

# Electrically Reconfigurable Plasmonic Metasurfaces Based on Phase-Change Materials $\text{Sb}_2\text{S}_3$

Zhuoxuan Han, Chensheng Li, Tengzhang Liu, Nannan Hu, Zhiqin Fan, Yang Guo, Baoli Liu, Haifang Yang, Aizi Jin, Baogang Quan, Shibing Tian, Yang Yang, Geng Li, Xiaofeng Fan, Sha Hu,\* Xin Huang,\* and Changzhi Gu\*



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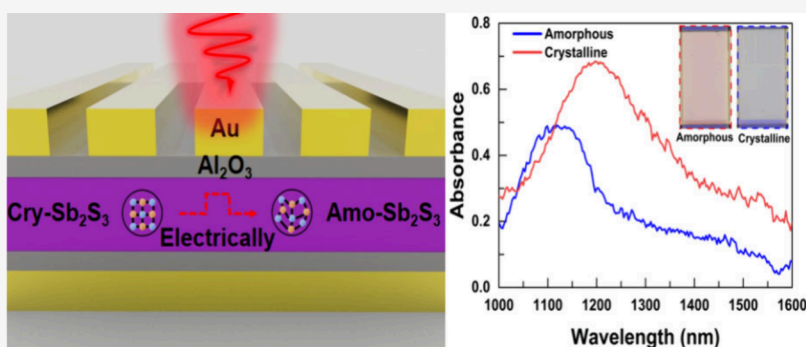
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**ABSTRACT:** Phase-change materials (PCMs) are widely used in active optical metasurfaces due to their large refractive index contrast and fast and stable phase-change properties. In this paper, an electrically reconfigurable plasmonic metasurface based on the PCM  $\text{Sb}_2\text{S}_3$  is proposed to achieve nonvolatile, reversible, and fast optical modulation in the near-infrared range. The designed metasurface can redshift the surface plasmon resonance peak from 1320 to 1480 nm through the phase transition of  $\text{Sb}_2\text{S}_3$  from amorphous to crystalline states. In addition, we further experimentally design an electrically reconfigurable platform. In a  $30\ \mu\text{m} \times 30\ \mu\text{m}$  region, the phase state of  $\text{Sb}_2\text{S}_3$  with a thickness of 60 nm is successfully and reversibly changed, which contributes to the dynamic modulation of gold gratings. This work has great application potential in reconfigurable optical filters and communication systems and adaptive optical imaging and sensing.

**KEYWORDS:** metasurface, electrical tuning, reversible switching, phase-change material

Metamaterials are artificial materials whose dimensions are smaller than the wavelength of external excitation. Unlike natural materials, whose electromagnetic properties are determined by the specific characteristics of their atoms or molecules, metamaterials can be artificially tailored to engineer desired electromagnetic responses.<sup>1</sup> Therefore, metamaterials can flexibly manipulate the properties of electromagnetic waves such as amplitude, polarization, and phase. Subsequently, extraordinary characteristics not found in natural materials can be achieved by metasurfaces, such as abnormal deflection,<sup>2,3</sup> metalenses,<sup>4,5</sup> and perfect absorption.<sup>6,7</sup> As a two-dimensional metamaterial, metasurfaces can not only flexibly manipulate electromagnetic wave characteristics<sup>8,9</sup> but also have the advantages of flexible design, small size, and easy integration compared with metamaterials.

Once most metasurfaces are fabricated, their functions will be fixed and lack dynamic tuning, which limits their breakthrough in practical applications. Active metasurfaces can dynamically modulate their optical responses, providing exciting opportunities for flexible manipulation of light

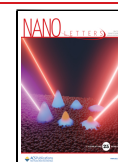
propagation.<sup>10,11</sup> So far, the active tuning of metasurfaces has been utilized through mechanical deformation,<sup>12–14</sup> electrochemical or chemical reaction,<sup>15</sup> electrical regulation,<sup>16–18</sup> and all-optical regulation.<sup>19</sup> In addition, liquid-crystal materials<sup>20</sup> and phase-change materials (PCMs)<sup>21–24</sup> are often used in active metasurfaces. In particular, PCMs have unique conditions for achieving active modulation due to the significant refractive index contrast brought about by their structural transitions. PCMs have been widely applied in reconfigurable photonic platforms, extending from programmable photonics, neuromorphic computing, and nonvolatile and rewritable data storage to tunable metasurfaces. For PCM-

