Combining Switchable Phase-Change Materials and Phase-Transition Materials for Thermally Regulated Smart Mid-Infrared Modulators

Xinrui Lyu, Andreas Heßler, Xiao Wang, Yunzhen Cao,∗ Lixin Song, Alfred Ludwig, Matthias Wuttig, and Thomas Taubner∗

Phase-change materials (PCMs) and phase-transition materials (PTMs) both show a large contrast in their respective optical properties upon switching, enabling compact optical components with diverse functionalities like sensing, thermal imaging, and data recording. However, their switching properties differ significantly, that is, the switching is non-volatile for PCMs while volatile for PTMs. Here, new-generation smart mid-infrared modulators with switchable transmission, reflection, and absorption are demonstrated conceptually and experimentally, which combine one PCM (Ge₃Sb₂Te₅ or In₃SbTe₂) with one PTM (VO₂) as two active layers. The bottom VO₂ layer is employed as a thermally regulated (modulated) dynamic mirror, facilitating the switching of transmission between “on” state (using VO₂ in its semiconducting state at temperatures below its phase transition temperature \( T_c \)) and “off” state (metallic VO₂ at temperatures above \( T_c \)). The PCM layer on top of the metallic VO₂ layer is used either for continuously adjusting the absorption peak spectrally (by up to 1.8 \( \mu \text{m} \) using different phases of Ge₃Sb₂Te₅) or for switching between absorption mode (\( A = 0.99 \) with amorphous In₃SbTe₂) and reflection mode (\( R = 0.85 \) with crystalline In₃SbTe₂). The presented concept of merging static, non-volatile thermal switching (via PCMs) with dynamic, volatile thermal modulation (via PTMs) empowers a new generation of optical devices for smart optical switching, for example in spectrally tunable safety optical switches.

1. Introduction

Because of the conservation of energy, the total amount of transmittance, absorptance, and reflectance of optical films equals unity (\( \text{Trans} + A + R = 1 \)), with these three quantities dependent on each other. The modulation of these quantities enables applications in optical components such as anti-reflective coatings,[1] optical filters,[2] and optical absorbers.[3] Generally, their optical properties can be tuned by designing the structural geometries and materials involved, which remain fixed after fabrication. Using phase-change materials (PCMs) and phase-transition materials (PTMs) as active optical layers provides the optical components with new features of switchable optical properties. PCMs have at least two (meta-)stable phases, that is, one amorphous phase and one or more crystalline phases with high contrast in their electrical and optical properties.[4] A PCM can be crystallized by a thermal, optical, or electrical stimulus which heats it above its glass transition temperature \( T_g \). The phase change is non-volatile, which means the switched phase is maintained even after the switching stimulus ends.[4,5] Common PCMs like GeTe and GeSbTe-derivatives are, for example, applied in rewritable data storage devices.[6–8] In contrast, PTMs also have a temperature-stimulated, but volatile phase transition, where the switched phase is only maintained at temperatures above its transition temperature \( T_c \). These materials automatically change back to the original phases when cooled down below \( T_c \). Therefore, their optical and electrical properties change reversibly with the temperature. A commonly used example of PTMs is vanadium dioxide (VO₂), which undergoes a semiconductor-metal phase transition.
phase transition at around 68 °C.\[^{16}\] Note that there is a hysteresis inherent to the VO\textsubscript{2} phase transition. The temperature-dependent optical properties of VO\textsubscript{2}\[^{16}\] enable, for example, applications like optical switches, thermal rectifiers, and smart radiative cooling coatings in the infrared\[^{11–15}\] and switchable quarter-wave plates in the THz range\[^{16}\].

