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# **Dynamic color generation with electrically tunable thin film optical coatings**

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## **ABSTRACT**

Thin film optical coatings have wide range of industrial applications from displays and lighting to photovoltaic cells. The realization of electrically tunable thin film optical coatings in the visible wavelength range is particularly important to develop energy efficient and dynamic color filters. Here, we experimentally demonstrate dynamic color generation using electrically tunable thin film optical coatings that consist of two different phase change materials (PCMs). The proposed active thin film nanocavity excites Fano resonance that results from the coupling of a broadband and a narrowband absorber made up of phase change materials. The Fano resonance is then electrically tuned by structural phase switching of PCM layers to demonstrate active color filters covering the entire visible spectrum. In contrast to existing thin film optical coatings, the developed electrically tunable PCM-based Fano resonant thin optical coatings have several advantages in tunable displays and active nanophotonic applications.

**KEYWORDS:** Thin film optical coatings, Electrical control of colors, Fano resonance, Phase change materials, Tunable optical structures, Color filters, Microheaters.

Dynamically tuning the optical properties of nanophotonic and plasmonic systems in the visible wavelength range is particularly important to develop reconfigurable optical components such as color filters for display and lighting applications<sup>1</sup>. In particular, the optical and plasmonic responses of these systems can be dynamically tuned by inducing a controllable change in the optical properties via an external stimulus<sup>2</sup>. Since the dynamic color generation requires two essential parts such as the coloration mechanism and the coloration control<sup>3</sup>, it can be achieved by integrating functional materials with nanophotonic and plasmonic systems. To date, various functional materials<sup>4-7</sup> including electrically tunable materials such as liquid crystals<sup>8, 9</sup>, electrochromic polymers<sup>10, 11</sup>, and phase change materials<sup>12, 13</sup> (PCMs) have been used to realize dynamic color generation. Among these, phase change materials based on chalcogenide semiconductor alloys have received great attention to tune the optical properties of nanophotonic systems in the ultraviolet to terahertz frequencies<sup>14-17</sup>, due to their substantial refractive index change when switched between two structural states. Chalcogenide PCMs provide several advantages because of their ability of reliable and repeatable non-volatile switching mechanism. This is important to develop dynamic color filters with non-volatile pixels as the pixels remain in the desired state after the external stimulus is removed. However, in most of the reported PCM based active optical systems, the active modulation was realized by utilizing bulky thermal annealing process and expensive optical writing techniques<sup>18</sup>. We note that electrically reconfigurable PCM-based nanophotonic components are particularly important for compact integration with flat optics and chip-scale reconfigurable photonic device applications, thus this field of research has recently received significant attention<sup>19-23</sup>.

In recent years, thin film optical coating based light absorbers have been used for numerous applications including structural coloring as they are scalable and lithographically-free method to

create cost-effective thin film optical devices<sup>24</sup>. In these systems, perfect absorption take place when the light is critically coupled to an absorber. The critical light coupling in thin films can be realized by suppressing reflection via thin film interference. Asymmetric Fabry–Perot metal–dielectric–metal (MDM) cavity is a well-studied thin film optical coating; wherein critical light coupling takes place due to amplitude splitting destructive interference such that light is entirely trapped inside the resonator and is dissipated due to the existence of losses<sup>24</sup>. Thus, different MDM cavities based optical coatings have widely been used for structural coloring applications<sup>25-28</sup>. More importantly, dynamic color filters have been developed using PCM-based tunable MDM cavities<sup>29-36</sup>. However, there are several drawbacks in color filters based on MDM optical coatings: (i) due to the existence of a top-metal layer, it can easily suffer from scratching and oxidation which will deteriorate the formed colors rapidly, (ii) most of the MDM cavities are iridescent, that is, polarization and incidence angle dependent, and (iii) MDM cavities provide different colors in the reflection and transmission mode.

