

Laser-induced reconfigurable wavefront control with a structured Ge₂Sb₂Te₅-based metasurface

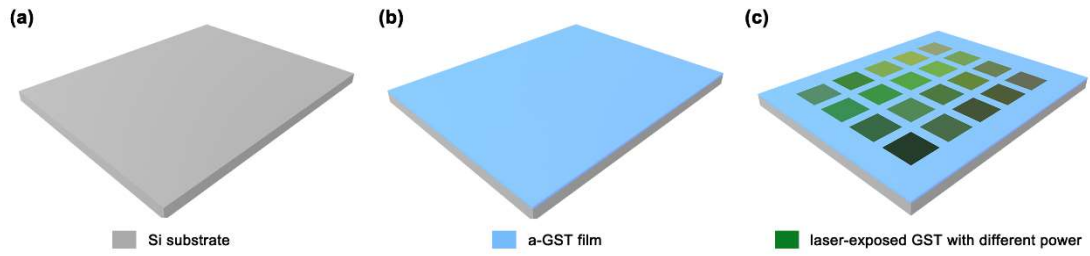
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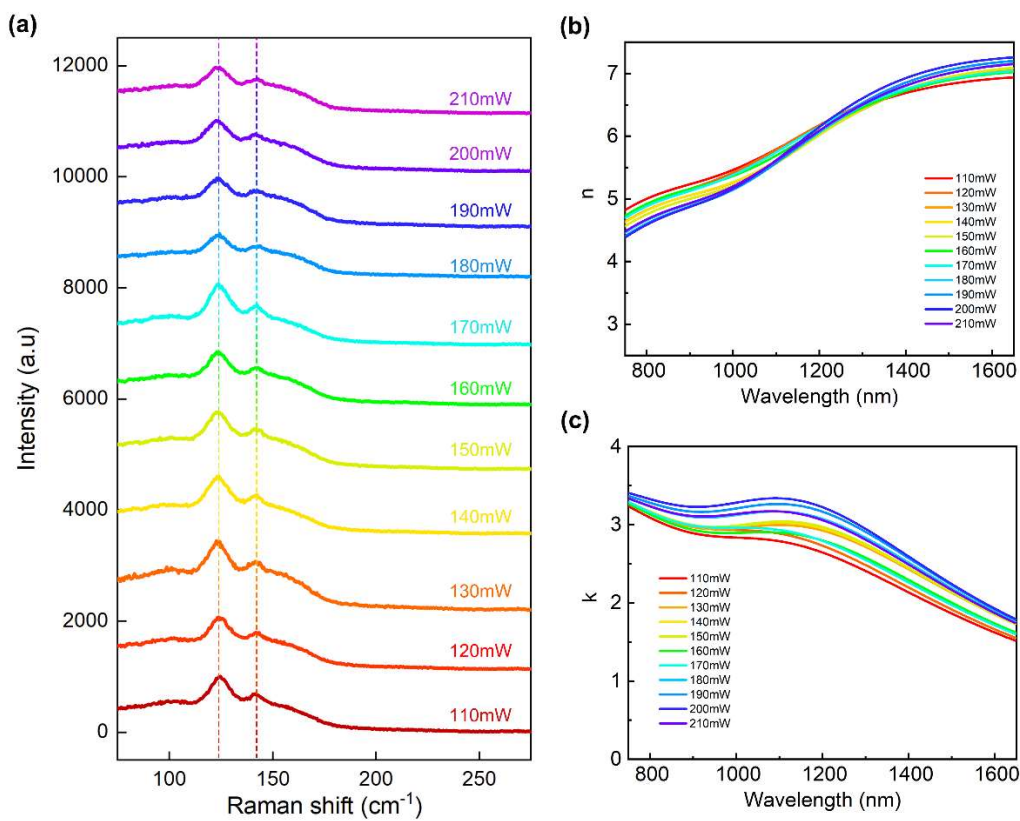
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Supplementary Note 1: Multi-level phase change of GST film controlled by laser power