Both PCMs and PTMs have been used as a single active layer in optical component designs with switchable optical properties. A perfect absorber using Ge\textsubscript{3}Sb\textsubscript{2}Te\textsubscript{6} as an active spacer layer showed a phase-change-induced spectral redshift in the mid-infrared (MIR) range, which demonstrated multispectral thermal imaging capability.\[^{17}\] VO\textsubscript{2} has also been employed as an active spacer in a metamaterial absorber, which shows a spectrally selective thermal emission with “on” (semiconducting VO\textsubscript{2}) and “off” (metallic VO\textsubscript{2}) states.\[^{18}\] Such switchable properties only enable single-feature adjustment: either spectrally shifting the resonance peak or switching between “on” and “off” states. Recently, a 4-level optical switch based on the combination of Ge\textsubscript{3}Sb\textsubscript{2}Te\textsubscript{6} and VO\textsubscript{2} was realized in the visible spectral range for wavelengths between 660 and 700 nm.\[^{19}\] There, three layers of VO\textsubscript{2}, Ge\textsubscript{3}Sb\textsubscript{2}Te\textsubscript{6}, and Al were illuminated through a SiO\textsubscript{2} substrate and the average reflectance in the studied spectral range was switched to four different levels between 56% and 100% by first heating the sample up to 190 °C and then cooling it back down to room temperature. Although achieving a respectable overall reflectance contrast of 44%, the switch could only be operated in a reflectance mode because it incorporated an inactive Al reflective layer. Moreover, it was only studied in a narrow spectral range in the visible regime. Here, we instead work in the broad infrared spectral range where thermal emission occurs and significantly extend this concept by using only a two-layer stack consisting of a PCM layer (for static tuning) on top of a VO\textsubscript{2} layer (for dynamic modulation) on a transparent CaF\textsubscript{2} substrate without the additional Al reflection layer. Our proposed smart optical modulators can switch between transmission, reflection, and absorption modes by temperature modulation. We additionally study their continuous spectral tuning through a broad MIR wavelength range and demonstrate designs using two PCMs of distinctly different optical properties, that is, Ge\textsubscript{3}Sb\textsubscript{2}Te\textsubscript{6} (GST) and In\textsubscript{1}SbTe\textsubscript{2} (IST).

![Image](image.png)

Figure 1 shows the concept of the thermally regulated smart MIR modulators. Two PCMs, GST and IST, are used in this paper to demonstrate different multi-feature modulations, which are caused by the very different optical properties of GST and IST (see Figure S1, Supporting Information). Both GST and IST have an amorphous phase, which reveals properties that are characteristic of covalently bonded compounds. These materials (in their amorphous phases) possess a moderate band gap and are poor electrical conductors.\[^{120–124}\] The crystalline phase of GST differs significantly from its amorphous counterpart due to a unique chemical bonding mechanism called “metavalent bonding”,\[^{25,26}\] which is neither compatible with metallic, covalent, or ionic bonding. This unique bonding mechanism is one of the characteristic features of PCMs.\[^{27,28}\] Interestingly, the crystalline phases of GST and IST also differ significantly: IST is much more conductive than GST. It is so conductive that it is best described as a “bad metal”.\[^{23}\] These differences provide exciting opportunities for optical modulators utilizing GST or IST with different functionalities.

In our first design (Figure 1b), GST and VO\textsubscript{2} are combined to form a thermally regulated MIR modulator. By heating the VO\textsubscript{2} bottom layer, the modulator can be dynamically switched between transmission mode ($T < T_g$, semiconducting VO\textsubscript{2}) and absorption mode ($T > T_g$, metallic VO\textsubscript{2}). Therefore, the VO\textsubscript{2} acts as a modulation layer. In both modes, the operation wavelength can also be tuned via switching the GST layer from the amorphous ($T < T_m$) to crystalline ($T > T_m$) phases. Therefore, the GST acts as the tuning layer. Because of the large optical contrast between the amorphous and crystalline phases of GST and the low optical losses compared with other PCMs in the infrared,\[^{29}\] our modulator achieves an improved optical contrast.

In our second design (Figure 1c), the novel PCM IST is used instead of GST, which uniquely features a non-volatile change from dielectric to metallic optical properties in the infrared upon crystallization. It was recently introduced as a programmable nanophotonics material platform for the infrared and can be classified as a switchable “bad metal”, as mentioned above.\[^{23}\] The IST/VO\textsubscript{2} modulator can dynamically switch between transmission ($T < T_g$, semiconducting VO\textsubscript{2}) and absorption ($T > T_g$, metallic VO\textsubscript{2}) modes at a fixed operation wavelength given by the IST layer thickness. In addition,
one can switch from either of these two modes to a reflection mode by crystallizing \( T > T_g \) and thus metalizing the top IST layer.

To experimentally demonstrate the multi-feature modulations of the smart optical modulators illustrated in Figure 1, a GST/VO\(_2\) layer stack and an IST/VO\(_2\) layer stack were sputtered, and their MIR reflectance and transmittance were measured and normalized to optically thick gold and transparent CaF\(_2\), respectively (see Experimental Section). To prevent the potential oxidation of GST and IST during crystallization (at 200 °C for GST and at 300 °C for IST in the experiment), 80 nm ZnS:SiO\(_2\) was sputtered on the top of the layer stacks as a capping layer, which has little influence on the MIR properties of these two layer stacks (see Figure S2, Supporting Information).

### 1.1. GST/VO\(_2\) Smart Modulator

Figure 2a shows a cross-sectional scanning electron microscope (SEM) image of the ZnS:SiO\(_2\)/GST/VO\(_2\) layer stack on a CaF\(_2\) substrate with thicknesses of 80, 150, and 250 nm, respectively. Figure 2b displays the measured transmittance spectra for the layer stack with semiconducting VO\(_2\) (s-VO\(_2\)) at the bottom. It shows a resonance peak in transmittance at 3.2 \( \mu m \) wavelength.

