A Reversible Tuning of High Absorption in Chalcogenide–Metal Stacked-Layer Structure and Its Application for Multichannel Biosensing

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Perfect absorption ranging from visible, infrared, terahertz, to microwave is desirable for solar cell, photodetection, telecommunications, and molecular sensing. Recently, the air/dielectric–metal stacks/substrate-based asymmetric Fabry–Pérot (FP) cavity has attracted much attention owing to lithography-free design which is scalable and low cost. Herein, a reversibly tunable asymmetric FP cavity high absorber in the near-infrared (NIR) region is experimentally demonstrated, relying on chalcogenide (Ge$_2$Sb$_2$Te$_5$)–metal (Au) stacked layers. It exhibits an extremely high absorptance of 0.99 at a resonant wavelength of 1180 nm for amorphous (AM) state; yet, the peak absorptance redshifts to 1680 nm for crystalline (CR) state. Importantly, it takes about 5 ns to reversibly transit the peak absorptance by reamorphizing the GST225. It is also experimentally shown the Brewster modes can be excited in the cavity absorber, and a wide tuning of Brewster modes (from 820 to 1500 nm) is realized as switching the state between AM and CR. The Goos–Hänchen (GH) shift can be observed at the Brewster angle for each state. For the proof of concept multibiosensing application, the cavity to detect two different solutions of the copper sulfate and glucose in both AM and CR states is numerically functionalized, respectively.

1. Introduction

In the past decade, high absorption of light becomes a rising research field which has been intensely explored in many applications, including solar cell, photodetectors, thermal imaging, color filters, and biosensing.\cite{1-6} Recently, metamaterial (MM)-based absorbers have attracted much attention because they can achieve nearly high absorptance.\cite{7} For example, photonic hypercrystals, a combination between hyperbolic MMs and photonics crystals, were proposed to offer wideband enhancement in light outcoupling and spontaneous emission rate.\cite{8} A ribbon array with multiple resonances was demonstrated to reversibly switch extinction in the midinfrared (MIR) region of 3–5 μm through doping control.\cite{9} However, most of the meta-absorbers are lithographically intense, i.e., the subwavelength-scale resonator is required to be made using focused ion beam (FIB) milling\cite{9} or electron-beam lithography (EBL).\cite{9} This severely hinders the scalability and increases the cost of meta-absorber particularly for the high-frequency regions such as near-infrared (NIR).\cite{10} The NIR regime is, however, a crucial spectrum in which there are molecular vibrational fingerprints.\cite{11,12} Very recently, perfect absorptance in the visible (vis)–NIR region has been both numerically predicted\cite{13,14} and experimentally illustrated\cite{15,16,17} by using lithography-free stacked layers that exhibit the robust interaction effects. For example, a few strategies have presented high absorptance in metal–dielectric–metal Fabry–Pérot (FP)-like cavities, operating in not only wideband\cite{17} but also narrowband\cite{18} regions. Other approach with a similar design but using periodic arrays of dielectric–graphene stacked film grating to couple the light to the waveguide modes to obtain wideband absorptance in the visible region was also shown.\cite{19} However, in current state-of-the-art designs, most of the FP cavity light absorbers are restrained to a fixed spectrum. It is because the optical response of FP cavity absorber is decided by its dielectric and metal elements and the filling ratio, which is fixed after the fabrication. Recently, a bismuth (Bi)/Al$_2$O$_3$/Ag trilayered FP cavity has been proposed to obtain a high absorptance, where the peak absorptance can be spatially tuned in the MIR region by varying the thicknesses of Bi nanofilm and Al$_2$O$_3$ spacer.\cite{20} For the NIR light, in order to vary the working spectrum or function, one has to engineer the geometry of the FP cavity with nanosize accuracy, which increases the cost, time, and difficulty of tunable optical system. There are, however, numerous applications where promptly tunable photonic devices are in high requirement. In that regard, functional materials with either electrically or optically tunable characteristics are desired.
The discovery of chalcogenide alloys of Ge–Sb–Te (GST) opens the door for not only photonic data storage but also tunable photonic devices. The GST possesses two stable structural phases of amorphous (AM) and crystalline (CR) and undertakes a reversible phase change under external stimulus, which is either optical or electrical. For example, a prototypical GST alloy, Ge$_2$Sb$_2$Te$_5$ (GST225), undergoes a thermally driven AM-to-CR structural state transition above 433 K taking the GST225 from AM state to polycrystalline cubic state. On the contrary, one can reversibly transit the phase of GST225 by melt-quenching the lattice into the AM state. In this reamorphization processing, the GST225 film needs to be heated above a temperature of 913 K by nanosecond (ns) pulsed electrical or optical heating. During structural changes, the GST225 can pronounce its thermal, electrical, and optical characteristics. Particularly, the GST225 is a promising active component for switchable and ultrafast photonic devices owing to its high cyclability, ultrafast phase change, and excellent thermal stability. For instance, the GST alloys have been integrated with the photonics structures to modulate their optical features such as negative refraction, perfect lens, chirality, nonlinearity, thermal radiation, plasmon modulator, and beam steering. Very recently, the GST layer has also been proposed as a suitable functional material to perform tunable near-perfect absorbance both in periodic metasurfaces and in planar multilayered structures without lithographic patterns. However, thus far very few efforts were made to obtain a reversibly tunable absorbance. There is, nevertheless, realization of reversible tuning of high light absorbance which is important for many applications, including selective thermal emission, electro-optic switching and tunable detection.