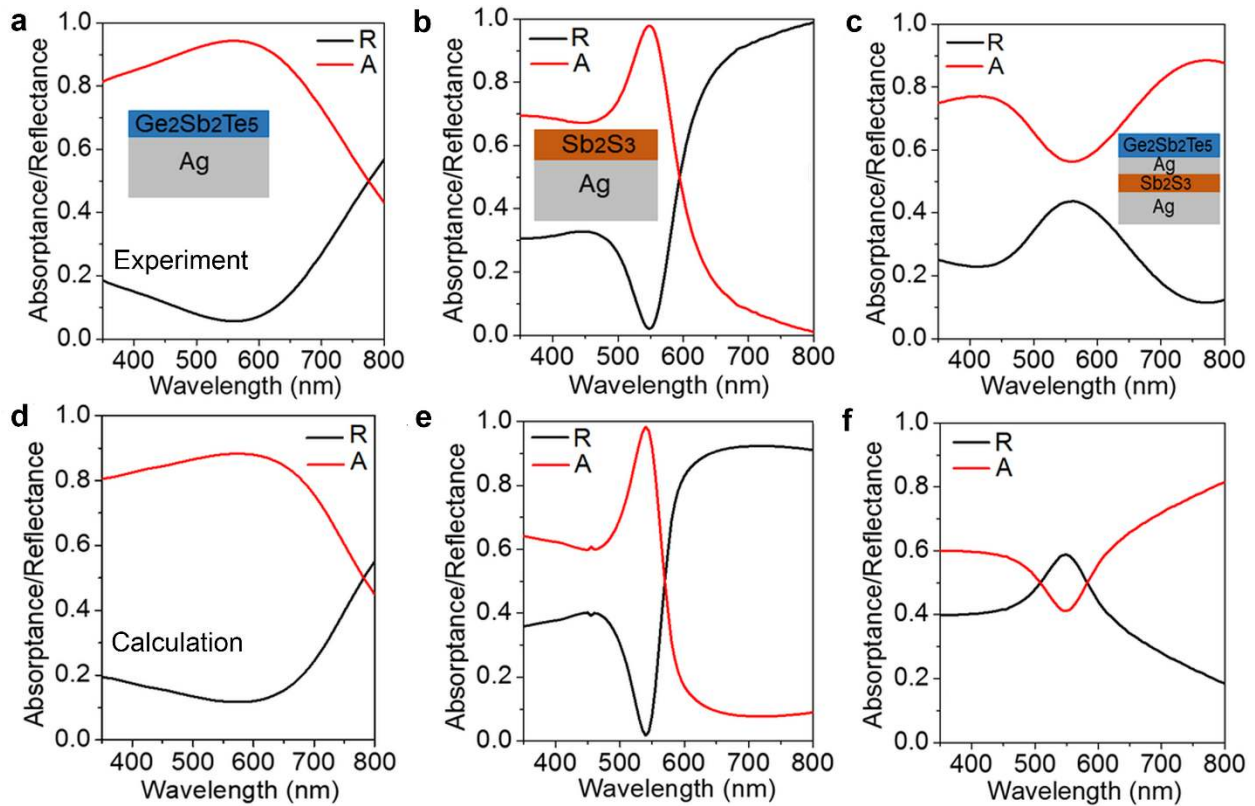
In this work, we propose and experimentally demonstrate dynamic color generation using active PCM-based thin film optical coatings. The proposed multilayered thin film optical coating consists of a broadband and a narrowband absorber with a lossy and lossless PCM, respectively. In comparison to conventional Fabry–Perot MDM cavities, this cavity exhibits Fano resonance due to the coupling between the broadband and narrowband absorbers. By switching the structural states of PCM layers in the cavity from amorphous to crystalline, the Fano resonance supported by the cavity is tuned in wavelength and bandwidth, as a result wide color tunability is realized. More importantly, electrically forward continuous tuning of colors at low DC current is realized by integrating the thin optical coatings with a microheater device. As an advantage, these coatings provide wide spectral tunability in the visible wavelength range with same color in the reflection

and transmission mode, and they are iridescence-free. Since the top layer of the optical coating is not a metal, the scratching and oxidation issues can be overcome to a large extent.

