Formation of a crystalline and amorphous periodic structure by femtosecond pulse excitation of GeTe

Takanori Morita, Yosuke Katsumata, Yusuke Morimoto, and Toshiharu Saiki
Graduate School of Science and Technology, Keio University
3-14-1 Hiyoshi, Kohoku, Yokohama, Kanagawa 223-8522, Japan
takanori.morita@saiki.elec.keio.ac.jp

ABSTRACT

By multiple femtosecond laser pulse amorphization of a GeTe thin film, we found that a self-organized periodic structure in a single amorphous mark was formed. Micro-Raman imaging unveiled that the periodic structure is consisted of crystalline and amorphous phases. For a $Ge_2Sb_2Te_5$ thin film, however, we obtained homogeneous amorphous marks and did not observe similar fringe pattern even over a wide range of excitation conditions. This probably originates from difference between threshold properties of nonthermal amorphization and thermal crystallization.

Key words: femtosecond laser pulse, nonthermal process, GeTe, periodic structure

1. INTRODUCTION

Recently we have demonstrated ultrafast nonthermal amorphization of $Ge_{10}Sb_2Te_{13}$ on sub-picosecond time scale by femtosecond laser pulse excitation, and found that this nonthermal phase change has distinct threshold property [1]. In this study, inspired by the formation of laser-induced periodic surface structures by ultrafast pulse excitation, we came to the idea that the collaboration of the threshold behavior with large contrast in refractive index between crystalline and amorphous phases will generate a self-organized spatial pattern in a single amorphous mark even if spatially homogeneous excitation is provided.

2. EXPERIMENTS

The sample was a 10 nm thick GeTe (GT) film and a $Ge_2Sb_2Te_5$ (GST225) film deposited by sputtering on a glass substrate. These films were covered with 10 nm thick SiO_2 layer and were annealed at 250°C to obtain a crystalline phase. The excitation source for amorphization was a mode-locked Ti:Sapphire laser operating at 800 nm central wavelength with a pulse duration of 260 fs. The laser beam was focused onto the sample surface using an objective with a numerical aperture (NA) of 0.15 for writing marks. Scanning laser microscopy and micro-Raman imaging spectroscopy were performed to analyze the constituents of spatial pattern.

3. RESULTS & DISCUSSION

Figure 1 (a) shows a laser scanning micrograph of amorphous mark of GT. This mark was obtained by irradiation of 100 femtosecond laser pulses with a pulse energy of 1.4 nJ. We found that the direction of the periodic structure is always parallel to the polarization of the incident light, and that several tens of pulse irradiation was needed to form the structure. The period is estimated to be 580 nm, which is smaller than the wavelength of irradiation light. On the other hand, any periodic structure was not observed in GST225 (Fig. 1(b)). Figures 1(c) and 1(d) show micro-Raman spectra and spatial mapping of crystalline and amorphous phases of GT respectively. The green and red areas in Fig. 1(d) represent the amorphous and crystalline constituents, respectively. From the result we concluded that the periodic structure is alternation between crystalline and amorphous phases.

We also confirmed that overwriting of periodic structure with different polarization direction is possible. Fig. 2(a) is an amorphous mark formed by 100 pulse irradiation of vertically polarized light. The amorphous mark in Fig. 2(b) was obtained by irradiating another 100 pulses with horizontal polarization onto the amorphous mark in Fig. 2(a). The direction of periodic structure completely altered from vertical to horizontal direction. The demonstration of overwriting implies that the formation of periodic structure does not involve any ablation.

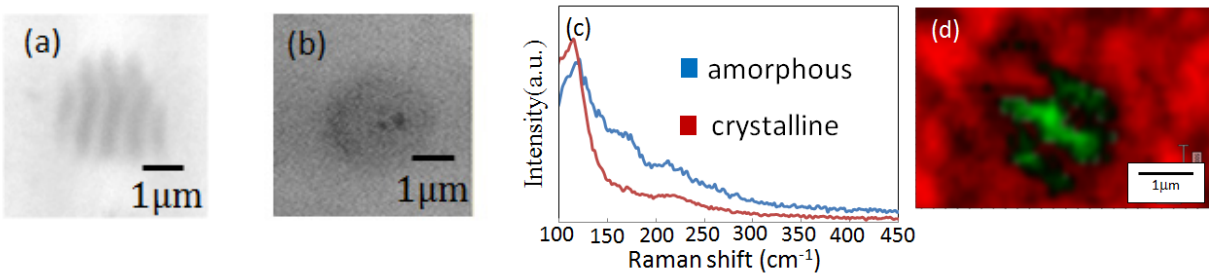

Fig. 1 Scanning laser microscopy images of amorphous marks of (a) GT and (b) GST225. (c) Micro-Raman spectra of crystalline and amorphous phases of GT. (d) Micro-Raman spatial mapping of an amorphous mark of GT (green: amorphous dominant, red: crystalline dominant).

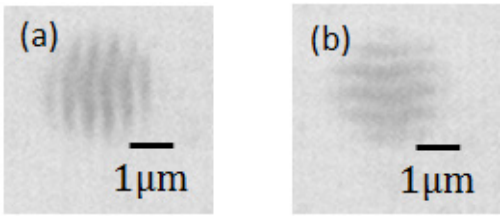

Fig. 2 (a) Amorphous mark of GT induced by vertically polarized 100 femtosecond laser pulses. (b) Additional 100 pulses with horizontal polarization are irradiated onto the amorphous mark in (a).

4. CONCLUSION

A self-organized periodic structure in a GeTe thin film induced by multiple pulse amorphization was investigated. Micro-Raman spatial mapping clarified that the periodic structure was constituted from crystalline and amorphous phases. We demonstrated complete overwriting of periodic structure for the irradiation of two orthogonal polarized pulses.

ACKNOWLEDGEMENT

We are grateful to T. Shintani (Hitachi, AIST) for providing us with GT and GST225 thin film samples. This research was supported by a Grant-in-Aid for Scientific Research (S) from the Ministry of Education, Culture, Sport, Science, and Technology of Japan.

REFERENCES

1. M. Konishi, H. Santo, Y. Hongo, K. Tajima, M. Hosoi, and T. Saiki: “Ultrafast amorphization in $Ge_{10}Sb_2Te_{13}$ thin film induced by single femtosecond laser pulse”, Appl. Opt. **49** (2010) 3470.