A Simple Method to Reversibly Switch the Reflectance Spectrum of a Layered Structure Consists of an Ultra-Thin Film Phase-Change Material GST

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Abstract. By applying pulsed laser and CW laser to irradiate the top layer phase-change material GST, we realized reversible optical switching of reflectance of a layered structure. The FTIR measured reflectance spectrum proved that the reflectivity at 8μm could be changed reversibly from 0.19 to 0.90. Our research may have some inspiration for active thermal control and other applications.

1. Introduction

Controlling the reflective properties of objects at infrared wavelengths is of great importance for applications such as radiative cooling [1], thermal extraction [2, 3], designing absorbers for detection [4], molecular sensing [5], or thermal voltaic energy conversion [6].

Phase change materials (PCMs) [7], which are typically based on the combinations of the elements germanium, antimony, tellurium, and indium, show a pronounced change of their dielectric function during the phase transition between the amorphous and the crystalline state. Among these PCMs, Ge$_2$Sb$_2$Te$_5$ (GST) is a good candidate for infrared applications because of its high contrast in refractive index and extinction coefficient between amorphous state (aGST) and crystalline state (cGST) and repeatable tenability [8].

GST is usually sputter-deposited onto a substrate as a thin film at the amorphous state. If a long and high energy stimulus (i.e. thermal, electrical or optical) was applied to the aGST to heat the material above its crystallization temperature ($T_c \approx 160^\circ$) and hold the temperature above $T_c$ for a short time, the GST will be tuned to the crystalline state. In contrast to the crystallization process, the re-amorphization process needs a short and higher energy pulse to heat the cGST above its melting temperature ($T_m \approx 600^\circ$) and then a sudden cooling down within the crystallization time. In previous work, a cooling rate of 1°per nanosecond is needed for the melt quenching of GST [9]. The considerable number of vacancies in the crystalline GST contributes to the free charge carriers and associated absorption below the energy gap [10]. Different from other PCMs like vanadium dioxide [11, 12], the phase of GST is stable after removing the stimuli. Besides amorphous state and crystalline state, stepwise intermediate states can also be realized in thermal or optical approaches which enlarge the potential for reconfigurable photonic applications [13]. In most of the previous works, the reversible optical induced phase change of GST was done by applying femtosecond laser [14].

In this paper, we propose an innovative but rather simple method that using a laser diode to reversibly tune the reflectance spectrum of a layered structure that consists of an ultra-thin film of phase-changing material GST. With the laser direct writing system, patterns with a minimum pixel...
size about 1 micron can be written. By measuring the absorption spectrum of the sample, we proved accurately that the a-c-a phase transitions happened. Finally, by writing some random patterns with different a/c area ratios, we realized intermediate reflectance spectra between aGST sample and cGST sample stepwise. Our research may be useful for thermo-photonic applications.

2. Phase-Changing Experiment and Results

The sample we use in the experiment is a multilayer structure that consists of a highly reflective Au layer, a ZnS layer and a thin GST layer, and the designed thicknesses are 100nm, 800nm and 25nm respectively. But due to the fabrication imperfection, the real thicknesses are 50nm, 620nm and 30nm respectively as shown in figure 1(a). The experiment setup includes mainly a 405nm laser diode and an arbitrary signal generator employed to modulate the laser, a piezoelectric ceramic displacement platform and a mechanical displacement platform as shown in figure 1(b).

![Figure 1](image)

Figure 1. Sample and experiment setup. (a) SEM image of the fabricated sample comprising a thin layer GST, a ZnS layer about 750nm in thickness and a high reflective Au layer on a silicon wafer. (b) Schematic of the phase changing experiment. Laser modulated by the arbitrary signal generator irradiates the sample surface after being focused by the 100× objective.