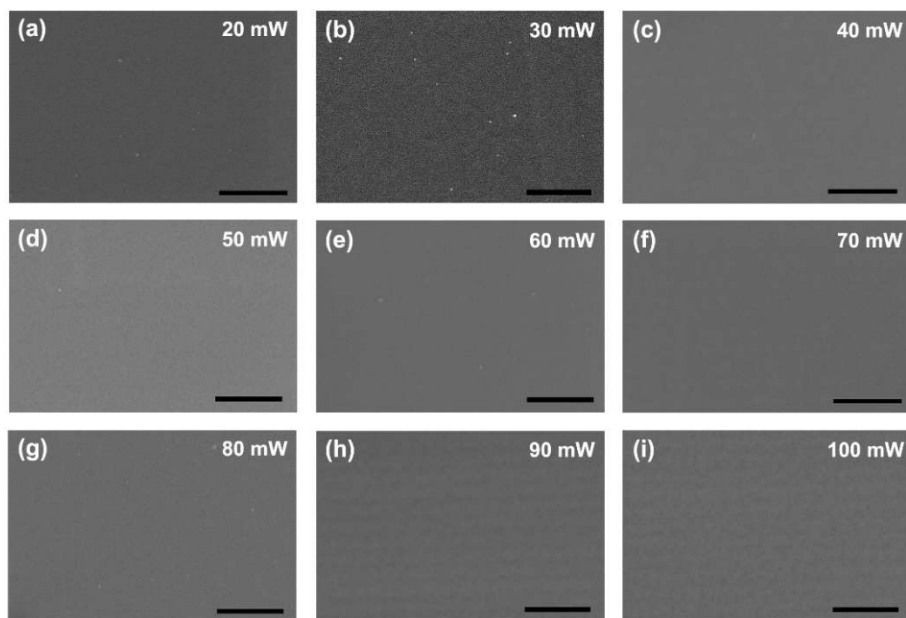


Supplementary Figure 1. Schematic of the experimental process for laser-induced phase change of GST film. **a** Cleaned Si substrate. **b** GST film deposited on the Si substrate by magnetron sputtering. **c** Laser-induced phase change of GST film by a laser writing system (DWL 66⁺). 5×4 areas of the GST film are exposed by DWL 66⁺ system with laser powers in 10 mW increments in the range of 20 mW-210 mW. Each area is up to 2 mm × 2 mm and spaced 1 mm apart from its neighboring areas.

By controlling the power of laser source, multi-level switching between the amorphous and crystalline states can be achieved, as shown in Fig. 2. Supplementary Figure 2a shows the measured Raman spectra of GST under laser irradiation of 110-210 mW. When the laser power exceeds 100 mW, there exist two peaks in the Raman spectra and no obvious change is observed. The corresponding refractive index and extinction coefficient are shown in Supplementary Figure 2b, c. Similarly, only slight changes exist in both refractive index and extinction coefficient. That's to say, the phase state of GST keeps stable with laser power exceeding 100 mW.



Supplementary Figure 2. **a** Raman spectra of laser-irradiated GST thin films. Measured **(b)** refractive index (n) and **(c)** extinction coefficient (k) of GST films exposed by laser source. Here, the laser power varies from 110 mW to 210 mW.



Supplementary Figure 3. The SEM images of GST films exposed by various laser power. The scale bars are all 1 μm. When laser power varies from 20 mW to 100 mW, the GST films keep uniform.