In this work, a tunable high absorbance was illustrated in the air/Ge$_2$Sb$_2$Te$_5$–Au stacks/SiO$_2$ substrate-based asymmetric Fabry–Pérot (FP) cavity, where the GST225 and Au films have subwavelength thickness. The absorbance mechanism is caused by strong absorbance effects in the thin GST225 layers due to strong interaction between multiple reflected light beams. The high absorbance in the NIR region is very sensitive to variations in the optical properties of GST225, which can be reversibly tuned by switching the state of GST225 between AM and CR. The high absorbance in the lithography-free planar multilayer structure is omnidirectional for transverse magnetic (TM)–incident polarizations. We also perform a numerical simulation relied upon the finite-difference time-domain (FDTD) method to validate the experimental measurement. The nonvolatile feature of reversible tunable absorber makes it only need the energy for the phase switching process and not for maintaining a certain structural state. Namely, once the asymmetric FP cavity absorber is switched, it can maintain its optical characteristic until it is switched again that is attractive from a green technology prospect. Furthermore, the Brewster mode can be excited in the phase change cavity absorber, and a wide tuning of the Brewster mode from 820 to 1500 nm is experimentally realized as switching the state between AM and CR. We numerically demonstrate that Goos–Hänchen (GH) shift occurs at the Brewster angle for each structural state, and the cavity absorber with tunable Brewster mode can work as multichannel refractive index (RI) sensor. Different from most of existing RI sensor that can only measure the RI change of one type of solution, herein by leveraging the reversibly tunable high absorber platform, we present the sensing of two different solutions of the copper sulfate and glucose in both AM and CR states, respectively. The characteristic absorbance wavelengths of the copper sulfate and glucose solutions are of 800 and 1650 nm, respectively. Correspondingly, the Brewster mode (detection wavelength) of the cavity absorber is tuned from $\lambda_{ba} = 820$ nm to $\lambda_{bc} = 1500$ nm. The data explore the high sensitivity obtained under 78° incident angle at the wavelength of $\lambda_{ba} = 820$ nm for the AM state when the concentration of copper sulfate solution was changed. By transiting the state from AM to CR, the absorber can achieve the high sensitivity under 78° incident angle at the wavelength of $\lambda_{bc} = 1500$ nm for the various concentrations of glucose solution.