The structural parameters of the sample were designed to maximize the reflective difference of the system at 555nm wavelength in aGST or cGST states, which is advantageous to judge roughly whether the phase transition occurred under the microscope. The as-deposited aGST sample surface is darker than that of cGST because of the designed difference in the reflectivity at 555nm. As shown in figure 2(a), after applying continuous laser of proper power to a squared area, the irradiated area looks brighter than the surrounding area, which means the crystallization happened. For the re-amorphization of cGST, we applied a pulsed laser of about 10 nanoseconds to irradiate some part of the cGST to “write” a pattern, as illustrated in figure 2(b), the irradiated area looks as dark as the surroundings. To prove that the phenomenon of darkening is because of re-amorphization rather than surface damage we applied continuous laser again to “erase” the pattern, it became brighter as a result of crystallization as shown in figure 2(c). So far, a cycle of “write” and “erase” was completed.
Figure 2. “write” and “erase” process. (a) Crystallization of GST in a squared area induced by continuous laser. (b) A “GST” pattern written by a pulsed laser. (c) The area after erased by continuous laser.

Reflectance spectrums measured by Fourier Transform Infrared Spectrometer (FTIR) were used to analyze the phase transition of GST and reversible tunability of the reflectance spectrum of the sample for better accuracy. To find out the variation rule of reflection with the crystallization fraction of GST, we calculated the reflectance spectrum of the layered structure of different crystallization fractions using the Transfer Matrix Method (TMM), and the result is shown in figure 3(a). The dielectric function of intermediate state GST with crystallization fraction X was obtained using Lorentz-Lorentz relation [15, 16], with $\varepsilon_c$ and $\varepsilon_a$ are dielectric functions of GST at the crystalline state (X=1) and the amorphous state (X=0).

Figure 3(b) shows the measured absorption spectrum of the as-deposited aGST sample area and continuous laser irradiated crystalline area, as well as the pulsed laser-induced re-amorphous GST sample, and it is obvious that the irradiated part possesses lower reflectivity than the untreated part, which can prove the crystallization happened. After being irradiated by continuous laser, the absorption spectrum of the re-amorphous sample becomes nearly the same with that of the initial aGST sample.

As we can see from figure 3(a)(b), the measured reflectance spectrum is much lower than the calculated one, which may result from the inaccuracy of the cGST dielectric function we use in the calculation.

Figure 3. Calculated and measured reflectance spectrum. (a) Calculated reflectance spectrum using the Transfer Matrix Method. With the increasing of crystallization fraction, the reflectivity decrease. (b) Measured reflectance spectrum using Fourier Transform Infrared Spectrometer. The reflectivity decreases from 0.93 to 0.19 at 8\mu m wavelength after the continuous laser-induced crystallization, and increases from 0.19 to 0.90 after the pulsed laser-induced amorphization.
3. Realization of Intermediate Reflectance Spectrum

After the reversible phase transition has been realized, we write some random patterns in which the area proportions of cGST are different. The different area proportion is realized by applying pulsed laser to some points of the crystallized area to re-amorphized them and assume that the ratio of points number not irradiated by the pulsed laser to the total number is equal to the crystallization fraction X. The FTIR measured reflectance spectrum is shown in figure 4(a). We can see that the reflectance spectrum of the structure depends on the degree of crystallization, but not linearly proportional to the fraction of the crystalline phase in the top layer GST.

![Figure 4](image)

**Figure 4.** Reflectance of different crystallization fraction samples. (a) Measured reflectance spectrum of samples that crystallization fraction X = 0, 20%, 40%, 60%, 80% and 100%. (b) Maximum reflectivity at 8μm wavelength with crystallization fraction X.

4. Conclusion

In summary, a filmed structure with a top layer phase-change material GST was designed, whose reflectance spectrum could be tuned reversibly because of the laser-induced phase transitions between the amorphous state and the crystalline state of GST. Some random patterns of different crystallization areas were written and analysed by measuring the reflectance spectrum, and the results show that reflectivity from 0.19 to 0.90 could be tuned stepwise.

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6. References

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