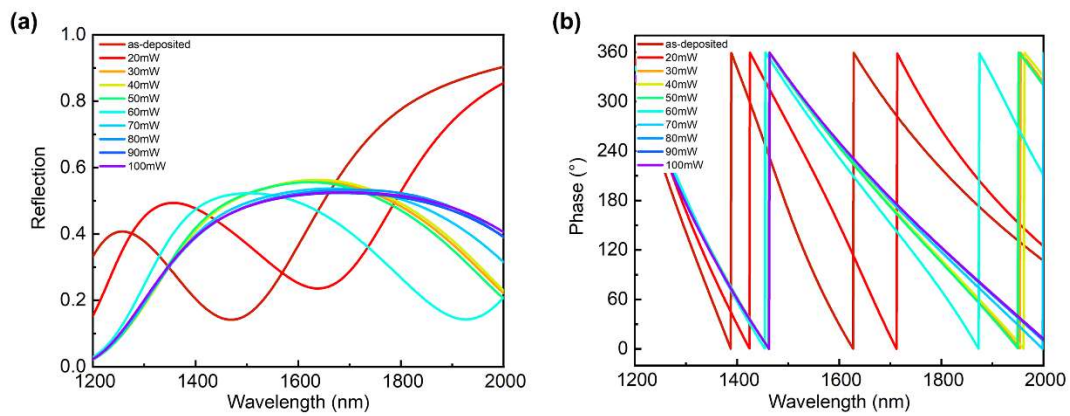
Supplementary Note 2: Complete phase and amplitude modulation of laser-driven GST-rod array

The crystallized GST delivers metallic character in the visible-NIR spectral range^{1,2}. Thus, the demonstrated metasurface can be considered as a typical metal/insulator/metal-like (MIM-like) structure, which consists of GST-rod array and a gold film separated by a ZnO layer, as schematically depicted in Fig. 3a. The MIM configuration has been used in photonics research with applications ranging from perfect absorption to phase modulation. The desired functionalities can be achieved through varying the geometrical and material parameters of MIM structures, which has been proved both experimentally and theoretically^{3,4}. Our proposed metasurface reconstruct multifunctionalities through controlling the material parameters of GST. Moreover, the gap-surface plasmon mode inside the MIM-like structure makes its reflection phase easily cover the full 2π range⁵.

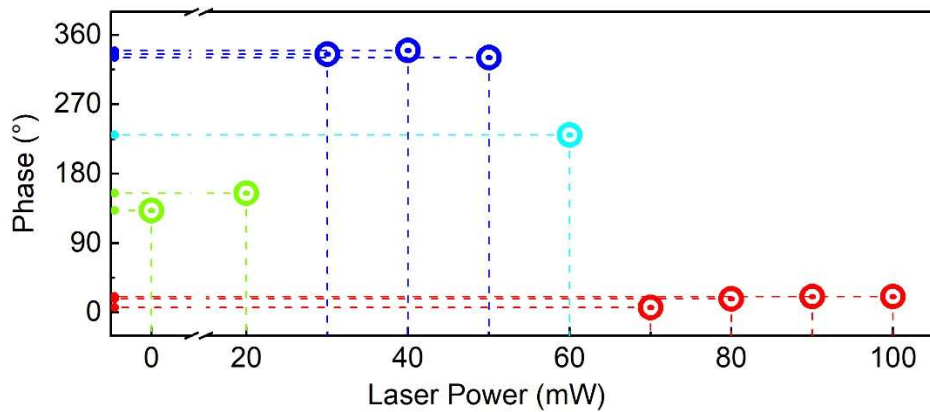
The resonances for a metasurface are closely related to the optical properties and the geometric size of meta-atoms. On one hand, the length L_x has great impact on the optical resonances of the GST-rod array due to the incident polarization along the x -axis. On the other hand, the resonances of GST-rod metasurface are directly dependent on the refractive index of GST, which is essentially determined by the laser power. Thus, we will focus on the effect of L_x and laser power on the phase and amplitude modulation of GST-rod array. For our demonstrated reconfigurable metasurface, the corresponding geometric parameters remain unchanged. The reconfigurability is achieved by changing the laser power. Subsequently, the reflection and phase spectra as a function of laser power are shown in Supplementary Figure 4. The lengths of the GST rods are set as $L_x = 500$ nm and $L_y = 300$ nm, respectively. As Fig. 3b, c show, efficient amplitude and phase modulation has been obtained at the wavelength 1370 nm and 1622 nm, respectively. In addition, Supplementary Figure 4 indicates that complete phase modulation and high amplitude control can be achieved in a broad bandwidth.