## Results and Discussion

In general, Fano resonance can be realized in nanophotonic systems by coupling the resonators having contrasting resonance linewidths<sup>37,38</sup>. Interestingly, Mohamed et al. recently demonstrated Fano resonance in thin film optical coatings by coupling a broadband (representing the continuum) and a narrowband (representing the discrete state) absorber<sup>39</sup>. However, the reported thin film coating is not tunable. Here, we introduce an active Fano resonant thin film coating for dynamic color generation and tuning by coupling PCM based broad- and narrow-band absorbers. Since  $\text{Ge}_2\text{Sb}_2\text{Te}_5$  (GST) is a lossy PCM in the visible wavelengths, we fabricated tunable broadband absorber by depositing a 20 nm of GST thin film on a 100 nm Ag film (Fig. 1a). The measured reflection and absorption spectrum of a GST-Ag stack at normal incidence is shown in Fig. 1a, which has a broad absorption bandwidth. We used  $\text{Sb}_2\text{S}_3$  PCM to fabricate tunable narrowband absorber because  $\text{Sb}_2\text{S}_3$  is a low-loss and high refractive index ( $>3$ ) PCM in the visible wavelengths. We realized narrowband absorber by depositing a 90 nm of  $\text{Sb}_2\text{S}_3$  thin film on a 100 nm Ag film (Fig. 1b). Figure 1b shows the measured reflection and absorption spectra of a  $\text{Sb}_2\text{S}_3$ -Ag stack at normal incidence that shows a narrow absorption bandwidth. The GST-Ag stack (broadband absorber) provides a nearly constant phase that represents the continuum, and  $\text{Sb}_2\text{S}_3$ -Ag stack (narrowband absorber) provides a steep phase change at the resonance (see Supporting Information Fig. S1). We used transfer matrix method<sup>40</sup> (TMM) to calculate the reflection spectrum and the phase change. In our calculations, experimentally determined optical constants of GST,  $\text{Sb}_2\text{S}_3$  and Ag thin films were used (see Supporting Information Fig. S2). In Figure 1c, we show the measured reflection and absorption spectra of a GST (20 nm)-Ag (20 nm)- $\text{Sb}_2\text{S}_3$  (30 nm)-

Ag (100 nm) stack at normal incidence. In comparison to GST-Ag and  $\text{Sb}_2\text{S}_3$ -Ag stacks, the combined GST-Ag- $\text{Sb}_2\text{S}_3$ -Ag stack results in destructive interference between the constant and steep varying phase of the overlapping broadband and the narrow-band cavities that lead to Fano type sharp resonance with asymmetric line shape. These asymmetric line shapes are then fitted using a Fano formula (see Supporting Information Fig. S3). In Figure 1d, 1e and 1f, we report the calculated reflection and absorption spectra of GST-Ag,  $\text{Sb}_2\text{S}_3$ -Ag, and GST-Ag- $\text{Sb}_2\text{S}_3$ -Ag stacks, respectively. As can be seen, the calculated and experimental results are in good agreement.