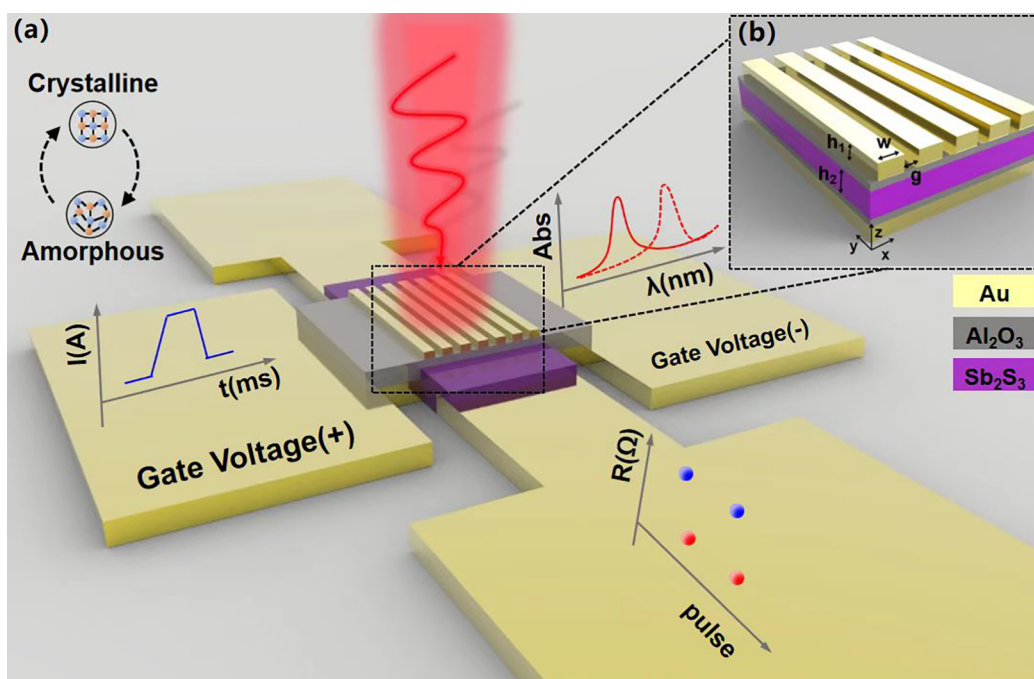
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**Figure 1.** Operation and optical response of an electrically tunable  $\text{Sb}_2\text{S}_3$ -based metasurface. (a) Schematic diagram of the electrothermal metasurface. The tunable metasurface makes the  $\text{Sb}_2\text{S}_3$  phase change by applying a voltage and shows a dynamic modulation of the plasmon resonance in the near-infrared band. The corresponding geometric parameters are shown in part b.

based metasurfaces, dynamic switching can be realized between amorphous and crystalline states by thermal annealing or laser pulse excitation. Additionally, the electrical switching of PCMs naturally favors compact integration with planar optics and anticipates miniaturization, leading to chip-level reconfigurable optical systems.