The wavefront manipulation of our proposed optical devices is based on resonant phase, which is closely associated with the geometric parameters of unit cell, operating

wavelength and the optical properties of materials. Assuming that the geometric parameters are fixed with $L_x = 250$ nm and $L_y = 450$ nm, the operating wavelength λ is considered to be 1623 nm. As shown in Supplementary Figure 5, a nearly full 2π phase range can be achieved by controlling laser power. That's to say, a nearly full 2π phase range can be obtained through optimizing geometric parameters and operating wavelength. What's more, through decreasing the change interval of laser power, a finer phase interval can be yielded.



Supplementary Figure 4. Simulated (a) reflection and (b) spectra of GST-rod array with the control of laser power. Here, L_x and L_y are set as 500 nm and 300 nm, respectively.

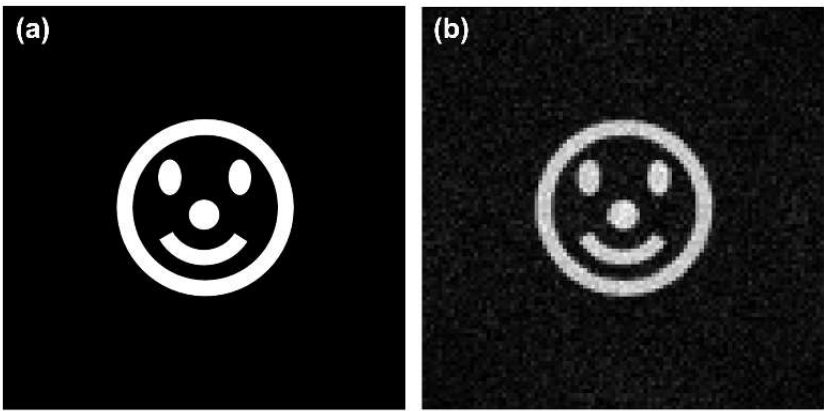


Supplementary Figure 5. Multi-level phase distribution with changing laser power from 20 mW to 100 mW. Here, the geometric parameters are fixed with $L_x = 250$ nm and $L_y = 450$ nm, and the operating wavelength λ is considered to be 1623 nm.

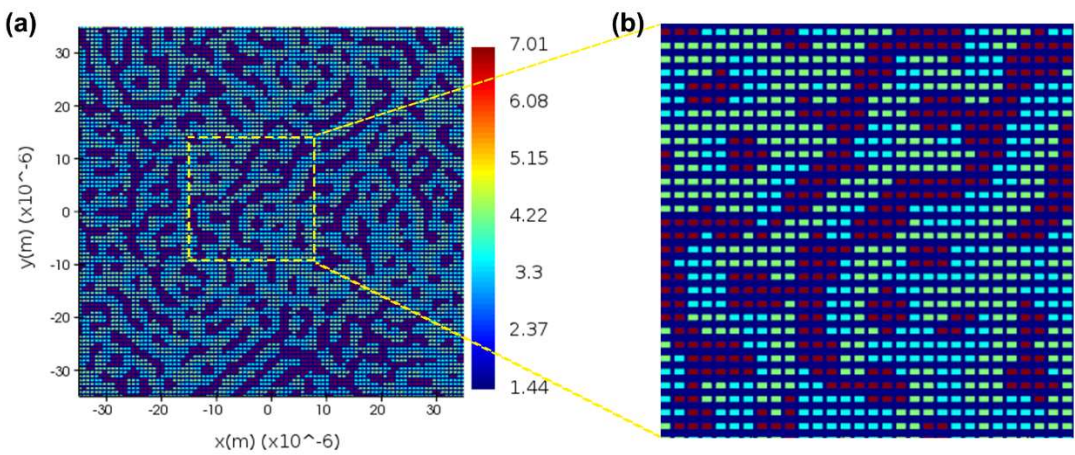
Supplementary Note 3: Reconfigurable optical devices based on multi-level phase and amplitude modulations

Supplementary Table 1. The selected three-level phase modulation and their corresponding reflections and laser powers.