2. Results and Discussion

A scheme of the reversible tunable high absorber in the NIR region is shown in Figure 1a. The two periods of Au/GST225 stacked layers were deposited on the SiO$_2$ substrate using a physical vapor deposition technique (see Device Fabrication in Experimental Section). The thickness of each layer is 40 nm. Our proposed absorber can be treated as an asymmetric FP cavity consisting of air/GST225–Au stacks/SiO$_2$ substrate, where a nonvolatile bidirectional tuning function can be achieved via a reversible state transition. In Figure 1b, we show a focus ion beam (FIB) cross section of the multilayered absorber in which each GST225 and Au films is distinguished. In Figure 1c, we present the complex RI of the 40 nm-thick planar GST225 layer for both the as-deposited (AD) AM (top panel) and CR (bottom panel) states. The refractive indices of GST225 ($n = n_c + i \times k_c$) were measured via an infrared variable angle spectroscopic ellipsometer (VASE). It was seen that the $n$ exhibits a weakly dispersive $n_c$ (solid lines) and a relatively large $k_c$ (dashed lines) in the NIR region. The large $k_c$ indicates an intense optical loss in GST225 film. Meanwhile, a rapid increasing of $n_c$ (overall higher than 2) can provide a remarkable optical tuning, once the phase of GST225 changes from AD-AM to CR. Such a huge contrast of $n_c$ between structural phases is owing to a variation in the chemical bonding from covalent in the AM phase to the resonant bonding after crystallization. Thus, the GST225 plays an important role in not only attaining a high absorbance but providing tunable response by state change. The planar multilayer absorber is thermally heated under $T_g = 473$ K inside Ar atmosphere for 30 min. Such a temperature is beyond the crystallization temperature of $T_m = 433$ K whereas smaller than the melting temperature of $T_m = 903$ K, which transits the structural state from AD-AM to CR. A Drude model was used to fit the measured permittivity of the Au film (Figure S1, Supporting Information). Our proposed asymmetric multilayer FP cavity possesses near-perfect light absorbance due to the presence of high optical losses in the GST225 films amplified by the FP cavity resonance. Herein, the absorbance amplification is caused by robust interaction effects owing to the phase accumulation as the light transmits through the absorbing GST225 films; it cancels the reflectance while increasing the absorbance. The
mechanism of reflection cancellation is similar to that occurring in λ/4 films, where a phase shift of π takes place between partially and primary reflected light beams. In Figure 1d, we directly measure the reflectance (R(ω)) using VASE under incident angle of 20° (see Optical Characterization in Experimental Section). The multilayer absorber exhibits a null transmittance that simplifies the absorptance to A(ω) = 1 − R(ω). In the top panel, we experimentally measure the A(ω) for the two different structural states. As is seen, the FP cavity absorber has a near-perfect absorptance of A(ω) = 0.99 around the resonant wavelength of λ₁ = 1180 nm for the AM state. As changing the structural phase from AM to CR, the peak A(ω) massively redshifts to λ₂ = 1680 nm while having the high absorptance of A(ω) = 0.88. In the bottom panel, we have numerically simulated the A(ω) spectra using the Lumerical Solution based upon FDTD solver. The absorber is simulated under a vertical plane wave illumination (see Numerical Simulation in Experimental Section). The numerical simulation validates the spectral shifting trend by transiting the structural phase and it illustrates that the simulated A(ω) well matches the measured A(ω). To simplify the numerical model, we do not consider the fabrication defects and surface roughness. This simplification causes the differences between the simulation and measurement. In addition, the A(ω) spectra are measured under an oblique incidence of 20° in the far-field. Nevertheless, in the numerical model, the A(ω) spectra are calculated under normal incidence in the near-field, which also induces the differences. The GST layer supports a large quantity of switching cycles between AM and CR states, which is a crucial merit for tunable photonic devices. Herein, the reversible state transition provides a significant variation in the complex RI of GST225, enabling a reversible tuning of peak absorptance. In Figure S2, Supporting Information, the treatment of reversible tuning of Au/GST225 stacked-layer absorber was schematically demonstrated. In Figure 2a, we show a home-built laser system made of Q-switch pulsed laser (Quintel, Q-smart 850) working at λ = 1064 nm, and 5 ns pulse duration with a 10 Hz repetition rate is used to reamorphize the multilayer absorber. The highest output energy of the laser pulse is 850 mJ with a circular beam diameter of 5 mm. Focusing lens was not used for the irradiation. To optimize the switching energy, we have fabricated a few laser amorphized marks on the surface of the absorber by using the various energy densities. These marks are made on the various regions by using the translation stage to shift the sample. The
Figure 2. a) Schematic illustration of the laser-induced reamorphization of multilayer FP cavity. b) The absorptance spectra of the phase change absorber with the four different states of AD-AM and CR (top panel), MQ-AM and R-CR (middle panel). In the bottom panel, the trajectory of movement of resonant wavelength is presented. c–f) The optical images of the absorber with the structural states of c) AD-AM, d) CR, e) MQ-AM, and f) R-CR (scale bar, 5 mm).

