Crystallization behavior of amorphous GeSbTe nonthermally amorphized by femtosecond laser pulse irradiation

Kazuyuki Tajima, Nariaki Kitamura, Yuki Hongo, and Toshiharu Saiki
Graduate School of Science and Technology, Keio University
3-14-1 Hiyoshi, Kohoku, Yokohama, Kanagawa 223-8522, Japan
tajima@saiki.elec.keio.ac.jp

ABSTRACT

By multi-femtosecond pulse irradiation we investigated crystallization process of GeSbTe (GST) in various amorphous states. We compared the number of pulses required for crystallization of amorphous GST prepared by three different ways: as-deposited, amorphized by single-femtosecond (higher-fluence) pulse irradiation, and amorphized by multi-femtosecond (lower-fluence) pulse irradiation. We found that the amorphous GST nonthermally amorphized by lower fluence pulses was recrystallized most efficiently in terms of the number of irradiation pulse.

Keywords: femtosecond laser pulse, nonthermal amorphization

1. INTRODUCTION

High speed crystallization in GeSbTe-based phase change materials is one of the most critical factors to be addressed for rewritable optical and electronic memory applications. Recently we have found that a single femtosecond laser pulse induces nonthermal amorphization, which takes place below the melting temperature [1]. In a pump-probe measurement in transient reflectivity, the reflectivity dropped abruptly within 500 fs after excitation by a single pulse. This ultrafast amorphization indicates that switching of Ge atoms can be initiated by photoexcitation of electrons to break the weaker Ge−Te bond, and that it is completed within a subpicosecond time scale. If this scenario is true we would then expect that photoexcitation also makes a significant contribution to the switch-back of Ge atom: recover to the crystalline phase.

In this study we performed crystallization of amorphous states by multi-pulse irradiation of femtosecond pulse. We found that the amorphous state induced by femtosecond pulse irradiation can be much more easily crystallized compared with as-deposited and melt-quenched amorphous phases.

2. EXPERIMENTS

The sample investigated was a $Ge_2Sb_2Te_5$ (GST) thin film with a thickness of 20 nm sputtered on a glass substrate. The GST film was covered with a 5-nm SiO_2 protection layer. The main excitation source was a Ti:Sapphire laser system operating at 800 nm central wavelength with a pulse duration of 170fs. In order to compare crystallization behavior depending on initial amorphous states, we amorphized the GST film with three different ways: (i) as-deposited, (ii) amorphized by single-femtosecond (higher-fluence) pulse irradiation, and (iii) amorphized by multi-femtosecond (lower-fluence) pulse irradiation.

For crystallization of the individual amorphous states, a train of weak femtosecond pulse with fluence of 6.5 mJ/cm^2 was focused onto the amorphous area of the sample. A He-Ne laser was used as a probe beam to measure the change of transmission upon crystallization after each femtosecond pulse excitation.

3. RESULTS & DISCUSSION

Figures 1(a)-(c) show normalized transmissions of probe beam as a function of the number of femtosecond pulses irradiated for different initial amorphous states. In the case of amorphous state induced by multi-femtosecond pulse irradiation, only 25 pulses are sufficient to reach 10% decrease in transmission whereas 150 pulses are required for the as-deposited amorphous state. The difference mainly comes from the fact that, in the case of as-deposited, the first 70 pulses serve to create nucleation centers for crystallization.

It is also important to note that 120 pulses are needed to crystallize the amorphous state induced by single-femtosecond pulse irradiation, where a higher fluence pulse is applied compared to the case of multi-femtosecond pulse irradiation. The reason for this would be as follows: at the center of irradiated area the temperature reaches above the melting point and melt-quenched amorphization takes place. Similar to the as-deposited amorphous state, in the case of melt-quenched state, a few tens of pulses are needed for the creation of nuclei and crystallization proceeds more slowly.

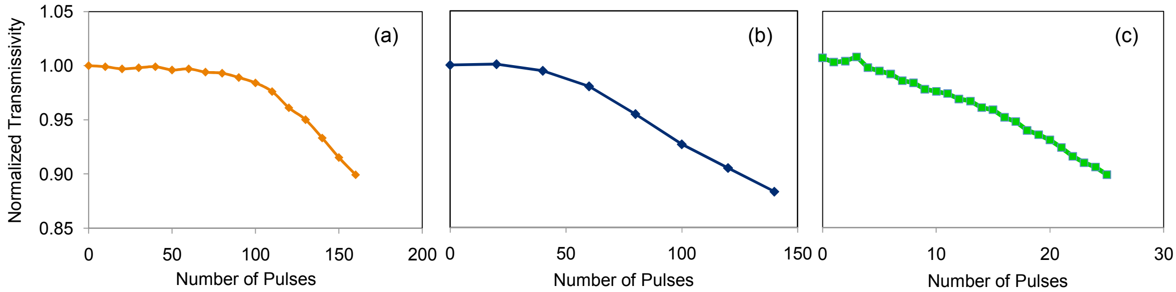

Figure 1: Normalized transmissions of probe beam as a function of the number of femtosecond pulses irradiated for three different initial amorphous states: (a) as-deposited, (b) amorphized by single-femtosecond pulse irradiation, and (c) amorphized by multi-femtosecond pulse irradiation.

4. CONCLUSION

Under multi-femtosecond pulse irradiation we compared crystallization behavior of GeSbTe thin films with different amorphous phases. We found that crystallization of the amorphous state nonthermally created by low-fluence femtosecond pulse irradiation is the most efficient in terms of the number of irradiation pulse. The result suggests that the photoexcitation of high density electron can be a trigger for fast and efficient crystallization.

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