonant transmission is expected to decrease quickly via two mechanisms. First, the resonance is frequency shifted by the large change in the real part of the refractive index of VO$_2$ ($\Delta n > 100$% in the mid infrared[26]). Second, the increasing loss in the VO$_2$ further suppresses the amplitude of the resonant transmission. These two mechanisms result in a large reduction of the device transmittance as the VO$_2$ undergoes the IMT. For the substrate, we chose undoped GaAs (001) since it is mostly transparent across the mid infrared.[35] We used finite-difference time-domain (FDTD) simulations (implemented in Lumerical FDTD) for design and optimization.

The structure was optimized for maximum transmittance at a target wavelength when in the open state, and maximum reflectance when in the limiting state. Specifically, we
considered three figures of merit: a) high open-state transmittance that ensures high transmission efficiency of low-intensity light; b) low limiting-state transmittance that ensures effective limiting of high-intensity light; and c) low limiting-state absorbance that enables a high damage threshold. For this paper, we chose $\lambda = 10.6 \mu m$ as the central wavelength of the open-state resonant transmittance.

Using the temperature-dependent refractive indices of VO$_2$ that were characterized in our previous work (ref. [26]), we optimized the structural parameters of the aperture antennas, including length ($d_1$) and width ($d_2$) of the aperture antenna and the periodicity ($D$). Note that there are tradeoffs between our three figures of merit. For example, a larger $d_1$ or a smaller $D$ helps to improve the open-state transmittance but also increases the limiting-state absorbance (see detailed discussion in Supporting Information S1).

After considering these tradeoffs during optimization, we settled on $d_1 = 3.1 \mu m$, $d_2 = 0.2 \mu m$, and $D = 3.5 \mu m$. The thicknesses of gold or VO$_2$ have much less influence on our figures of merit because they are much smaller than the wavelength (thickness of $\approx 100$ nm $\ll$ operational wavelength of 10.6 $\mu m$). Therefore, we chose the thicknesses of gold and VO$_2$ to be 50 and 100 nm, respectively. The resulting structure features a broad transmittance band (FWHM $> 2 \mu m$) centered at $\lambda = 10.6 \mu m$ in the open-state and close-to-zero transmittance ($\approx 0.008$) in the limiting state (Figure 1b). The limiting-state absorbance ($\approx 0.06$ at all wavelengths) is significantly reduced compared to that of the bare VO$_2$ film with no FSS ($\approx 0.2$, Supporting Information S2, and more details in Analysis and Discussion), thus enhancing the damage threshold of the device. The limiting-state absorbance can be further decreased by reducing the area density of the aperture antennas; however, this comes at the cost of the amplitude transmittance decrease for $\lambda = 10.6 \mu m$ of the metal-phase VO$_2$ are possibly due to a very thin surface oxide of V$_2$O$_5$ on the as-grown film [26,36] (more details in Supplementary Information S3). We note that, as shown later, such thin surface oxide does not substantially affect the performance of our fabricated limiter. The absorbance spectrum at 100 $^\circ C$ (absorbance $= 1 -$ transmittance $-$ reflectance) indicates that the metal-phase VO$_2$ has a relatively high absorbance of $\approx 0.3$ at all wavelengths.

Then, we built a 50-nm gold FSS on top of the as-grown VO$_2$ via steps as shown in Figure 2a (see details in Experimental Section). At 30 $^\circ C$ (device in the open state), the fabricated FSS-VO$_2$ limiter features a peak transmittance of $0.45$ at $\lambda = 9.8 \mu m$, while the transmittance decreased to $\approx 0.03$ when the temperature was increased to 100 $^\circ C$ (device in the limiting state) (Figure 2d). The measured reflectance in the limiting state is $>0.9$ at all wavelengths (Figure 2f), indicating that the limiting-state absorbance is $<0.06$ (Figure 2d), which is substantially reduced compared to that of the bare VO$_2$ film (Figure 2c). To demonstrate that our design is feasible for wavelengths across the mid infrared, we also fabricated samples with different aperture lengths. The measurements (Figure 2g) showed that our limiters with different aperture lengths ($d_1 = 1, 1.5, 2, 2.5,$ and $3 \mu m$) have central wavelengths of resonant transmittance ranging from 4 to 11 $\mu m$, as expected based on the simulations in Figure 1d. We attribute the transmittance decrease for $d_1 = 3 \mu m$ to the increasing optical loss in VO$_2$ at $\approx 11 \mu m$ [26].