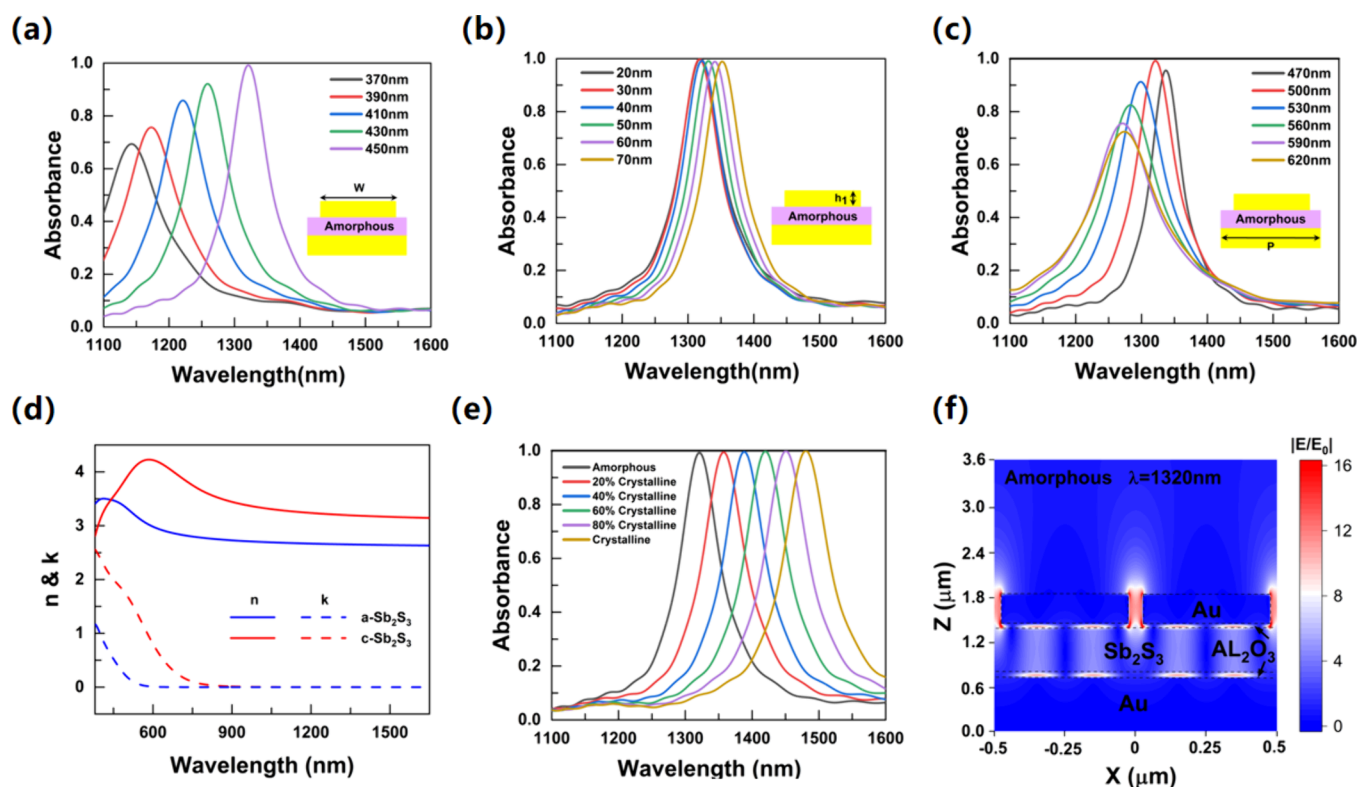
$\text{Ge}_2\text{Sb}_2\text{Te}_5$  (GST), one of the most common PCMs, has a large refractive index contrast in the near-infrared band. GST-based metasurfaces have a wide range of applications in holographic imaging,<sup>25,26</sup> perfect absorption,<sup>27</sup> and other fields. However, the crystalline GST has a large loss, which also limits its performance in the field of photoelectric detection. Antimony sulfide ( $\text{Sb}_2\text{S}_3$ ) has recently been classified as a PCM.<sup>28–30</sup> Compared with GST,  $\text{Sb}_2\text{S}_3$  has almost zero loss in the near-infrared band. This type of material can be stimulated between its amorphous and crystalline phases through thermal or laser irradiation, requiring modulation of its refractive index in an ultrashort time (on the order of picoseconds).<sup>31,32</sup> However, both methods require bulky external equipment, namely, heating stages or ultrafast lasers. Therefore, electric drive regulation becomes the best choice for the active regulation of PCM metasurfaces. Compared with the electric regulation of other materials,<sup>33,34</sup> PCMs have the advantages of fast response speed. Currently, there have been reports of electrical-driven control based on GST.<sup>35,36</sup> However, the electrical control of  $\text{Sb}_2\text{S}_3$  still poses challenges because the crystallization temperature of  $\text{Sb}_2\text{S}_3$  is around 270 °C, which is much higher than that of GST. Additionally, high temperatures can cause thermal deformation of the surrounding nanostructures. Nevertheless, recent advancements have achieved electrothermal control of  $\text{Sb}_2\text{S}_3$  using microheaters.

In this work, we propose a metal–insulator–metal (MIM) structure that consists of a gold (Au) substrate, PCM  $\text{Sb}_2\text{S}_3$ , and a gold grating. For the proposed metasurface, the plasmon resonance of the surface grating is dynamically adjusted by the