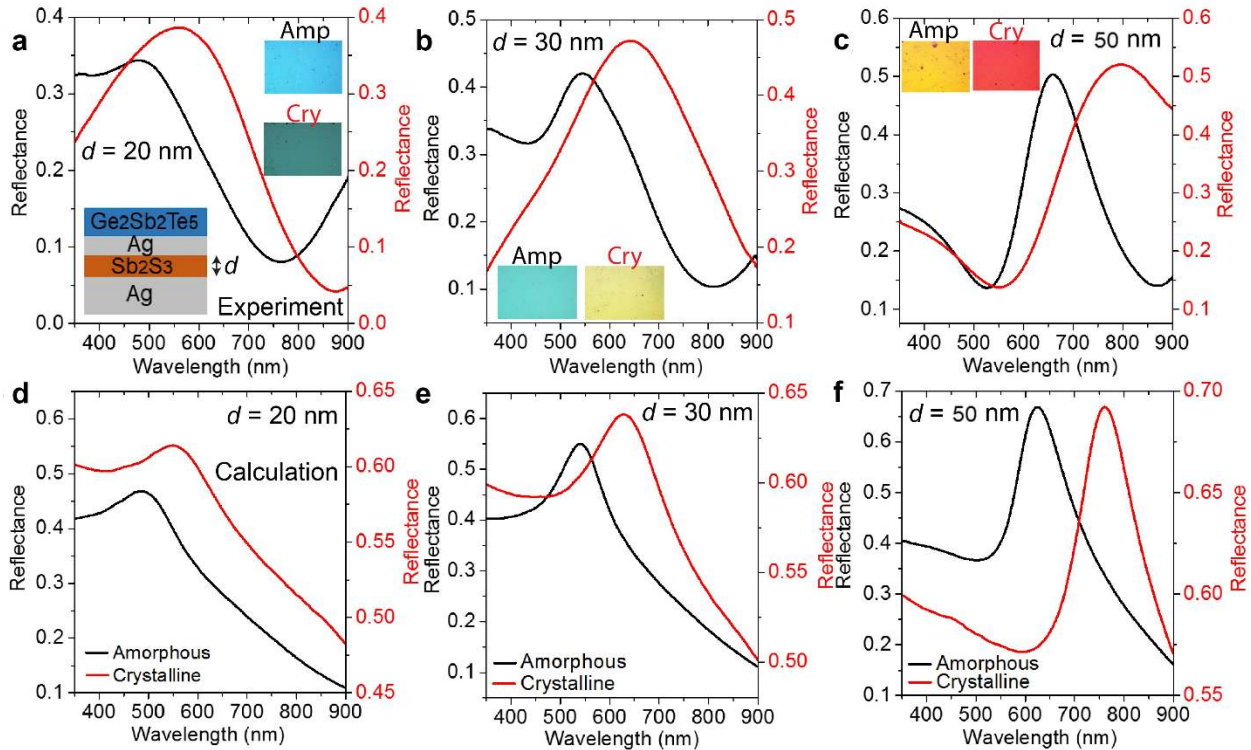
**Figure 1 Demonstration of Fano resonance in active nanophotonic cavities.** Measured reflectance and absorptance spectra of an active (a) broadband absorber (GST-Ag), (b) narrowband absorber ( $\text{Sb}_2\text{S}_3$ -Ag) and (c) Fano resonant thin film coating (GST-Ag- $\text{Sb}_2\text{S}_3$ -Ag). Calculated reflectance and absorptance spectra of an active (d) broadband absorber (GST-Ag), (e) narrowband absorber ( $\text{Sb}_2\text{S}_3$ -Ag) and (f) Fano resonant thin film coating (GST-Ag- $\text{Sb}_2\text{S}_3$ -Ag).

An interesting feature of the Fano resonant thin film optical coating is that it reflects and transmits the same color<sup>39</sup>. The reflectance and transmission spectra of a semi-transparent active thin film coating (GST (20 nm)-Ag (15 nm)-Sb<sub>2</sub>S<sub>3</sub> (50 nm)-Ag (30 nm)) is shown in Supporting Information Fig. S4, where the peak transmission and reflection wavelengths overlap. We note that the thickness of top GST layer is important to realize Fano resonance. The resonance of the cavity switches from Fano to conventional MDM at a critical thickness of GST layer, which is >40 nm (see Supporting Information Fig. S5). Since Sb<sub>2</sub>S<sub>3</sub> is the cavity layer of the coating, the Fano resonance is exactly realized at the resonance wavelength of the narrowband absorber (Sb<sub>2</sub>S<sub>3</sub>-Ag stack), thus the Fano resonance linewidth and resonance wavelength can be tuned by changing the thickness of Sb<sub>2</sub>S<sub>3</sub> layer (see Supporting Information Fig. S6). It should be noted that the developed thin film coating is independent of incidence angle ( $\pm 80^\circ$ ) and polarization (p- and s-polarization) due to the high refractive index of Sb<sub>2</sub>S<sub>3</sub> layer (see Supporting Information Fig. S7). Thus, the proposed thin film coating is highly suitable for structural color applications overcoming the major issue of iridescence in the existing color filters.