![Figure 2](image-url)

**Figure 2.** The switchable transmission and the spectrally tunable absorption using a GST/VO\(_2\) layer stack. a) A cross-sectional SEM image of the layer stack of 80 nm ZnS:SiO\(_2\) capping layer, 150 nm Ge\(_3\)Sb\(_2\)Te\(_5\), and 250 nm VO\(_2\) layers deposited on a CaF\(_2\) substrate. b) The transmittance spectra of a-GST/s-VO\(_2\) (blue curve) and c-GST/s-VO\(_2\) (green curve) layer stacks measured at 50 °C. c,d) The transmittance spectra of c-GST/VO\(_2\) stacks measured at 50 °C < \( T < 65 \) °C. e) The reflectance spectra of a-GST/m-VO\(_2\) (solid blue curve), c-GST/m-VO\(_2\) (solid green curve), and partially crystallized pc-GST/m-VO\(_2\) (dashed cyan curve, which is prepared by annealing a-GST at 160 °C for 150 s) measured at 90 °C. f) The tuning of the reflectance minimum position measured at 90 °C using partially crystalline GST annealed at 160 °C with different annealing time.
with a transmission of about \( T_n = 0.91 \). By crystallizing the top GST layer, the transmittance can be tuned in peak position to about 4.2 \( \mu m \) with reduced peak intensity. This results in a high transmittance contrast of 0.67 (\( T_n = 0.91 \) for a-GST/s-VO\(_2\) and \( T_n = 0.24 \) for c-GST/s-VO\(_2\)) at 3.2 \( \mu m \). Since both a-GST and c-GST are almost lossless in MIR,[29,30] the generally lower transmittance of c-GST/s-VO\(_2\) is due to the larger refractive index of c-GST (compared with a-GST) and thus higher reflectance of c-GST/s-VO\(_2\), and not due to additional absorption (see Figure S3, Supporting Information). When increasing the temperature, the transmitance spectra of both a-GST/VO\(_2\) (Figure 2c) and c-GST/VO\(_2\) (Figure 2d) show red-shifted peak positions and decreased amplitudes until the transmittance reaches zero at 65 °C. The decreased transmittance is caused by the VO\(_2\) transition from its transparent semiconducting to its reflecting metallic phase.

At temperature \( T = 90 \, ^\circ C > T_c \), the VO\(_2\) layer becomes a reflective layer and the modulator is in an absorption mode (Figure 2e). The reflectance of the a-GST/m-VO\(_2\) stack (solid blue curve) is near zero at 4.1 \( \mu m \), which corresponds to a near-unity absorbance at this wavelength since no light can pass through the 250 nm-thick m-VO\(_2\) mirror (\( A = 1 - R = \text{Trans} \), \( T = 0 \)). After crystallizing the GST at 200 °C for 10 min on a hot plate, the reflectance minimum redshifts to 5.9 \( \mu m \) (solid green curve), corresponding to a tuning figure of merit TFOM = \( \Delta \lambda / \text{FWHM} = 1 \) (\( \lambda \) is the wavelength shift and FWHM is the full width at half maximum). This large shift by one-peak width is caused by the large contrast in the refractive index of the two phases of GST (\( n = 3.5 \) for a-GST to \( n = 6.1 \) for c-GST at 5 \( \mu m \)).[30] Importantly, the reflectance minimum position of GST/m-VO\(_2\) can be continuously tuned within the range of 4.1 to 5.9 \( \mu m \) by employing a partially crystallized GST.[31] For example, the dashed cyan curve exhibits a reflectance minimum at 5.1 \( \mu m \) after heating at 160 °C for a total amount of 150 s. Figure 2f shows the minimum position as a function of the annealing temperature of GST. By increasing the annealing time at intervals of 30 s, the wavelength of the reflectance minimum is gradually redshifted (saturated growth) by increasing the degree of crystallization.

Numerical simulations with CST Microwave Studio, a commercial electromagnetic wave simulation software, show good agreement with the experimental spectra. Additionally, they indicate that the spectral tuning range can be tailored during fabrication by changing the thickness of the GST layer (see Figure S4, Supporting Information). By varying the GST thickness from 30 to 270 nm, the reflectance minimum can be redshifted by 4 \( \mu m \) for a-GST/m-VO\(_2\), and even by 6.7 \( \mu m \) for c-GST/m-VO\(_2\). This also implies that the spectral tuning range enabled by partial crystallization in Figure 2f can be expanded via changing the GST thickness.