Figure 3. The VASE measurement of incident angular-dependent absorptance spectra for the phase change cavity residing on a SiO$_2$ substrate under the illumination of TM-polarized (left column) and TE-polarized (right column) incidence for a) AM and b) CR states. The Brewster modes for both of the structural states under TM-polarization are marked in red dots.
energy density of 208.9 mJ cm\(^{-2}\) is chosen to reamorphize the absorber (see Figure S3, Supporting Information). Note that, the melting temperature of Au is \(\approx 1336\) K\(^{[54,55]}\) that is much higher than the melting temperature of GST225 of \(T_m = 903\) K. This, together with the nanosecond heating/cooling processing, forbids the Au from melting. In Figure 2b, we experimentally realized a reversible tuning of peak absorptance for the NIR region using a reversible state transition of GST225 between the AM and CR. The absorptance spectra are measured in the different states of AD-AM, hot-plate annealed CR, laser-induced melt-quenched (MQ) AM, and hot-plate annealed recrystalline (R-CR). The spectra for the AD-AM and CR are presented in top panel, and in the middle panel the spectra for the MQ-AM and R-CR are shown. In the bottom panel, we mark trajectory of movement of resonant wavelength of peak absorptance as reversibly switching the state between AM and CR. As was seen, the resonant wavelength redshifts from \(\lambda_1 = 1180\) nm to \(\lambda_2 = 1680\) nm by changing the state from AD-AM to CR. The crystallized structure is then reamorphized to the MQ-AM phase, which blueshifts the resonant wavelength from \(\lambda_2 = 1680\) nm to \(\lambda_3 = 1280\) nm. The MQ structure was subsequently recrystallized to shift the resonant wavelength from \(\lambda_3 = 1280\) nm to \(\lambda_4 = 1680\) nm. The alternations in the position of resonant wavelength indicate reversible shifts, corresponding to reamorphization and recrystallization of the GST225 films, respectively. In Figure 2c–f, we present the microscope pictures of the absorber with the four different phases, accordingly. The CR absorber (Figure 2d) is brighter than the AM one (Figure 2c) owing to the lower absorptance. As melt-quenching the CR absorber, the structure changes back to a dark color (circled by dotted line in Figure 2e) due to an increasing absorptance. As shown in Figure 2f, the recrystallized structure exhibits a similar color with the crystallized one in the first writing. Although the resonance wavelength of the CR state (\(\lambda_2 = 1680\) nm) well matches the R-CR state (\(\lambda_4 = 1680\) nm), a difference takes place in the resonant wavelengths between the AD-AM (\(\lambda_1 = 1180\) nm) and MQ-AM (\(\lambda_3 = 1280\) nm) phases. It is because the crystallization kinetic of the AD-AM state is different with the MQ-AM state.\(^{[56]}\) The numbers of cyclability of crystallization–reamorphization are \(10^5\) and \(10^7\) for optical and electrical data storages, accordingly.\(^{[57]}\) However, we anticipate that phase-change switching cyclabilities in our multilayer absorber are not high. It is because the material fatigue can result from interdiffusion of films during repeated melt/quench cyclabilities.\(^{[58,59]}\) The reamorphization

![Figure 4](image-url)
of GST225 may not be possible after several crystallization–reamorphization cycles, owing to effect of metal induced crystallization. To solve the problem, for future study, an optimization of laser pulse energy density and pulse repetition rate for crystallization are useful for achieving a large number of switching cycles. Moreover, an ultrathin buffer layer can be introduced that may increase the device lifetimes and offer stable operations across many phase-change switching cycleabilities.