We demonstrate wide color tunability from blue to red by slightly changing the thickness of Sb<sub>2</sub>S<sub>3</sub> thin layer and switching the structural phase of PCM layers from amorphous to crystalline. We note that the crystallization temperature of GST and Sb<sub>2</sub>S<sub>3</sub> is entirely different, that is,  $\geq 150^\circ\text{C}$  for GST and  $\geq 280^\circ\text{C}$  for Sb<sub>2</sub>S<sub>3</sub>. Therefore, in our thermal switching experiments, we annealed the thin film samples at a moderate temperature of 220 °C to avoid the possible heat induced damage of GST thin film. It indicates that Sb<sub>2</sub>S<sub>3</sub> layer could be partially crystallized by annealing at 220 °C. In Figure 2a-c, we show the measured reflectance spectrum of thin film coating for different thicknesses (20-50 nm) of Sb<sub>2</sub>S<sub>3</sub> cavity layer and both phases (amorphous and crystalline) of the thin layers of GST and Sb<sub>2</sub>S<sub>3</sub>. As can be seen, the Fano resonance peak wavelength red shifts with



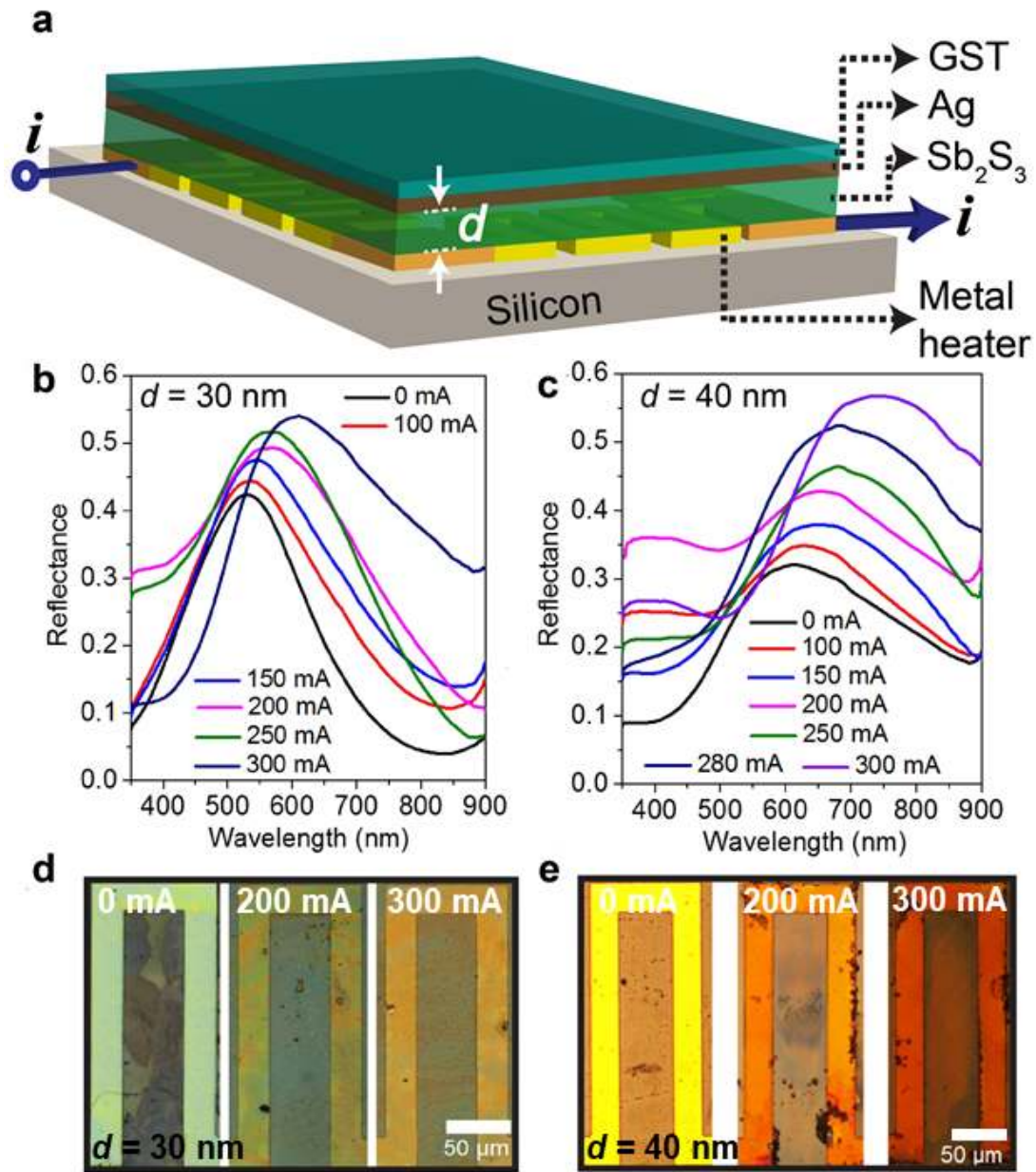
increasing the thickness of  $\text{Sb}_2\text{S}_3$  layer as well as by switching the phase of PCMs, as a result wide color tunability from blue to red is realized. The spectral wavelength shifts obtained for the  $\text{Sb}_2\text{S}_3$  thickness of 20 nm, 30 nm and 50 nm are 80 nm, 100 nm, and 140 nm, respectively, as