It should be mentioned that the optical thickness \( d \) of the a-GST layer to achieve a near-unity absorption at \( \lambda_p = 4.1 \, \mu m \) in Figure 2e is two times smaller than the optical thickness \( \lambda_p / 4 \) of conventional quarter-wave anti-reflection coating, that is \( n d = \lambda_p / 8 \) (\( n = 3.5 \) and \( d = 150 \, \mu m \)). The much thinner GST thickness is caused by the usage of m-VO\(_2\) (lossy metal with finite conductivity in the MIR) instead of noble metal as a reflective mirror. This replacement results in a non-trivial (not 0 or \( \pi \)) reflection phase[32,33] at the interface between GST and VO\(_2\), reducing the required thickness of the GST layer for perfect absorption.

The simple GST/VO\(_2\) layer stack with multi-feature switching properties, which is easy to fabricate, has potential for next-generation, smart optical switches. One possible application is a safety optical switch (see Figure S6, Supporting Information), which protects the optical system from excessive power (via the phase transition of VO\(_2\) at 65 °C) and cools down automatically once the “safety” has been engaged (via the phase change of GST at 150 °C).

1.2. IST/VO\(_2\) Smart Modulator

The former GST/VO\(_2\) modulator demonstrates the functions of dynamically switchable (VO\(_2\)) and spectrally tunable transmission and absorption (GST). The switching/tuning is attributed to the phase change of GST between its two dielectric phases with greatly different permittivities. In the next modulator, IST was used as a function-altering layer instead of GST. Its optical properties change from dielectric to metallic when switched from amorphous to crystalline.[23] This, therefore, provides the IST/VO\(_2\) modulator with different multi-feature switching properties.

The IST/VO\(_2\) layer stack (Figure 3) functions in three different modes (transmission, absorption, and reflection) in different temperature ranges. It consists of 80 nm Zn:S:SiO\(_2\), 100 nm IST, and 250 nm VO\(_2\) layers on a CaF\(_2\) substrate (Figure 3a). Similar to GST/VO\(_2\), the IST/VO\(_2\) modulator can also be dynamically switched between transmission and absorption modes via increasing the temperature from room temperature to the critical temperature \( T_c \) of VO\(_2\). In Figure 3b, the measured transmittance and reflectance of the modulator at room temperature (a-IST/s-VO\(_2\)) are shown. One can clearly see a peak in transmittance and a minimum in reflectance at a wavelength of about 2.7 \( \mu m \). Thus, the modulator is in transmission mode with a very high transmittance \( T = 0.97 \) (magenta curve). The high transmittance can be gradually modulated by increasing the temperature (from 0.92 at 50 °C to 0 at 65 °C) and using the intermediate states of VO\(_2\) (Figure 3c). When the temperature is above 65 °C, VO\(_2\) turns into its metallic state, and the bottom, opaque m-VO\(_2\) layer acts like a reflective mirror. This facilitates destructive interference for the a-IST/m-VO\(_2\) layer stack at 2.8 \( \mu m \), resulting in a high absorption \( A = 1 - R = 0.99 \) (Figure 3d, black curve). At significantly higher temperatures (300 °C), the top IST layer can be crystallized and it becomes highly reflective. The modulator (c-IST/m-VO\(_2\)) switches to a reflection mode with high reflectance \( R = 0.85 \) for a wide range of wavelengths \( \lambda \geq 2.8 \, \mu m \) (Figure 3e, black curve). Therefore, by putting an IST layer on top of a VO\(_2\) bottom layer and utilizing their different phase change/transition temperatures, one can switch between transmission mode (\( T = 0.97 \)), absorption mode (\( A = 0.99 \)), and reflection mode (\( R = 0.85 \)). Moreover, the reflection mode is persistent, that is, it remains even after cooling down the device below \( T_c \). The operation wavelength is fixed here but can be tailored during fabrication by changing the IST layer thickness.
Figure 3. Thermally-regulated mid-infrared transmission/absorption/reflection switch using an IST/VO$_2$ layer stack. a) A cross-sectional SEM image of the layer stack of 80 nm ZnS:SiO$_2$ capping layer, 100 nm In$_3$SbTe$_2$, and 250 nm VO$_2$ layers deposited on a CaF$_2$ substrate. b–e) The reflectance (black) and transmittance (magenta) spectra of the modulator at different temperatures. b) The transmission mode with an a-IST/s-VO$_2$ configuration measured at room temperature. c) The temperature-modulated transmittance of the a-IST/VO$_2$ stack measured at 50 °C < T < 65 °C. d) The absorption mode with an a-IST/m-VO$_2$ configuration measured at 90 °C. e) The reflection mode with a c-IST/m-VO$_2$ configuration after annealing at 300 °C.
2. Discussion

In addition to GST and IST, other PCMs can also be used as a top active layer in the design of a PCM/VO₂ smart modulator. Candidates may for example include lossless PCMs like GeSbTe[34] or lossy PCMs like AgInSbTe[35] Using PCMs with different stoichiometries and intermediate states (via different annealing processes) enables smart modulators with operating wavelengths covering the whole MIR spectral range.