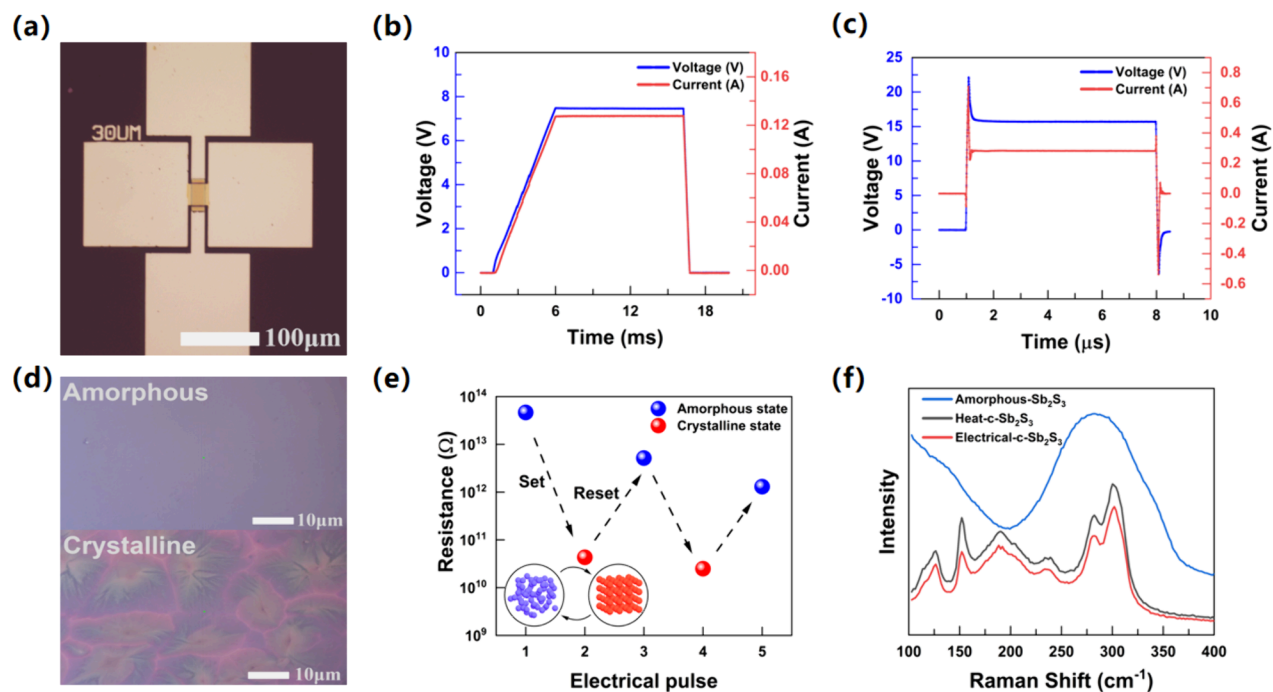
huge refractive index difference generated by the  $\text{Sb}_2\text{S}_3$  phase transition. Furthermore, we demonstrate on-demand optical modulation and wavefront engineering in a  $30\ \mu\text{m} \times 30\ \mu\text{m}$  area using electrical drive, achieving a fully reversible and reconfigurable  $\text{Sb}_2\text{S}_3$ -based metasurface. The design proposed in this paper can produce a spectral response that is more sensitive to incident polarization with potential applications in imaging, information transmission, and photoelectric detection.

An electrically tunable metasurface based on  $\text{Sb}_2\text{S}_3$  is demonstrated, as schematically shown in Figure 1a. The metasurface is composed of gold gratings and a microheater, which are separated by  $\text{Al}_2\text{O}_3$  and  $\text{Sb}_2\text{S}_3$  films. The gold grating controls the excitation and characteristics of the surface plasmon through varying specific structural parameters and environmental conditions. The microheater is composed of 5 nm of titanium and 60 nm of gold. The electrodes at the left and right ends are responsible for applying the voltage required for the phase transition of  $\text{Sb}_2\text{S}_3$ . The electrodes at the upper and lower ends are directly connected to  $\text{Sb}_2\text{S}_3$  to measure the resistance of  $\text{Sb}_2\text{S}_3$ . The entire microheater is mounted on a silica substrate, which effectively prevents heat loss. Under the conditions of external voltage pulse excitation, the  $\text{Sb}_2\text{S}_3$  film can be transformed between the amorphous phase and the crystalline phase. In addition, because  $\text{Sb}_2\text{S}_3$  is nonvolatile, crystalline and amorphous states can be activated and maintained. The phase transition of  $\text{Sb}_2\text{S}_3$  results in a great change of its dielectric constant changes, leading to a red shift of the resonance wavelength. The corresponding structural parameters of our proposed tunable metasurface are marked in Figure 1b, where  $h_1$  and  $h_2$  represent the height of the gold grating and  $\text{Sb}_2\text{S}_3$ , respectively,  $w$  is the width of the gold grating, and  $g$  is the spacing between the gratings.

The finite-difference time-domain (FDTD) method is used to numerically optimize the structure design. In this study, the



**Figure 2.** Simulated results of demonstrated metasurfaces under different structural parameters. Simulated absorbance spectra when the gold gratings have varied (a) widths, (b) periods, and (c) thicknesses. (d) Measured complex refractive indices for amorphous and crystallized films of  $\text{Sb}_2\text{S}_3$ . (e) Simulated absorbance spectra of the active phase-change metasurface with different crystallization fractions of  $\text{Sb}_2\text{S}_3$ . (f) Electric-field distribution of the metasurfaces in the a- $\text{Sb}_2\text{S}_3$  case at a wavelength of 1320 nm.