The measured spectra of the fabricated FSS-VO$_2$ limiter (Figure 2d) essentially agree with our simulations (Figure 1b), though there are discrepancies in the amplitudes and peak positions of the open-state transmittance. The amplitude of experimental open-state transmittance is lower than that of the simulation, likely because we did not consider the backside of the substrate in the FDTD simulation. After taking it into account, the simulated peak value matches well with that of the experimental result (Supporting Information S4). In practice, one can manufacture anti-reflection coatings on the backside of the GaAs substrate to achieve the performance similar to simulations. Also, the open-state central wavelength of the fabricated limiter is blue-shifted by $0.8 \mu m$ compared to our simulation results primarily due to two experimental errors. First, the aperture length and width in the fabricated FSS are $3 \mu m$ and $0.17 \mu m$, respectively (Supporting Information S4), both smaller than those we used in simulations. Second, the refractive indices of VO$_2$ that we used for simulation were not extracted from the film that we used for fabrication. Small differences in refractive-index values are expected between VO$_2$ films, though they share similar tendencies in this wavelength region [26].

Despite the shift of the transmittance peak, our fabricated limiter still features reasonable open-state transmittance of $0.36$ at $\lambda = 10.6 \mu m$, enabling us to use a continuous-wave CO$_2$ laser to test our design. In our intensity-dependent transmission measurement setup (Figure 3a), the sample was mounted on a heat stage with a through-hole aperture used to thermally bias the device, which was necessary since the maximum incident
Figure 2. a) Fabrication flow: we first synthesized \( \approx 100 \text{-nm VO}_2 \) on GaAs wafer using magnetron sputtering. The as-grown film went through an e-beam lithography and development process, resulting in an array of cross-shape blocks of PMMA. Then, we evaporated 50 nm of gold on top, and performed a lift-off process, leaving an array of cross-slit apertures within the gold film. b) A schematic of simplified optical path of our transmission and reflection measurements, using an infrared microscope (Bruker Hyperion 2000) attached to a Fourier-transform spectrometer (Bruker Vertex 70). c) Transmittance and absorbance spectra of the as-grown VO\(_2\) film, for both insulating (30 °C) and metallic (100 °C) phases. d) Transmittance and absorbance spectra of the FSS-VO\(_2\) limiter at 30 °C (open state) and 100 °C (limiting state). e,f) Transmittance and reflectance measurements of our fabricated FSS-VO\(_2\) limiter when VO\(_2\) in its pure insulating phase (30 °C), pure metallic phase (100 °C), and intermediate phases across the IMT (at 72, 74, 76, and 78 °C). g) Measured transmittance of the FSS-VO\(_2\) limiters with aperture lengths \( d \) of 1, 1.5, 2, 2.5, and 3 \( \mu \)m, for both open (30 °C) and limiting (100 °C) states. The top surfaces of the inset figures in (c–g) are SEM images of the corresponding fabricated samples.

We measured the transmission of both the bare VO\(_2\) film and fabricated FSS-VO\(_2\) limiter for a series of incident powers and at different temperatures (i.e., temperature of the stage). For the bare VO\(_2\) film (Figure 3b), we did not observe nonlinear transmission (i.e., limiting behavior) when the temperature was set at or below 70 °C, which is at the edge of the IMT for increasing temperature (Figure S3c, Supporting Information). Limiting behavior became apparent when the film was thermally biased to 74 °C, which is already an intermediate temperature within the IMT. The throughput power was limited to \( \approx 55 \text{ mW} \) at high-power incidence (i.e., incident power greater than 120 mW). In contrast, the FSS-VO\(_2\) limiter featured obvious nonlinear transmission at \( \approx 70 \text{ °C} \) for incident power starting at 30 mW (the pink curve in Figure 3c). The transmitted power was limited to \( \approx 25 \text{ mW} \) for incident power from 90 to 190 mW. Furthermore, at a lower biasing temperature of 62 °C, which is far outside the IMT temperature range, we also observed limiting behavior when the incident
power was increased above 150 mW. Such limiting behavior was not found in the bare VO₂ film using the same experimental conditions.

4. Analysis and Discussion

Our intensity-dependent transmission measurements (Figure 3b,c) can be qualitatively understood using our previous FDTD simulation (Figure 1) and temperature-dependent measurements using FTS (Figure 2c-f and Figure 4a,b). As expected from the initial simulations (Figure 1b), the introduction of the FSS resulted in much lower limiting-state absorption and higher transmittance contrast between the two states compared to the bare VO₂ film. Somewhat counterintuitively, the introduction of the FSS also resulted in limiting action starting at lower incident powers or lower bias temperature (i.e., a reduction of the limiting threshold). As shown in Figure 3d, when biased at 70 °C, the bare VO₂ film featured constant (i.e., linear) transmittance at all input powers, while the FSS-VO₂ limiter featured a decreasing (i.e., nonlinear) transmittance as the incident power increases, indicating that the limiter was photothermally heated to intermediate temperatures in the IMT region at the beam spot. For each transmittance, we can approximate the corresponding local temperature within the VO₂ film at the beam spot using our temperature-dependent transmittance measurements shown in Figure 2e.

