Single-shot Real-time Observation of Ultrafast Amorphization in $Ge_2Sb_2Te_5$ Thin Film

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Abstract: Ultrafast dynamics of photo-induced amorphization in $Ge_2Sb_2Te_5$ thin film has been studied using broadband single-shot real-time pump-probe imaging spectroscopy. We successfully observed the transient absorption changes accompanied with the ultrafast amorphization with a single-shot detection.
OCIS codes: (260.7120) Ultrafast phenomena; (300.6500) Spectroscopy, time-resolved; (210.4810) Optical storage-recording materials

1. Introduction

$Ge_2Sb_2Te_5$ (GST) is one of the rapid phase-change materials, which has been applied to rewritable optical media such as digital versatile disc-random access memory (DVD-RAM). The phase-change from the amorphous to the crystalline phase is induced by an irradiation of a cw-laser through transient temperature ramping [1]. The phase-change from the crystalline to the amorphous phase, on the other hand, is induced by an irradiation of a single femtosecond laser pulse through a non-thermal process [2]. A model of the amorphization was proposed by Kolobov *et al*. [3]; the relatively weaker and longer Ge-Te bond is broken due to an intense photoexcitation, and subsequently, the amorphization occurs due to displacement of the Ge atoms from octahedral to tetrahedral arrangement with a subpicosecond time scale (the inverse of the phonon frequency).

In order to detect the photo-induced amorphization in GST, a measurement of transient absorption or transient reflectivity changes by using pump-probe spectroscopy is required. However, the amorphization is irreversible, and therefore, it is difficult to reveal the details of the dynamics with the conventional pump-probe spectroscopy that requires many repetitions of pump-probe sequence. In the present work, we performed real-time measurements of the transient absorption changes in GST using single-shot real-time pump-probe imaging spectroscopy [4-6].

2. Experimental method

A schematic configuration of the single-shot real-time pump-probe imaging spectroscopy is depicted in Fig. 1. The key aspect of this technique is the use of an echelon mirror, which is fabricated on a Ni block (40×40×20 mm^3) with 500 steps having 80 μm step-width and 20 μm step-height. As shown in Fig. 1, using the double four focusing configuration, the echelon mirror does not act as diffractive optics but generates spatially encoded delay times for the probe pulse. The probe pulse with spatially encoded delay times is focused onto a sample together with a pump pulse. After passing through the sample, the probe pulse is imaged onto an entrance slit of a spectrometer coupled with a two-dimensional (2D) charge coupled device (CCD) detector (1340×1300 pixels). The vertical axis of the CCD detector corresponds to the delay time, whereas the horizontal axis corresponds to the detected wavelength. Using this method, we are able to obtain time-frequency 2D image of transient absorption changes on a single-shot basis.

The light source was a Ti:sapphire regenerative amplifier system with center wavelength of 800 nm and pulse duration of 100 fs. The probe pulse was focused into CaF_2 thin plate to generate a white-light continuum by self phase modulation. As a result, we could perform the single-shot mapping of the transient absorption changes in the range of 530-660 nm and the time window of 15 ps. The sample used in this study was a 10 nm thick GST film deposited on a glass substrate.

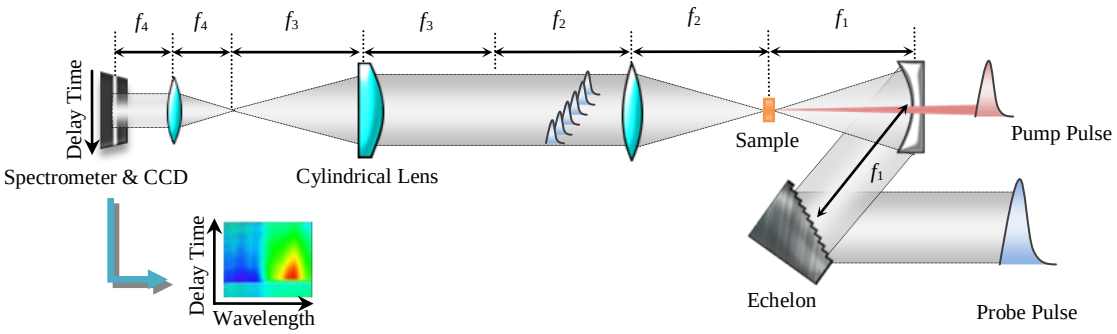

Fig. 1. Schematic configuration of single-shot real-time pump-probe imaging spectroscopy.