**Figure 3.** Electrically driven phase change of the  $\text{Sb}_2\text{S}_3$  layer based on a microheater. (a) Optical micrograph of the sample. The applied current and voltage of the (b) “set” and (c) “reset” pulses varying with time. (d) Optical micrograph of amorphous (up) and crystalline (down)  $\text{Sb}_2\text{S}_3$ . (e) Measured resistance of the  $\text{Sb}_2\text{S}_3$  film after each pulse, where blue dots represent the resistance with “reset” pulse and red dots denote the resistance with “set” pulse. The robust and reversible switching between the crystalline and amorphous phases is confirmed by resistance changes exceeding 1 order of magnitude. (f) Raman spectra of amorphous  $\text{Sb}_2\text{S}_3$ , high-temperature-annealed crystalline  $\text{Sb}_2\text{S}_3$ , and electrically controlled crystalline  $\text{Sb}_2\text{S}_3$ .

1300–1500 nm communication band, which is the most common working wavelength in integrated photonics, is chosen. Moreover, the incident light is polarized along the  $x$  direction. Parts a–c of Figure 2 display the simulated absorption spectra when the metasurface has varied structural parameters and is in the amorphous state of  $\text{Sb}_2\text{S}_3$ . The period was first fixed to 500 nm, with  $h_1 = 40$  nm and  $h_2 = 60$  nm, and the widths of the gold gratings varied from 370 to 450 nm. As shown in Figure 2a, when the width increases, the absorption peak becomes narrower and stronger, resulting in a higher absorbance. The absorbance reaches nearly one unit at  $w = 450$  nm, so the width is fixed at 450 nm. Under the same geometric conditions, the absorption spectra of crystalline  $\text{Sb}_2\text{S}_3$  with geometric parameter  $w$  are also discussed in Figure S1. It indicates that the results are consistent with those of the amorphous state, as shown. The influence of the thickness of the top gold nanoantenna in Figure 2b was also discussed ( $w = 450$  nm and  $g = 50$  nm). The corresponding full width at half-maximum and absorbance are detailed in Figure S1. The simulated spectra indicate that 40 nm is the optimal thickness. Figure 2c shows the absorption spectra under different periods. When the width of the gold grating is fixed at 450 nm,  $h_1 = 40$  nm, it can be clearly seen that the absorption spectra have an increasing trend by reducing the period. When the period is 500 nm, the absorbance reaches almost 1. When the period continues to decrease, the absorption spectra show a downward trend. Therefore, the period is fixed at 500 nm. Through structural optimization, the parameters of the metasurface were finally determined to be  $h_1 = 40$  nm,  $h_2 = 60$  nm,  $w = 450$  nm, and  $g = 50$  nm.

Subsequently, the effect of the phase state of  $\text{Sb}_2\text{S}_3$  on the absorption resonance of the designed metasurface is analyzed. The measured refractive index and extinction coefficient of  $\text{Sb}_2\text{S}_3$  are displayed in Figure 2d, which show a strong refractive index contrast in the near-infrared band. The resonance peak can be regulated due to the refractive index contrast between crystalline  $\text{Sb}_2\text{S}_3$  (c- $\text{Sb}_2\text{S}_3$ ) and amorphous  $\text{Sb}_2\text{S}_3$  (a- $\text{Sb}_2\text{S}_3$ ). Furthermore, the effective modulation of the resonance wavelength can be achieved when  $\text{Sb}_2\text{S}_3$  is in different degrees of crystallization. As shown in Figure 2e, the resonance peak moves from 1320 to 1480 nm before and after the phase transition of  $\text{Sb}_2\text{S}_3$  and undergoes a shift of about 160 nm. The whole structure is in the communication band, which is of great significance for high-performance near-infrared photoelectric detection. Figure 2f shows the electric-field distribution of the designed metasurface with amorphous  $\text{Sb}_2\text{S}_3$  at 1320 nm. It indicates that the electric field is mainly concentrated on the two edges of the metal nanoantenna, which satisfies the typical characteristics of plasma resonance. Our structural configuration significantly enhances the interaction between light and the  $\text{Sb}_2\text{S}_3$  layer. When the  $\text{Sb}_2\text{S}_3$  layer is in different crystalline components, the effective refractive index of the surface plasmon gap changes significantly, thereby expanding the wavelength range of spectral tuning.

