Self-organization of a fringe pattern between amorphous and crystalline phases in GeTe induced by femtosecond laser pulse amorphization

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ABSTRACT

By multiple femtosecond pulse amorphization of a GeTe thin film, we formed a self-organized fringe pattern in a single amorphous mark. A micro-Raman measurement unveiled that the fringe is a periodic alternation between crystalline and amorphous phases. The period of fringe is smaller than the wavelength of irradiation light and the direction is perpendicular to the polarization. For a $Ge_2Sb_2Te_5$ thin film, however, we obtained homogeneous amorphous marks and did not see similar fringe pattern even over a wide range of excitation conditions. This probably originates from difference in threshold properties in both nonthermal amorphization and thermal crystallization.

Key words: femtosecond laser pulse, self-organization, nonthermal process

1. INTRODUCTION

GeTe-Sb_2Te_3 pseudo-binary alloys are widely used as phase change materials for commercial products due to their reliability and fast switching speed. We have demonstrated ultrafast nonthermal amorphization of GeSbTe on sub-picosecond time scale using femtosecond laser pulse excitation [1]. Although its microscopic mechanism has not been fully understood yet, the nonthermal phase change process leads to a steep threshold behavior, which is advantageous for a variety of storage and computational applications. In this study we came to the idea that the collaboration of the threshold behavior with distinct contrast in refractive index between crystalline and amorphous phases will generate a self-organized spatial pattern in a single recording mark even if spatially homogeneous excitation is provided. To demonstrate the scenario we performed high-resolution microscopy and spectroscopy of recording marks for various pulse fluence, pulse number, and polarization. We also compared GeTe and $Ge_2Sb_2Te_5$ (GST225) expecting that differences in the threshold behavior will be visualized.

2. EXPERIMENTS

The sample investigated was a 10 nm thick GT film and a 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. For writing marks the laser beam was focused onto the sample surface using an objective with a numerical aperture (NA) of 0.13 for GT and 0.25 for GST225. A scanning laser microscopy and micro-Raman imaging spectroscopy were performed to analyze the constituents of spatial pattern.

3. RESULTS & DISCUSSION

Figure 1 shows scanning laser microscopic images of amorphous mark of GT and GST225. These marks were obtained by irradiation of 200 femtosecond laser pulses with a pulse energy of 3.0 nJ (NA 0.13) for GT and 0.7 nJ (NA 0.25) for GST225. While a homogeneous amorphous mark was generated in GST225, a fringe pattern was organized in GT. The period of fringe is 570nm, which is smaller than the wavelength of irradiation light. The direction of fringe is always perpendicular to the polarization. Figure 2(a) shows micro-Raman spectra of crystalline and amorphous phases. The spatial mapping of these two components in Fig. 2(b) indicates that the fringe is a periodic alternation between crystalline and amorphous phases. We also confirmed that no ablation took place.

To discuss the mechanism of fringe formation, we observed recording marks obtained with different irradiation pulse numbers (Fig. 3). Starting from a rather homogeneous and small amorphous mark, the crystalline stripe grows from the edge to the inside, and at the same time the amorphous stripe grows outside. A FDTD simulation exhibits that for both crystalline and amorphous stripes the electric field locally and slightly increases at their edges (boundary between crystalline and amorphous phases). This field distribution and threshold behavior in both nonthermal amorphization and thermal crystallization contribute to the self-organization of fringe structure. The period of fringe is probably determined by the interference between scattered and incident light fields according to the discussion on the laser induced periodic structure formation, which is usually accompanied by ablation of materials.

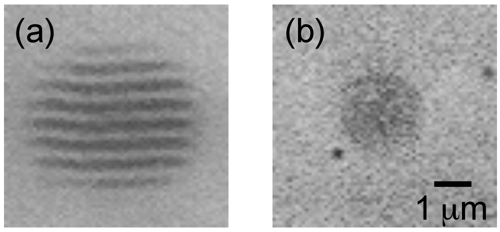

Fig. 1 Scanning laser microscopy images of amorphous marks of (a) GT and (b) GST225.

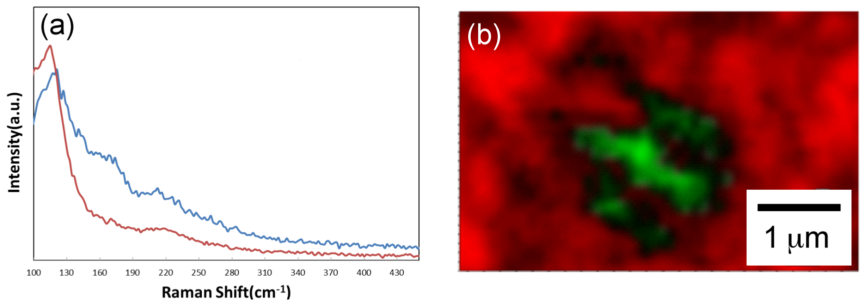

Fig. 2 (a) Micro-Raman spectra of crystalline (red) and amorphous (blue) phases. (b) A micro-Raman mapping of an amorphous mark (green: amorphous dominated, red:crystalline dominated).

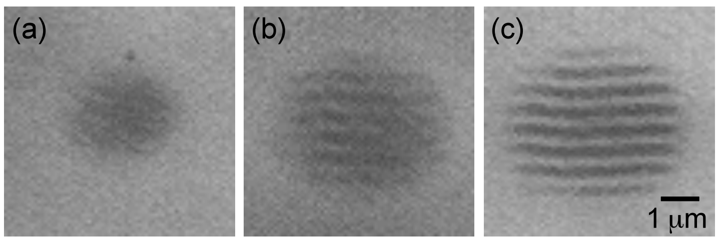

Fig. 3 Fringe patterns obtained with different irradiation pulse numbers. (a) single, (b) 30, and (c) 200 pulses.

4. CONCLUSION

A self-organized fringe structure in a GeTe thin film induced by multiple pulse amorphization was investigated. A localized electric field distribution due to the distinct refractive index contrast between crystalline and amorphous phases is a trigger for inhomogeneous amorphous mark formation. The scattered field due to the inhomogeneity and the incident light interferes and generates neighboring crystalline stripes. The threshold switching of GeTe provides a positive feedback mechanism for the growth of both crystalline and amorphous stripes in the direction of irradiation light polarization. The comparison between GeTe and $Ge_2Sb_2Te_5$ must be informative to understand the difference in threshold properties.

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