In Figure 3, we experimentally demonstrate the absorptance spectra as increasing the incident angle ($\theta_i$) from 20° to 80° with a step of 2°, where (a) and (b) present the absorptance for the AM and CR phases, respectively, under both the TM-incident (left column) and TE-incident (right column) polarizations. The cavity absorber possesses maximum absorptance for the different polarizations, particularly for TM-polarization at the larger $\theta_i$ and for TE-polarization at the smaller $\theta_i$, which is numerically validated (see Figure S4, Supporting Information). As the near-perfect absorptance is based on critical coupling, the cavity is polarization independent only under the smaller $\theta_i$, and exhibits the maximum absorptance only for TM-polarization over the larger $\theta_i$ that indicates the excitation of Brewster mode.$^{[5,60]}$ As was observed, the absorptance spectra can be redshifted as transiting the state from AM to CR. Particularly, we obtain the maximum absorptance for the AM state at the Brewster mode of $\lambda_{ba} = 820$ nm and Brewster angle of 78° (marked by red dot in the left column of Figure 3a); such a Brewster mode can redshift to $\lambda_{bc} = 1500$ nm while maintaining the Brewster angle at 70° after the crystallization of the structure (marked by red dot in the left column of Figure 3b).

In Figure 4, we have further investigated the effect of Brewster mode in the phase change cavity. In Figure 4a, for the AM state the VASE was used to measure the TM-polarized (blue line) and TE-polarized (blue dashed line) absorptance as a function of $\theta_i$, corresponding to the Brewster mode of $\lambda_{ba} = 820$ nm. It shows that the near-perfect absorptance can be achieved for the TM-polarization at the Brewster angle of $\theta_i = 78^\circ$. We then measure

![Figure 5](image_url)

**Figure 5.** a) A scheme of a flow microchannel-based biosensor by integrating the microfluidic system with the multilayered phase change absorber. b,d) The FDTD simulated spectra of incident angle-dependent GH shift b) for the AM absorber at Brewster mode of $\lambda_{ba} = 820$ nm by varying the concentration of copper sulfate solution, and d) for the CR absorber at Brewster mode of $\lambda_{bc} = 1500$ nm by varying the concentration of glucose solution. c,e) The FDTD simulated GH shift change with increasing RI of c) copper sulfate solution for the AM state and e) glucose solution for the CR state.
the phase difference ($\Delta \varphi$) between the TE- and TM-polarized light (red line), and a phase shift of $\approx \pi$ is realized around $\theta_i = 78^\circ$. In Figure 4b, we numerically simulate the spectra that have a good agreement with the measured ones. In Figure 4c,d, we both experimentally and numerically present the incident angle-dependent spectra of absorbance and $\Delta \varphi$ of the CR absorber, respectively. As was seen, the $\Delta \varphi$ around 70° varies sharply ($\approx \pi$) at $\lambda_{bc} = 1500$ nm. As the value of the GH shift is determined by the phase derivative at the Brewster angle, the Brewster mode can be used to enhance the GH shift.\(^{[61]}\) Herein, our proposed Au–GST225 stacked-layer absorber presents a reversible tuning of the enhanced GH shift by exciting the Brewster modes for both structural states. In Figure 4e,f, we numerically show the spectra of incident angle-dependent GH shift for both AM state at $\lambda_{ba} = 820$ nm and CR state at $\lambda_{bc} = 1500$ nm, respectively. We derive the GH shift from $l = -\frac{1}{\lambda} \frac{d\varphi}{d\theta}$, where $k$ is the wavevector of the incident medium. As was seen, for both the AM and CR states, the largest GH shifts are obtained at the Brewster angle in which a maximum phase variation ($\Delta \varphi$) occurs. The GH shift can be tuned by changing the structural state of GST225 in the multilayer absorber.