the phase changes from amorphous to crystalline form. These values are considered to be substantial in the visible wavelengths. In Fig. 2d-f, we show the corresponding calculated results, and the results agree very well with the experiment. We also simulated the intensity field distribution at the resonance wavelength for different thicknesses of  $\text{Sb}_2\text{S}_3$  cavity layer and confirmed that the field is tightly confined inside  $\text{Sb}_2\text{S}_3$  layer for both phases of PCMs (see Supporting Information Fig. S8). Moreover, we realized the same spectral shifts with color tunability by replacing the GST layer with another PCM layer such as  $\text{Sb}_2\text{Te}_3$ , however, smaller wavelength shift is obtained by replacing the GST layer with a Ge layer (see Supporting Information Fig. S9). It shows the importance of two PCMs in the thin film coating for dynamic color generation.



**Figure 2 Demonstration of tunable Fano resonance with color change.** Measured reflectance spectrum of Fano resonant thin film coating for amorphous and crystalline phases of PCM layers with different thicknesses of  $\text{Sb}_2\text{S}_3$  layer (a) 20 nm, (b) 30 nm and (c) 50 nm. Calculated reflectance spectrum of Fano resonant thin film coating for amorphous and crystalline phases of PCM layers with different thickness of  $\text{Sb}_2\text{S}_3$  layer (d) 20 nm, (e) 30 nm and (f) 50 nm.