Due to their relatively thin PCM layer thickness (about 2 times smaller than for conventional quarter-wavelength antireflection coatings), the modulators have a relatively wide angular bandwidth. The reflectance in the absorbing states stays near-zero in its minimum for angles of incidence between 0° and about 60° (see transfer matrix method calculations in Figure S5, Supporting Information).

Our presented experiments, performed on a hot plate, are intended as proof-of-principle experiments and thus are not focused on reaching high modulation or switching speeds. For reaching short modulation times (∼10 ns) for the VO₂ layer, one could, for example, use electrical heating by adjacent electrodes.[36,37] The spectral tuning via the PCM layer can also be realized continuously with a much faster, well-defined, multilevel switching by applying electrical or optical methods.[7,18,19] In principle, very fast electrical and optical switching in the ns-range has been shown for memory applications.[6,40] Mostly on sub-micron length scales. In these applications, the speed-limiting factor is normally the crystallization speed and the size of the devices. By optical switching with a pulsed, focused laser, crystallization can be achieved on the scale of ~100–1000 ns.[23] To reamorphize the PCM layer, it usually has to be heated above its melting point (~600 °C)[23] and then rapidly quenched. Current research is working on the challenging topic of scaling up the switchable areas. In addition to the classical approaches of electrical heaters[41,42] and optical pulses,[43] recently also ion beam irradiation[44] was demonstrated. To ease the optical reamorphization in our design, one could decrease the PCM layer thickness (this would also speed up the crystallization) which would, in turn, shift the resonances to smaller wavelengths (see Figure S4b, Supporting Information). Alternatively, by interspersing very thin, additional layers of thermally conductive dielectrics within the PCM, reversible switching could be enabled for even effectively thicker PCM layers.

Local, optical switching of PCM layers has been demonstrated previously to strategically adapt the optical properties pixel-by-pixel without the need for additional patterning via nanofabrication techniques.[23,45–49] This idea could be applied to our proposed modulators to achieve optical programming by locally switching the PCM over the whole modulator surface while the VO₂ layer can still be modulated via temperature. Patterning the modulator layerstack should also be beneficial for decreased switching times.[50]

Since both PCMs and VO₂ show a high endurance to phase change/transition cycling,[31,37] the demonstrated modulators are expected to exhibit a long cycling lifetime.

The presented concepts of combining PTMs and PCMs, that is, volatile and non-volatile switching mechanics, within the same device are not limited to the shown smart modulators and could, for example, be extended to polariton nanophotonics.[31–54] Exploiting both the advantages of static function tuning and dynamic modulation, they promise fundamentally increased tuning capabilities of nanophotonic components.

3. Conclusion

The dynamic mid-infrared modulators with a switchable reflection, transmission, and absorption response to a thermal stimulus were created by combining a static but reconfigurable PCM layer (Ge₂Sb₂Te₅ and In₃SbTe₂ as two examples) with a dynamic VO₂ layer. The GST/VO₂ modulator demonstrates dynamic switching between a transmission mode and an absorption mode with a spectrally tunable operation wavelength. At T = 50 °C, the VO₂ is transparent and the transmission could be decreased (from 0.91 to 0.24) and redshifted (from 3.2 to 4.2 µm) by crystallizing the top GST layer, resulting in a transmittance contrast of 0.67 at 3.2 µm. At T ≥ 65 °C, the VO₂ is in its metallic state and the modulator shows a spectrally tunable absorption, which can be continuously tuned by up to 1.8 µm (from 4.1 to 5.9 µm) via the crystallization of the GST layer. The IST/VO₂ modulator demonstrates a thermally regulated switching between a transmission mode (T = 25 °C, Trans = 0.97), an absorption mode (T > 65 °C, A = 0.99) and a reflection mode (T ≥ 300 °C, R = 0.85) at 2.8 µm by first turning the VO₂ layer metallic and then the IST layer metallic. In general, the concept of combining non-volatile adjustment (PCMs) and dynamic temperature-dependent modulation (VO₂) in one system creates a new platform for smart (reacting to thermal stimulus) and tunable (spectrally shifting) optical components.