For our proposed tunable metasurface, an electrothermal approach is employed to achieve dynamic and reversible phase change of the  $\text{Sb}_2\text{S}_3$  layer. The electrically driven phase change is based on a microheater, whose optical microscopy image is shown in Figure 3a. Different sizes of square  $\text{Sb}_2\text{S}_3$ -based metasurfaces were prepared. However, with expansion of the aperture size of the metasurface, the mitigation of thermal inhomogeneity becomes a key challenge. Therefore, the size of

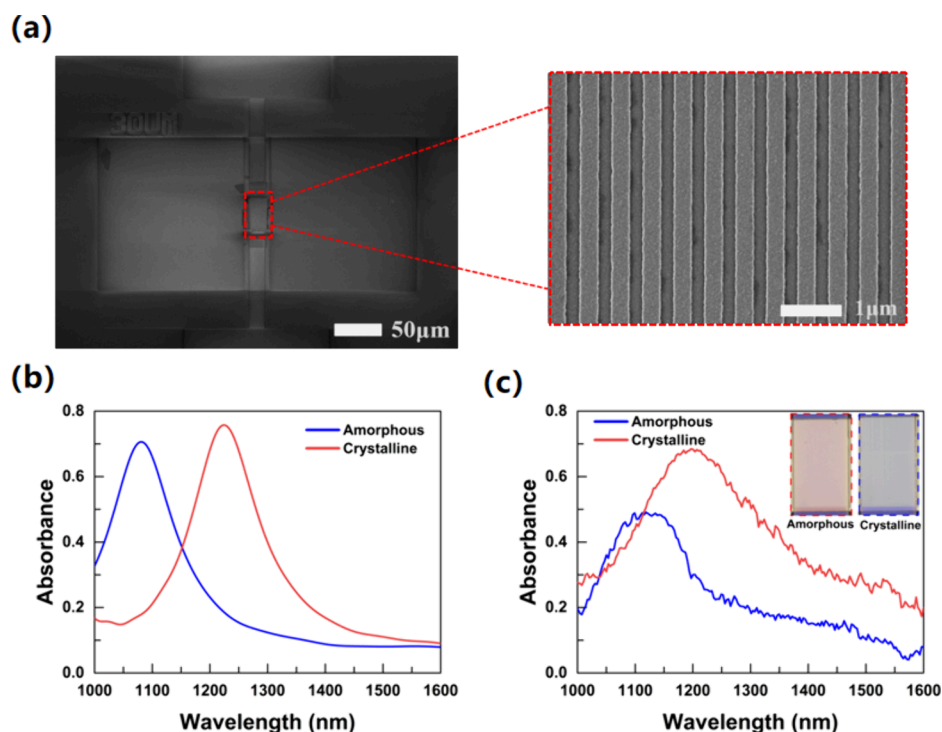
the heating area is  $30 \mu\text{m} \times 30 \mu\text{m}$ , and the area of  $\text{Sb}_2\text{S}_3$  is  $20 \mu\text{m} \times 30 \mu\text{m}$ . It can be seen from Figure 3a that the electrodes at both ends are directly connected to the microheating plate. Among them, the microheating plate is responsible for applying an electric pulse to actively regulate the phase state of  $\text{Sb}_2\text{S}_3$  in the central heating area. The upper and lower electrodes are directly connected to  $\text{Sb}_2\text{S}_3$ , which is responsible for measuring the resistance of  $\text{Sb}_2\text{S}_3$  to determine whether it is a phase transition. The manufacturing process is described in detail in Figure S3.

In each device, the phase state of  $\text{Sb}_2\text{S}_3$  is jointly controlled by electrical pulses, i.e., “set” and “reset”. Long and low voltage pulses (16 ms,  $<7.5$  V) trigger crystallization by Joule heating, as shown in Figure 3b. The short and high voltage pulse (8  $\mu\text{s}$ , 15.7 V) reamorphizes the elemental atoms ( $\sim 550$  °C) through the melt-quenching process, as displayed in Figure 3c. It is worth noting that the high-voltage pulse has extremely short front and rear edges ( $\sim 0.05 \mu\text{s}$ ). This allows  $\text{Sb}_2\text{S}_3$  to be rapidly heated and melted and then rapidly cooled to solidify amorphously. Compared with the previously reported GST regulation,<sup>35,36</sup> our regulation time is a little longer. The time factor is attributed to the size of the heater region, the material, and the thickness of the PCM. Furthermore, under identical conditions,  $\text{Sb}_2\text{S}_3$  exhibits slower switching speeds than GST due to its significantly higher phase transition temperature ( $\sim 270$  vs  $\sim 160$  °C for GST).