Phase (°)	55.0	185.7	299.8
Reflection (%)	52.6	42.5	23.9
Laser Power (mW)	80	0	20



Supplementary Figure 6. **a** Target image and **(b)** the calculated three-level holographic image by using GS algorithm.



Supplementary Figure 7. **a** Refractive index distribution for the proposed three-level hologram of a smiling face. **b** Partial detailed refractive index distribution.

Supplementary Table 2. The selected four-level reflection modulation and their corresponding laser powers.

Reflection (%)	25.7	34.2	42.9	49.2
Laser Power (mW)	0	80	60	20

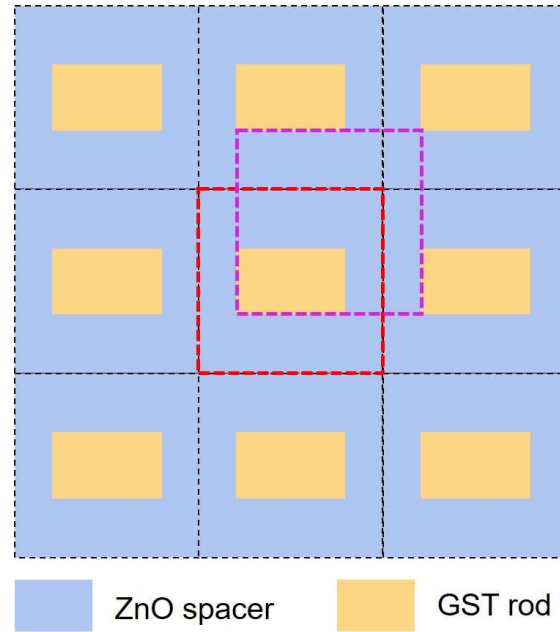
Supplementary Note 4: Potential manufacturing process for the proposed reconfigurable GST-rod metasurface.

The potential manufacturing process for the proposed reconfigurable GST-rod metasurface is as follows. First, the 100 nm gold film is deposited on a quartz substrate by electron beam evaporation. Subsequently, the ZnO spacer with a thickness of 40 nm is deposited on the gold film by atomic layer deposition. Then, electron beam lithography is performed to pattern the rod array with the PMMA resist. After this, the rod pattern is transferred to GST by magnetron sputtering and lift-off processes. In the next step, a 120 nm SiO₂ layer is deposited onto the GST-rod array as the protective layer by plasma-enhanced chemical vapor deposition. Finally, the laser writing system, which can precisely control the stage surface with low positioning errors, is used to modify GST-rod metasurface as desired. It should be noted that the laser power shall not be too high. Too high laser power will make the laser induced temperature higher than the melting point of GST and result in the decrease of crystallinity⁶.

The alignment accuracy is the key implementation point to achieve selective phase change of GST rods. The required alignment accuracy is determined by both the feature size of laser writing and geometric size of meta-atom. A suitable write mode enables the exposures of structures with a feature size of 750 nm, which is equal to the period. The optimized lengths of the GST rods are set as $L_x = 500$ nm and $L_y = 300$ nm, respectively. In this case, the alignment accuracy along x - and y -directions should be controlled within 125 nm and 225 nm, respectively, as indicated in Supplementary Figure 8.

Although the repeatability of GST has not been studied experimentally in our paper, the highly reproducible switching of GST by applying laser irradiation has been proved in previous reports. For examples, GST films have been widely applied in memory devices, which can operate over many write-erase cycles⁷⁻⁹. Particularly, the structured GST-based meta-devices have been demonstrated to realize dynamic wavefront manipulation via laser irradiation. For the measured switching cycles, the wavefront modulations keep almost constant¹⁰⁻¹². These studies indicate that the laser-

induced switching process of GST is highly reproducible.



Supplementary Figure 8. The scheme of the required alignment accuracy to achieve the structured $\text{Ge}_2\text{Sb}_2\text{Te}_5$ -based metasurface. The black dashed rectangle denotes the boundary of meta-atoms. The red dashed rectangle indicates the perfect alignment while the purple dashed rectangle represents a critical alignment situation.

Supplementary References

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