4. Experimental Section

Fabrication of Layer Stacks: All layer stacks were sputtered on CaF₂ substrates. The VO₂ bottom layer, about 250 nm thick, was fabricated by reactive sputtering from an elemental V target of 99.99% purity using a magnetron sputter system (AJA International, USA) at 520 °C. Pulsed DC power of 300 W (pulsing frequency of 10 kHz and a reverse time of 5 μs) was applied to the V target during sputtering under a chamber pressure of 0.4 Pa with 80 sccm of Ar. A plasma emission monitor system (Flotron, Nova Fabrika, Lithuania) was used to dynamically control the oxygen flow rate,[35] which was necessary to reproducibly obtain stoichiometric VO₂ films. Because the VO₂ layer was relatively thick at 250 nm, detrimental effects of stress on the film quality were negligible. The amorphous Ge₂Sb₂Te₅ and In₃SbTe₂ layers were sputtered using stoichiometric targets of 99.99% purity by direct current magnetron sputtering with a working pressure of 0.6 Pa and 20 sccm Ar flow. A (2x5)nm (SiO₂)₉ capping layer was then sputtered using radio frequency magnetron sputtering on top of the PCM layers to avoid oxidation. GST and IST layers were crystallized by heating at 200 and 300 °C in air for 10 min on a hot plate. The intermediate states of GST were obtained by successive annealing steps at 160 °C from 90 to 300 s on the hot plate.

SEM and Fourier-transform Infrared (FTIR) Spectroscopy Measurements: The thickness and morphology of the layer stacks were characterized by SEM (JEOL JSM-7200F). The reflectance and transmittance spectra of the layer stacks were measured by an FTIR spectrometer (Bruker Vertex 70) connected with an infrared microscope (Bruker Hyperion 2000) using one (for reflectance) or two (for transmittance spectra) 15 × objectives. The spectral resolution and scan time were 16 cm⁻¹ and 800 scans for both reflectance and transmission modes. The reflectance was normalized using an optically thick gold standard, and the transmission
was normalized using a bare CaF₂ substrate with the same thickness as that used for the samples. The operating temperature was set and controlled with a Linkam TP95 thermal stage.

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

Acknowledgements
X.L and A.H. contributed equally to this work. X.L. acknowledges the funding from the CAS-DAAD (Chinese Academy of Sciences – German Academic Exchange Service) scholarship for doctoral students. T.T., M.W., and A.H. acknowledge funding from the German Federal Ministry of Education and Research within the funding program Photonics Research Germany (contract number 13N14151) and the DFG (German Research Germany (contract number 13N14151) and the DFG (German Science Foundation) within the collaborative research center SFB 917 “Nanoswitches.” X.W. and A.L. acknowledge the funding from DFG (project LU1175/18-1) and the support in SEM measurements from ZGH (the Center for Interface-Dominated High Performance Materials). The authors acknowledge Denise Kühn for sputtering the Ge₃Sb₂Te₅ and In₅SbTe₅ films.

Open access funding enabled and organized by Projekt DEAL.

Conflict of Interest
The authors declare no conflict of interest.

Data Availability Statement
Research data are not shared.

Keywords
optical modulators, phase-change materials, phase-transition materials, switchable absorbers, thin film interference

Received: February 26, 2021
Revised: April 25, 2021
Published online: May 31, 2021