To illustrate the potential of our absorber working as a RI sensor for label-free multichannel biosensing applications. Figure 5 shows the proof-of-principle simulation, displaying the GH shift with the solutions of copper sulfate and glucose on the absorber surface. The rapid phase variation around the Brewster angle can be used to monitor slight alternation in the electromagnetic (EM) environment of the multilayer absorber, offering a label-free sensing platform. In Figure 5a, we schematically present a flow microchannel-based biosensor by integrating the microfluidic system on top of the phase change absorber. In particular, by switching the structural state between AM and CR, the Brewster mode can be tuned to measure the different kinds of biosamples. To detect multiple target biosamples, the spectral absorbance of the multilayer cavity needs to be designed to match the characteristic absorbance bands of various target biomolecules in the NIR region.\(^{[62]}\) We selected copper sulfate and glucose as the two target samples. The characteristic absorbance wavelengths of the two kinds of biosample solutions are 800 nm (copper sulfate) and 1650 nm (glucose), respectively. Correspondingly, the Brewster modes of the cavity absorber are tuned to be $\lambda_{ba} = 820$ nm (AM state) and $\lambda_{bc} = 1500$ nm (CR state), respectively. In Figure 5b,c, we first conducted the detection of concentration variation of copper sulfate by using the AM absorber. Figure 5b shows the simulated GH shift spectra of the detector filled with aqueous solutions of copper sulfate with the different concentrations. There is a large GH shift around the Brewster angle. The zoomed-in picture clearly presents that the GH shift can be changed at 78° by changing the concentration of copper sulfate, owing to the RI change in copper sulfate solution. In Figure 5c, we present the variation of GH shift with RI change in copper sulfate solution. It demonstrates that the change in GH shift changes linearly with the increasing of analyte RI. In Figure 5d, we then show the GH shift spectra associated with glucose solution at $\lambda_{bc} = 1500$ nm for the CR state, and the change in GH shift also linearly depends on the RI change in glucose solution, as shown in Figure 5e.

3. Conclusions

In summary, we present a versatile platform for creating a reversible tuning of high light absorption across the NIR regime. The exceptional presentation of the absorber is obtained by using an asymmetric FP cavity composed of air/GST225–Au stacks/SiO$_2$ substrate. We show that the peak absorbing wavelength can be redshifted from 1180 to 1680 nm as switching the GST225 state from AM to CR, and a backward tuning of peak absorption can be experimentally realized in just 5 ns by melt-quenching the CR GST225. We also experimentally demonstrate that the Brewster mode can be excited in the multilayer FP cavity under the TM-polarized incidence, and such a Brewster mode can be tuned to cover the full NIR spectrum via the phase transition of GST225. We further numerically illustrate that the label-free multichannel biosensing is possible by using our phase change absorber. We design the multilayer absorber whose Brewster modes for both AM and CR states are around the characteristic absorbance wavelengths of two target biomolecules: copper sulfate and glucose solutions, respectively. The numerically model can predict the GH shift of the two Brewster modes channel based upon the concentrations of two target biosamples. Our proposed structure is lithography-free that possesses a main advantage over microfluidics integrated nanophotonics sensor.

4. Experimental Section

Device Fabrication: The physical vapor deposition method was used to fabricate the planar multilayered structure. We deposited the GST225 layers by radio frequency (RF) sputtering and Au films by direct current (DC) sputtering. Atomic force microscope (AFM) was used to calibrate the deposition rates. The silica was chosen as the substrate. The cross-sectional picture of the planar multilayered absorber was obtained by the FIB (Helios C4 UX) system.

Optical Characterization: Variable angle spectroscopic ellipsometry (J. A. Woollam Co., Inc., V-VASE) was used to optically characterize the spectra of multilayer FP in the spectral range of 400–2400 nm with a spectroscopic resolution of 10 nm. The angle- and polarization-dependent spectra of the multilayer structures were measured as varying the incident angle from 20° to 80° at room temperature. The spectra were recorded with 2° angular resolution.

Numerical Simulation: We used a commercial FDTD simulation software package (Lumerical Solutions, version 8.15.736) to simulate the multilayer FP cavity absorber. The simulation was conducted in a 3D layout. We used periodic boundary conditions in the $x$-$y$ plane and perfectly matched layers along the $z$-direction. The reflection and the transmittance spectra were recorded by the power monitors positioned above and below the multilayered absorber, respectively. We used Bloch boundary condition for normal incidence while a broadband fixed angle source technique (BFAST) for oblique incidence. In the model, the complex RIs of GST225 were measured using an infrared variable angle spectroscopic ellipsometer. A Tauc–Lorentz or Lorentz–Lorentz model can be used for the fitting.\(^{[63]}\)

Supporting Information

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

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Conflict of Interest
The authors declare no conflict of interest.

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Research data are not shared.

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