The amorphization process requires a sufficiently fast cooling rate. Due to the poor thermal conductivity of silicon oxide, this is also an important factor in the selection of silicon oxide for our substrate. In addition, when the phase of  $\text{Sb}_2\text{S}_3$  is switched, a rapid color change occurs due to the change of its optical constant, as shown in Figure 3d. Optical micrographs of amorphous and crystalline  $\text{Sb}_2\text{S}_3$  films are shown (the up side is the amorphous state, and the down side represents the crystalline state). The surface of crystalline  $\text{Sb}_2\text{S}_3$  shows obvious crystalline silk, and the change of the surface morphology indicates that the phase transition of  $\text{Sb}_2\text{S}_3$  can be regulated by an electric pulse.

The upper and lower electrodes monitor the resistance of the  $\text{Sb}_2\text{S}_3$  film after an electrical pulse. The switching behavior of  $\text{Sb}_2\text{S}_3$  was further analyzed, as shown in Figure 3e. The resistance after each set pulse continuously reduced by at least 2 orders of magnitude, while the resistance after each reset pulse is opposite. It is confirmed that reversible switching between the a- $\text{Sb}_2\text{S}_3$  and c- $\text{Sb}_2\text{S}_3$  stages can be achieved. The slight change in resistance during the reset process may indicate that c- $\text{Sb}_2\text{S}_3$  is not completely reamorphous, as shown in Figure S4.

Figure 3f shows Raman spectra evidencing the transition from amorphous to crystalline  $\text{Sb}_2\text{S}_3$  states. The blue line is the Raman spectrum of the amorphous  $\text{Sb}_2\text{S}_3$  film, which has two broad bands at  $\sim 290$  and  $\sim 100 \text{ cm}^{-1}$ . These bands correspond to the vibrational Sb–Sb bonds in the  $\text{S}_2\text{Sb–SbS}_2$  structural unit and the  $\text{Sb}_2\text{S}_3$  pyramid, respectively.<sup>37,38</sup> Black and red lines represent the Raman spectra of crystalline  $\text{Sb}_2\text{S}_3$  annealed at 300 °C on a hot plate and electrically tuned by a microheater, respectively. The band at  $290 \text{ cm}^{-1}$  is split into two sub-bands, and the band at  $\sim 100 \text{ cm}^{-1}$  is further split into many small peaks. These characteristics are consistent with the Raman spectra of the reported crystalline-state  $\text{Sb}_2\text{S}_3$  film.<sup>39</sup> Therefore, the feasibility of electrically tuning  $\text{Sb}_2\text{S}_3$  is also confirmed.



**Figure 4.** Electrolally reconfigurable plasmon metasurface. (a) SEM images of actively regulated plasmonic metasurfaces. (b) Simulated absorption spectra of the a-Sb<sub>2</sub>S<sub>3</sub> and c-Sb<sub>2</sub>S<sub>3</sub> metasurfaces with a period of 500 nm and a gold grating width of 340 nm. (c) Measured absorption of the metasurface after a set pulse.

Due to the limitation of experimental conditions, a grating structure with  $p = 500$  nm and  $w = 340$  nm was fabricated. Figure 4a shows a scanning electron microscopy (SEM) image of the combination of a gold grating and a microheater. The simulated absorption spectra of the Sb<sub>2</sub>S<sub>3</sub>-based metasurface before and after the phase transition are shown in Figure 4b. The resonance peak shifts to 140 nm after the phase transition. The corresponding measured absorption spectra are displayed in Figure 4c. The peak position and intensity of the experimental spectra are basically consistent with those of the simulated results. The peak is 1109 nm in the amorphous state. After the reset pulse, the resonance peak red-shifts to 1206 nm, and the resonance peak moves 97 nm before and after the phase transition. Because the melting point of gold nanorods is much lower than that of bulk gold, the displacement difference is partly caused by thermal melting of the top gold grating,<sup>40–42</sup> as shown in Figure S5. In addition, Figure 4c also shows the optical micrographs of the Sb<sub>2</sub>S<sub>3</sub> metasurface before and after the phase transition. It can be seen that the color of the metasurface changes significantly after the reset pulse is applied, which is also caused by the Sb<sub>2</sub>S<sub>3</sub> phase transition.