[1] H. Kumar Raut, V. Anand Ganesh, A. Sreekumar Nair, S. Ramakrishna, *Energy Environ. Sci.* 2011, 4, 3779.
[2] H. A. Macleod, *Thin-Film Optical Filters*, CRC Press, Boca Raton, FL 2010.
[3] M. A. Kats, F. Capasso, *Laser Photonics Rev.* 2016, 10, 735.
[4] M. Wuttig, H. Bhaskaran, T. Taubner, Nat. Photonics 2017, 11, 465.
[5] D. Loke, T. H. Lee, W. J. Wang, L. P. Shi, R. Zhao, Y. C. Yeo, T. C. Chang, S. R. Elliott, *Science* 2012, 336, 1566.
[6] M. Wuttig, N. Yamada, *Nat. Mater.* 2007, 6, 824.
[7] Z. Cheng, C. Rios, N. Youngblood, C. D. Wright, W. H. P. Pernice, H. Bhaskaran, *Adv. Mater.* 2018, 30, 1802435.
[8] C. Rios, M. Stegmaier, P. Hosseini, D. Wang, T. Scherer, C. D. Wright, H. Bhaskaran, W. H. P. Pernice, *Nat. Photonics* 2015, 9, 725.
[9] N. F. Mott, L. Friedman, *Philos. Mag.* 1974, 30, 389.
[10] H. W. Verleur, A. S. Barker, C. N. Berglund, *Phys. Rev.* 1968, 172, 788.
[11] C. Wan, E. H. Horak, J. King, J. Salman, Z. Zhang, Y. Zhou, P. Roney, B. Gundlach, S. Ramanathan, R. H. Goldsmith, M. A. Kats, *ACS Photonics* 2018, 5, 2688.
[12] A. Thomas, P. Savaliya, K. Kumar, A. Ninawe, A. Dhawan, *J. Opt. Soc. Am. B* 2018, 35, 1687.
[13] B. A. Kruger, A. Joushangahi, J. K. S. Poon, *Opt. Express* 2012, 20, 23598.
[14] K. Sun, C. A. Rios, A. Urban, M. Simeoni, S. Mengali, M. Zalkovskij, B. Bilenberg, C. H. de Groot, O. L. Musken, *ACS Photonics* 2018, 5, 2280.
[15] R. Audhikhsa, M. L. Povinelli, *J. Appl. Phys.* 2019, 126, 063106.
[16] D. Wang, L. Zhang, Y. Gu, M. Q. Mehmood, Y. Gong, A. Srivastava, L. Jian, T. Venkatesan, C.-W. Qiu, M. Hong, *Sci. Rep.* 2015, 5, 15020.
[17] A. Tittl, A.-K. U. Michel, M. Schäferling, Y. Yin, B. Gholiapour, L. Cui, M. Wuttig, T. Taubner, F. Neubrech, H. Giessen, *Adv. Mater.* 2015, 27, 4597.
[18] H. Wang, Y. Yang, L. Wang, *Appl. Phys. Lett.* 2014, 105, 071907.
[19] Y. Meng, J. K. Behera, Y. Ke, L. Chew, Y. Wang, Y. Long, R. E. Simpson, *Appl. Phys. Lett.* 2018, 113, 071901.
[20] J. M. Vázquez-Limón, J. González-Hernández, J. J. Alvarado-Gil, L. Delgadillo, H. Vargas, *Phys. Rev. B* 1995, 52, 16321.
[21] E. T. Kim, J. Y. Lee, Y. T. Kim, *Phys. Status Solidi RRL* 2009, 3, 103.
[22] V. L. Deringer, W. Zhang, P. Rausch, R. Mazzarelli, R. Dronskowski, M. Wuttig, J. Mater. Chem. C 2015, 3, 9519.
[23] A. Heßler, S. Wahl, T. Leuteritz, A. Antonopoulos, C. Stergianou, C.-F. Schön, L. Naumann, E. Ecker, M. Lewin, T. W. Maß, M. Wuttig, S. Linden, T. Taubner, *Nat. Commun.* 2021, 12, 924.
[24] S. Selmo, R. Cecchini, S. Cecchi, C. Wiemer, M. Fanciulli, E. Rotunno, L. Lazarini, M. Rigato, D. Pogany, A. Augstein, M. Longo, *Appl. Phys. Lett.* 2016, 109, 213103.
[25] M. Zhu, O. Cojocaru-Mirédin, A. M. Mio, J. Keutgen, M. Küpers, Y. Yu, J.-Y. Cho, R. Dronskowski, *M. Wuttig, Adv. Mater.* 2018, 30, 1706735.
[26] J.-Y. Raty, M. Schumacher, P. Golub, V. L. Deringer, C. Gatti, M. Wuttig, *Adv. Mater.* 2019, 31, 1806280.
[27] M. Wuttig, V. L. Deringer, X. Gonze, C. Bichara, J.-Y. Raty, *Adv. Mater.* 2018, 30, 1803777.
[28] B. J. Kooi, M. Wuttig, *Adv. Mater.* 2020, 32, 1908302.
[29] A.-K. U. Michel, M. Wuttig, T. Taubner, *Adv. Opt. Mater.* 2017, 5, 1700261.
[30] A.-K. U. Michel, D. N. Chigrin, T. W. Maß, K. Schöner, M. Salinger, M. Wuttig, T. Taubner, *Nano Lett.* 2013, 13, 3470.
[31] Y. G. Chen, T. S. Kao, B. Ng, X. Li, X. G. Luo, B. Luk’yanchuk, S. A. Maier, M. H. Hong, *Opt. Express* 2013, 21, 13691.
[32] M. A. Kats, D. Sharma, J. Lin, P. Genevet, R. Blanchard, Z. Yang, M. M. Qazilbash, D. N. Basov, S. Ramanathan, F. Capasso, *Appl. Phys. Lett.* 2012, 101, 221101.
[33] M. A. Kats, R. Blanchard, P. Genevet, F. Capasso, *Nat. Mater.* 2013, 12, 20.
[34] Y. Zhang, J. B. Chou, J. Li, H. Li, Q. Du, A. Yadav, S. Zhou, M. Y. Shalaginov, Z. Fang, H. Zhong, C. Roberts, P. Robinson, B. Bohlín, C. Rios, H. Lin, M. Kang, T. Gu, J. Warner, V. Liberman, K. Richardson, J. Hu, *Nat. Commun.* 2019, 10, 4279.
[35] M. S. Arjunan, A. Mondal, A. Das, K. V. Adarsh, A. Manivannan, *Opt. Lett.* 2019, 44, 3134.
[36] M. Sadeghi, B. Janjan, M. Heidari, D. Abbott, *Opt. Express* 2020, 28, 9198.
[37] Y. Zhou, X. Chen, C. Ko, Z. Yang, C. Mouli, S. Ramanathan, *IEEE Electron Device Lett.* 2013, 34, 220.
[38] X. Li, N. Youngblood, C. Rios, Z. Cheng, C. D. Wright, W. H. P. Pernice, H. Bhaskaran, *Optics* 2019, 6, 1.
[39] G. A. Sevison, S. Farzinazar, J. A. Burrow, C. Perez, H. Kwon, J. Lee, M. Asheghi, K. E. Goodson, A. Sarangan, J. R. Hendrickson, I. Agha, *ACS Photonics* 2020, 7, 480.
[40] G. Bruns, P. Merkelbach, C. Schlockermann, M. Salinga, M. Wuttig, T. D. Happ, J. B. Philipp, M. Kund, Appl. Phys. Lett. 2009, 95, 043103.