Since electrically tunable devices are required for chip scale reconfigurable photonic device applications, we demonstrate electrically continuous forward (amorphous to crystalline) tuning of colors using microheater-integrated thin film optical coating devices (Fig. 3a). In particular, the whole sample is annealed continuously by electric current-induced heating through the Joule heating mechanism. We used optical lithography followed by metal deposition to fabricate microheaters (see Supporting Information Fig. S10a). The remaining three layers ( $\text{Sb}_2\text{S}_3$ , Ag and GST) of thin film coatings were directly deposited on the whole sample. That means, only the heater elements represent the Fano resonant optical coatings. Initially, we performed the temperature calibration of the microheater-integrated device and observed a linear variation of temperature (room temperature to 220°C) with applied DC current (0 to 325 mA). In Fig 3b and Fig 3c, we show the electrical forward tuning of Fano resonance of thin film coating with 30 nm and 40 nm thick  $\text{Sb}_2\text{S}_3$  layer, respectively. The continuous tuning of Fano resonance towards higher wavelength with increasing current (0 to 300 mA) is obtained. In Supporting Information Fig. S11, we plot the peak resonance wavelength with increasing current and the corresponding calculated wavelength shift (with respect to 0 mA), which shows a non-linear behavior. The corresponding color change for  $\text{Sb}_2\text{S}_3$  layer thickness of 30 nm (green to yellow) and 40 nm (yellow to red) is shown in Fig 3d and Fig 3e, respectively. A large area color tunability (pink to greenish yellow) of thin film coating with 15 nm thick  $\text{Sb}_2\text{S}_3$  layer is shown in Supporting

Information Fig. S12, where whole parts of the sample represent Fano resonant optical coatings. As can be seen, a DC current of  $\geq 200$  mA is required to observe the considerable color change, however, the current or voltage required to switch the phase of the PCMs in the forward direction can be further reduced by using longer electrical current pulses<sup>21-23</sup> (msec to  $\mu$ sec). Since the reversible switching of color is important for practical applications, it can be realized with bowtie type heater designs with shorter high energy electrical current pulses<sup>21-23</sup> ( $\mu$ sec to nsec), since the phase switching from crystalline to amorphous phase can only be achieved with transient melting followed by rapid cooling. The oxidation issue with chalcogenide PCMs (GST and  $\text{Sb}_2\text{Te}_3$ ) can be overcome by capping the top PCM layer with a thin dielectric such as  $\text{SiO}_2$  (Supporting Information Fig. S13). Since the switching speed of PCM based device depends on the switching area, the switching speed of the proposed thin film coating is of the order of hundreds of seconds. In short, the proposed active Fano resonant thin film coating is an ideal scalable nanophotonic system for dynamic structural coloring applications. Moreover, the active Fano resonant coating can be used for other potential applications including imaging, nonlinear optics, superlenses and sensing because the stacked ultra-thin films of dielectrics, semiconductors, and metals are considered as an alternative platform for metasurfaces, which performs unique or similar functionalities<sup>41-44</sup>.



**Figure 3 Demonstration of electrically continuous tunable Fano resonance with color change.**

(a) Schematic of fabricated microheater integrated Fano resonant thin film optical coating device.

Measured tunable reflectance spectra of thin film coatings with applied current (0- 300 mA) for different thicknesses of  $\text{Sb}_2\text{S}_3$  layer (b) 30 nm and (c) 40 nm. Optical microscopic image of color change observed on heater element with applied current for different thicknesses of  $\text{Sb}_2\text{S}_3$  layer

(d) 30 nm and (e) 40 nm.

## **Conclusion**

We experimentally demonstrated electrically tunable dynamic color generation using scalable active thin film optical coatings. In particular, tunable Fano resonance was realized in PCM-based thin optical coatings by the destructive interference between the resonance of a broad- and narrow-band absorber. We demonstrated wide color tunability covering whole visible spectrum by slightly changing the thickness of the cavity layer and switching the structural phase of PCM layers in the optical coatings from amorphous to crystalline. Moreover, we showed that Fano resonance with color can be electrically tuned in the forward direction using a microheater-integrated device at low DC currents. Electrically reconfigurable color generation is important for future applications of displays and lighting, which can be realized using heaters based on bowtie structures and high energy pulsed electrical excitation<sup>21-23</sup>. More importantly, we addressed the limitations of the existing thin film optical coating-based color filters such that the proposed active optical coating provides wide color tunability with same color in reflection and transmission mode, and it is iridescence-free and oxidation-resistant.

## **ASSOCIATED CONTENT**

### **Supporting Information**

Sample fabrication and characterization; spectroscopy ellipsometry characterizations; additional experimental and simulation results.

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### **Notes**

The authors declare no competing financial interests.

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