The limiting threshold and required thermal bias depend strongly on the open-state absorptance of the device. In the case of bare thin-film VO₂, the open-state absorptance is very low (≈0.02, Figure 4a), so a high incident power is required to trigger the IMT. Alternatively, the film can be thermally biased part of the way into the IMT (e.g., at 74 °C, the dark red curve in Figure 3b), where the absorptance has begun to rise even in the absence of incident light. Due to the absorptance of the nanostructured gold film, the introduction of the FSS results in much-higher open-state absorptance of ≈0.12 in our device (Figure 4a), enabling limiting action that begins far outside the IMT (e.g., at 62 °C, the purple curve in Figure 3c). Furthermore, in the FSS-VO₂ limiter, both the higher open-state absorptance and the increase of absorptance at the onset of the IMT (Figure 4a) result in faster turn-on compared to the bare VO₂ film. As the IMT evolves toward the pure metallic phase, the subsequent decrease of the absorptance starts to slow down the IMT, resulting in limiting performance for a broad intensity range (Figure 3c).

To quantitatively understand our intensity-dependent measurements, we built a photothermal simulation using COMSOL Multiphysics. Our model consists of an axisymmetric geometry of five domains centered on the center axis of the laser beam (Figure 4c). The heat generation within the device is due to laser power absorbed in the gold FSS and VO₂ layers (domains 1 and 3 in Figure 4c). The laser beam used in our model had a uniform intensity distribution within a radius of 50 µm. We used a continuous gold film to represent the gold FSS in our thermal modeling, which is expected to be a valid assumption because the area density of gold in the FSS is high (≈0.88) and the apertures are small.

We assumed the thermal conductivity and specific heat capacity of VO₂ to be 6 W (m·K)⁻¹ and 690 J (K·kg)⁻¹, respectively. Note that these values correspond to VO₂ in its insulating phase, but their change due to the IMT is not dramatic (less than 20% across the IMT), so for simplicity we set them to be constant. Thermal properties of gold and GaAs were taken from refs. [40, 41].

Our model works as follows to calculate the transmittance given a particular incident intensity for a given bias temperature. With no laser irradiation, the device is in thermal equilibrium at the given bias temperature. The transient thermal simulation is initiated by heat flux induced into the device by laser irradiation. For each increase in time, the simulation returns a transient temperature distribution within the device, which is used to update the absorptance based on the measured temperature-dependent absorption in Figure 4a. The resulting absorptance is then fed into the simulation at the next time step. This coupled
opto-thermal simulation loop iterates until the temperature distribution stabilizes. Finally, we can convert the stabilized temperature distribution to transmittance according to Figure 4b. Based on convergence tests, we chose to use a step of 0.1 µs and a total simulation time of 500 µs (see details in Supporting Information S6).

Both the limiting threshold and the trend of reduction of transmittance with incident intensity are well captured in our simulations (Figure 4d), and agree very well with experimental data extracted from Figure 3c (see Supporting Information S4 for more details). Our simulations also confirmed that our experiments never fully reached the complete metal phase of the VO₂ due to the limited incident intensities (<6.5 kW cm⁻²). One can expect that once the IMT is completed, the output power will start to increase again with a very slow rate of ≈0.03 (i.e., the limiting-state transmittance) as the input power increases.

Our opto-thermal modeling can be used to estimate the limiting threshold for a passive device (i.e., without thermal biasing) and the response time of the device (i.e., the time needed to reduce transmittance by e⁻¹), which are important parameters for practical use. In our experiments, we did not demonstrate nonlinear transmittance without thermal bias due to the limited output power of our laser. Our simulations predicted that, at room temperature (25 °C), the limiting state will be triggered when the incident intensity reaches ≈40 kW cm⁻² (blue curve in Figure 4d). The response time was obtained by converting the temperature distribution at each time step to the transmittance. For the limiter working at 25 °C, the time evolution of the transmittance for different incident intensities is shown in Figure 4e. For example, given an incident intensity of 60 kW cm⁻², the response time is ≈30 µs. The response time reduces to <4 µs when the incident intensity is greater than 150 kW cm⁻². Note that the recovery time (from the limiting state to the open state, after the light turns off) maybe much longer than the limiting response time, but the recovery time is not particularly important for limiting applications. Similarly, for a given incident intensity (e.g., 90 kW cm⁻²) one can expect to have a faster response at a higher bias temperature (Figure 4f). However, passive devices are more practical. In the absence of thermal bias, the limiting threshold can be engineered via a reduction of the IMT temperature using doping[42] or defect engineering.[43] On the other hand, to increase the limiting threshold, one can reduce the open-state absorption by redesigning the FSS.[44]