[41] Y. Zhang, C. Fowler, J. Liang, B. Azhar, M. Y. Shalaginov, S. Deckoff-Jones, S. An, J. B. Chou, C. M. Roberts, V. Liberman, M. Kang, C. Rios, K. A. Richardson, C. Rivero-Baleine, T. Gu, H. Zhang, J. Hu, Nat. Nanotechnol. 2021, https://doi.org/10.1038/s41565-021-00881-9.

[42] Y. Wang, P. Landreman, D. Schoen, K. Okabe, A. Marshall, U. Celano, H.-S. P. Wong, J. Park, M. L. Brongersma, Nat. Nanotechnol. 2021, https://doi.org/10.1038/s41565-021-00882-8.

[43] A.-K. U. Michel, P. Zalden, D. N. Chigrin, M. Wuttig, A. M. Lindenberg, T. Taubner, ACS Photonics 2014, 1, 833.

[44] M. Hafermann, P. Schöppe, J. Rensberg, C. Ronning, ACS Photonics 2018, 5, 5103.

[45] P. Li, X. Yang, T. W. W. Maß, J. Hanss, M. Lewin, A.-K. U. Michel, M. Wuttig, T. Taubner, Nat. Mater. 2016, 15, 870.

[46] Q. Wang, E. T. F. Rogers, B. Gholipour, C.-M. Wang, G. Yuan, J. Teng, N. I. Zheludev, Nat. Photonics 2016, 10, 60.

[47] K. Dong, S. Hong, Y. Deng, H. Ma, J. Li, X. Wang, J. Yeo, L. Wang, S. Lou, K. B. Tom, K. Liu, Z. You, Y. Wei, C. P. Grigoropoulos, J. Yao, J. Wu, Adv. Mater. 2018, 30, 1703878.

[48] A.-K. U. Michel, A. Heßler, S. Meyer, J. Pries, Y. Yu, T. Kalix, M. Lewin, J. Hanss, A. De Rose, T. W. W. Maß, M. Wuttig, D. N. Chigrin, T. Taubner, Adv. Mater. 2019, 31, 1901033.

[49] A. Leitis, A. Heßler, S. Wahl, M. Wuttig, T. Taubner, A. Tittl, H. Altg, Adv. Funct. Mater. 2020, 30, 1910259.

[50] H. Wang, Y. Yang, L. Wang, J. Appl. Phys. 2014, 116, 123503.

[51] K. Chaudhary, M. Tamagnone, X. Yin, C. M. Spägele, S. L. Oscurato, J. Li, C. Persch, R. Li, N. A. Rubin, L. A. Jauregui, K. Watanabe, T. Taniguchi, P. Kim, M. Wuttig, J. H. Edgar, A. Ambrosio, F. Capasso, Nat. Commun. 2019, 10, 4487.

[52] N. C. Passler, A. Heßler, M. Wuttig, T. Taubner, A. Paarmann, Adv. Opt. Mater. 2020, 8, 1901056.

[53] H. Sumikura, T. Wang, P. Li, A.-K. U. Michel, A. Heßler, L. Jung, M. Lewin, M. Wuttig, D. N. Chigrin, T. Taubner, Nano Lett. 2019, 19, 2549.

[54] T. G. Folland, A. Fali, S. T. White, J. R. Matson, S. Liu, N. A. Aghamiri, J. H. Edgar, R. F. Haglund, Y. Abate, J. D. Caldwell, Nat. Commun. 2018, 9, 4371.

[55] X. Wang, Y. Cao, Y. Zhang, L. Yan, Y. Li, Appl. Surf. Sci. 2015, 344, 230.