In summary, we have experimentally demonstrated an electrically reconfigurable Sb<sub>2</sub>S<sub>3</sub>-based metasurface. The metasurface adopts an MIM-type structure, and multistage continuous operation of infrared resonance absorption can be achieved by changing the crystal state of Sb<sub>2</sub>S<sub>3</sub>. In addition, compared with vanadium oxide, Sb<sub>2</sub>S<sub>3</sub> is not volatile and can activate and maintain crystalline and amorphous states. Compared with GST, the near-infrared extinction coefficient of Sb<sub>2</sub>S<sub>3</sub> is almost zero. In the experiment, an electric microheater was designed to quasi-continuously, reversibly, and nonvolatily drive the phase change of Sb<sub>2</sub>S<sub>3</sub>. The wavelength shift in the near-infrared spectral range has been

experimentally demonstrated to be achieved by using an electric Sb<sub>2</sub>S<sub>3</sub> metasurface. The measured red shift of the wavelength is 97 nm. Our findings can open up new directions for reconfigurable optical devices that can manipulate the main properties of light for various applications, including imaging, computing, and near-infrared photodetectors.

## ■ ASSOCIATED CONTENT

### Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.nanolett.5c00929>.

Simulation part, experimental preparation, sample characterization, optimization of the PCM metasurface, characterization of electrically tunable Sb<sub>2</sub>S<sub>3</sub> surface morphology, experimental results of the Sb<sub>2</sub>S<sub>3</sub>-based metasurface, and the reflection spectrum under  $y$  polarization (PDF)

## ■ AUTHOR INFORMATION

### Corresponding Authors

**Sha Hu** – School of Physics, Henan University of Technology, Zhengzhou, Henan 450001, China; [orcid.org/0000-0002-3851-8003](https://orcid.org/0000-0002-3851-8003); Email: [husha@haut.edu.cn](mailto:husha@haut.edu.cn)

**Xin Huang** – Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China; School of Physical Sciences, CAS Key Laboratory of Vacuum Physics, University of Chinese Academy of Sciences, Beijing 100190, China; Email: [xinhuang@iphy.ac.cn](mailto:xinhuang@iphy.ac.cn)

**Changzhi Gu** – Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China; School of Physical Sciences, CAS Key Laboratory of Vacuum Physics, University of

Chinese Academy of Sciences, Beijing 100190, China;  
orcid.org/0000-0002-2689-2807; Email: czgu@iphy.ac.cn

## Authors

**Zhuoxuan Han** – Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China; School of Physics, Henan University of Technology, Zhengzhou, Henan 450001, China; orcid.org/0009-0007-1462-5799

**Chensheng Li** – Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China; orcid.org/0000-0002-8902-6594

**Tengzhang Liu** – Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China

**Nannan Hu** – Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China; School of Physics, Henan University of Technology, Zhengzhou, Henan 450001, China

**Zhiqin Fan** – School of Physics, Henan University of Technology, Zhengzhou, Henan 450001, China

**Yang Guo** – Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China; School of Physical Sciences, CAS Key Laboratory of Vacuum Physics, University of Chinese Academy of Sciences, Beijing 100190, China; orcid.org/0000-0001-8975-9387

**Baoli Liu** – Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China; School of Physical Sciences, CAS Key Laboratory of Vacuum Physics, University of Chinese Academy of Sciences, Beijing 100190, China

**Haifang Yang** – Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China

**Aizi Jin** – Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China

**Baogang Quan** – Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China

**Shibing Tian** – Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China

**Yang Yang** – Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China; orcid.org/0000-0003-3785-9233

**Geng Li** – School of Physical Sciences, CAS Key Laboratory of Vacuum Physics, University of Chinese Academy of Sciences, Beijing 100190, China; orcid.org/0000-0002-3347-7222

**Xiaofeng Fan** – Key Laboratory of Automobile Materials (Jilin University), Ministry of Education, and College of Materials Science and Engineering, Jilin University, Changchun 130012, China; orcid.org/0000-0001-6288-4866

Complete contact information is available at:

<https://pubs.acs.org/10.1021/acs.nanolett.5c00929>

## Author Contributions

Z.H. and X.H. conceived the initial idea of electrically reconfigurable plasmonic metasurfaces based on the PCM

$\text{Sb}_2\text{S}_3$ . Z.H. and X.H. developed the theory and performed numerical simulations with the help of T.L., N.H., and Z.F.. Z.H. and S.H. prepared the manuscript. Z.H. conducted the experiment and performed the tests with the help of C.L., Y.G., B.L., H.Y., A.J., B.Q., S.T., Y.Y., G.L., and X.F.. Z.H., S.H., and C.G. supervised the overall project. All of the authors analyzed the data and discussed the results.

## Notes

The authors declare no competing financial interest.

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