3. Results and Discussion

Figure 2(a) shows the time-frequency 2D image of the absorption change in the crystalline GST film measured by a single-shot detection. Here, the excitation laser fluence was 23.4 mJ/cm^2 where the recording mark associated with amorphization is formed. As shown by a dotted line, the absorption rapidly decreases at the time origin and the change does not recover even after long delay time, suggesting that the irreversible phase-change to the amorphous phase takes place. Figure 2(b) shows the time evolution of the absorption change obtained by cutting off the 2D image at 650 nm. The fast rise time was evaluated by a fitting procedure convoluted by the system response function, and was estimated to be 150 fs as shown by a dotted curve. The estimated rise time of 150 fs is consistent with the order of the inverse of the phonon frequency [7,8]. This fact strongly indicates that the amorphization in GST is due to the rearrangement of Ge atoms as shown in Fig. 3.

We also observed the excitation power dependence of the absorption change, which shows saturation behavior without a critical threshold. On the other hand, the recording mark due to amorphization is engraved with a clear threshold laser fluence. This fact suggests that the stabilization of the amorphous phase might occur in the longer time scale.

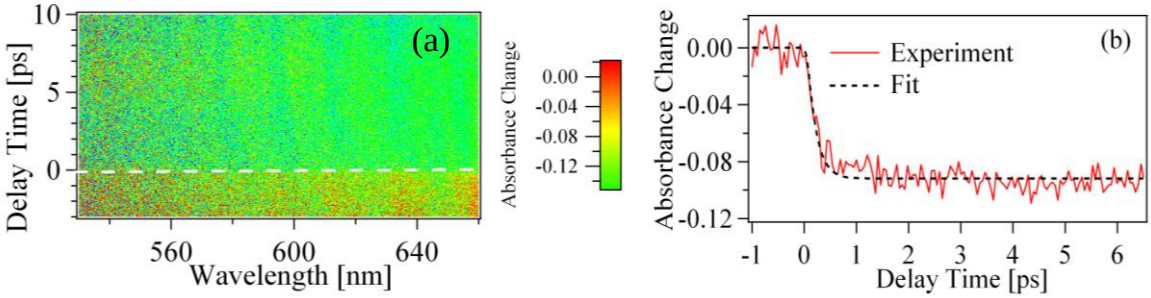

Fig. 2. (a) Time-frequency 2D image of the absorption change obtained in GST. (b) Time evolution of the absorbance change obtained by cutting off the 2D image at 650 nm. The solid and dotted lines are experimental data and the best fits to the data, respectively.

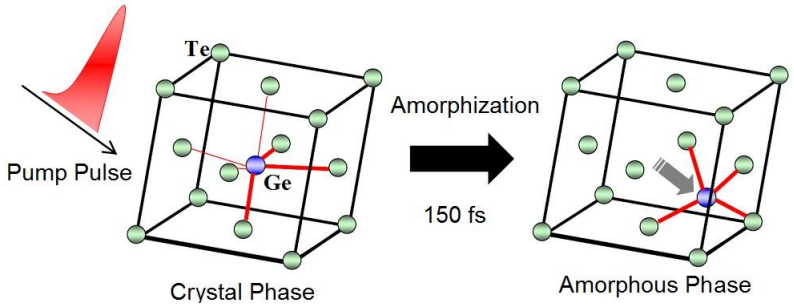

Fig. 3. Diagram of the photo-induced phase-change in GST.

4. Summary

In conclusion, we successfully observed ultrafast amorphization in GST for the first time using single-shot real-time pump-probe imaging spectroscopy with an echelon mirror. The phase-change occurs within the inverse of the phonon frequency, suggesting that the amorphization is due to the rearrangement of Ge atoms.

5. Acknowledgment

We would like to thank T. Shintani for providing the GST thin film sample.

6. Reference

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