Compared to conventional optical limiters that are based on nonlinear absorption such as reverse saturable absorption or multi-photon absorption, our reflective limiter has two advantages: low triggering threshold and high damage threshold. Due to the ultrahigh photothermally induced nonlinearity of VO₂, our limiter can feature limiting behavior for incident intensities of ≈1–100 kW cm⁻² (as shown in Figures 3 and 4), while nonlinear-absorption materials usually need the intensity to be higher than 10⁷ kW cm⁻² to trigger the nonlinearity.[8,10] We note that Howes et al.[45] recently demonstrated an optical limiter by incorporating VO₂ with dielectric metasurfaces, which also has a similar low
triggering threshold as that of our design. Moreover, our limiter is designed to be highly reflective in the limiting state to avoid the damage (such as melting or delamination) that can be caused by the absorption of the intense light. On the other hand, conventional nonlinear-absorption limiters have their own advantages such as low fabrication cost and broad bandwidth since they usually do not require optical-resonance structures.

For practical applications, it is also important to consider the effect of the hysteresis in VO₂. For many applications that may be enabled by VO₂, like modulators or switches, it is necessary that the device can be turned on and off quickly. On the other hand, in an optical limiter, the return to the open state when no longer illuminated by high-power light is ideal but not absolutely necessary. Nevertheless, hysteresis can be overcome in our optical limiters if the device is biased outside of the hysteresis loop (e.g., temperatures smaller than 52 °C according to Figure S3c in Supporting Information). Our simulations predict that, when biased at 52 °C, the nonlinear transmittance can be achieved when the incident intensity surpasses 15 kW cm⁻² (the cyan curve in Figure 4d). Alternatively, one can also consider exploring phase transitions with minimal hysteresis (e.g., the IMT in samarium nickelate).

Finally, we note that in our experimental demonstration, the IMT in VO₂ was photothermally triggered by a continuous-wave laser. Therefore, the device speed is limited to approximately microseconds. However, the IMT in VO₂ can actually be triggered nonthermally on time scales as short as tens of femtoseconds using high-intensity optical pulses, and therefore we anticipate that VO₂-based limiters can also limit very-high-intensity pulses at these time scales. The optimization of VO₂-based limiters for ultrafast operation may require a redesign, optimizing for higher electric-field concentration within the VO₂ layer to more-easily trigger the IMT.

5. Conclusion

Reflective optical limiters, which are transmissive at low input powers but reflecting at high powers, are a promising technology for sensor protection, because they can avoid damage due to light absorption in the limiter itself. Here, we explored the use of resonant transmission through metallic frequency-selective surfaces (FSSs) as the basis for reflective limiters, using the IMT in vanadium dioxide (VO₂) to make the resonant transmittance sensitive to incident intensity. The IMT in VO₂ can be a source of giant photothermal nonlinearity, which enabled the use of low-quality-factor FSS resonances in our design, resulting in open-state transmittance with large angular and temporal bandwidth. Our prototype reflective limiter designed for a wavelength of 10.6 μm has high open-state transmittance (≈0.7), low limiting-state transmittance (<0.01) and absorbance (≈0.06), broad working bandwidth (FWHM > 2 μm), and functions for all polarizations up to an incident angle of ≈50°.

6. Experimental Section

VO₂ was deposited onto double-side-polished undoped GaAs (001) wafers via magnetron sputtering from a V₂O₅ target, with radio-frequency power of 100 W. During deposition, the chamber pressure was maintained at 5 mTorr with an Ar/O₂ gas mixture at a flow rate of 49.85/0.15 sccm. The substrate was heated to 700 °C to form the VO₂ phase.

We built a 50-nm gold FSS on top of the as-grown VO₂ via the following steps. First, a 250-nm PMMA (495 PMMA A4) was spin-coated onto the VO₂ film and the pattern was written using an e-beam lithography system (Elionix GS-100). After development in MIBK/IPA (volume ratio of 1/3), an array of cross-shape PMMA blocks were left on top of the VO₂ film. Then, 50 nm of gold was evaporated, and the sample was soaked in an acetone bath for a few minutes to lift-off the gold-PMMA blocks, leaving an array of cross-slit apertures within the gold film. Note that a 60-s sonication was necessary during the lift-off process.

Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.

Acknowledgements
M.A.K. is supported by the Office of Naval Research (ONR, N00014-16-1-2556 and N00014-20-1-2297), with partial support from the Air Force Office of Scientific Research (AFOSR, FA9550-18-1-0046). S.R. is supported by the AFOSR (FA9550-18-1-0259).

Conflict of Interest
The authors declare no conflict of interest.

Data Availability Statement
The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords
frequency-selective surfaces, optical limiter, optical nonlinearity, phase-change materials, vanadium dioxide

Received: January 1, 2021  
Revised: March 3, 2021  
Published online: April 22, 2021
