Ultrafast phase change in $Ge_2Sb_2Te_5$ superlattices monitored by coherent phonon spectroscopy

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ABSTRACT

We demonstrate in GST superlattice that the phase change from amorphous into crystalline states can be manipulated within only a few cycles of local lattice vibration (≈ 1 picosecond) by coherent excitation of the local lattice vibration using a pair of femtosecond laser pulses.

Key words: coherent phonon, femtosecond, superlattice, nonthermal phase change.

1. INTRODUCTION

A question arising from the dynamics of the phase change in $Ge_2Sb_2Te_5$ (GST) is how fast the phase transformation between the amorphous and the crystalline phases occurs [1]. Motivated by understanding the mechanism of the rapid phase change, extensive investigations on GST have been carried out using electrical and optical measurements, and the *ab initio* molecular dynamics and first principle simulations. Coherent phonon spectroscopy (CPS) is a powerful tool to study the ultrafast dynamics of structural phase transitions occurring on pico- and femtoseconds time scales. In the CPS, a pump pulse impulsively generates coherent lattice motion through impulsive stimulated Raman scattering. To investigate the dynamics of the phase transition in GST alloy films, Först *et al*. applied CPS and found that the appearance of the phonon modes was significantly modified upon structural change [2]. However, ultrafast control of coherent phonons and subsequent phase change was not examined. Here, we have used a femtosecond pump-pulse pair to demonstrate ultrafast switching of GST superlattice (SL) from the amorphous to the crystalline states. The phase change occurred within only a few cycle of the coherent motion of a $GeTe_4$ local vibration, indicating that the phase-change induced by the pump pulse-pair is a nonthermal process.

2. EXPERIMENTS

We fabricated GST SL thin films, which consist of the GeTe and the Sb_2Te_3 layers on silicon (100) substrate by using a helicon-wave RF magnetron–sputtering machine [3]. GST alloy films were also deposited on silicon (100) substrate as a reference sample. To measure time-resolved reflectivity change ($\Delta R/R$) of the sample as a function of the time delay, 20 fs-width optical pulses (λ = 850 nm) from a Ti:sapphire laser was utilized. A pair of pump-pulses was generated through a Michelson-type interferometer, in which the time interval (Δt) of the double pump-pulses was precisely controlled by moving a piezo stage.

3. RESULTS & DISCUSSION

Figure 1 shows the time-resolved $\Delta R/R$ signal observed in GST alloy films with amorphous (as-grown) and crystalline (annealed) phases at RT. After the transient electronic response due to the excitation of photo-carriers near, coherent phonon oscillations with a few picoseconds dephasing time appear. Fourier transformed (FT) spectra are shown in the inset of Fig. 1, in which the two broad peaks are observed at 4.70 THz and 3.66 THz in amorphous film, while a sharp peak at 3.66 THz and a broad weaker band at ≈ 4.9 THz are observed in crystalline film. These peaks in the amorphous film can be considered to be the A_1 optical mode due to tetrahedral $GeTe_4$ structure for the

3.66 THz peak [2,4], and the A_1 optical modes due to Sb_2Te_3 sub-lattice for the 4.70 THz peak [4]. Notice that the dephasing time of the coherent A_1 mode is longer in the case of the crystalline film.

Figure 2 presents the results of the double-pump-pulse excitation obtained from an amorphous GST SL film with the time interval Δt = 273 fs, while the fluences of the first and second pump-pulse were fixed at 76 and 64 μJ/cm^2, respectively. After the double-pump pulse excitation, the dephasing time (τ) of the coherent A_1 optical mode increases from 750 fs to 830 fs. More importantly, when the time interval Δt was scanned at the constant pump fluences, the frequency of the A_1 mode due to $GeTe_4$ structure, shifted from its original value (Ω_A = 3.83 THz) to that of the crystalline phase ($\Omega_C \approx$ 3.67 THz) as Δt changed from 141 to 276 fs. We attribute the frequency shift, from Ω_A to Ω_C, to the ultrafast crystallization. The results suggest that the ultrafast phase change was induced by the coherent excitation of the local A_1 mode. A possible mechanism of the phase change observed by the irradiation of the femtosecond pump-pulse pairs is the umbrella-flip of Ge atoms from octahedral ($GeTe_4$) into tetrahedral ($GeTe_6$) Ge-coordinations [5].

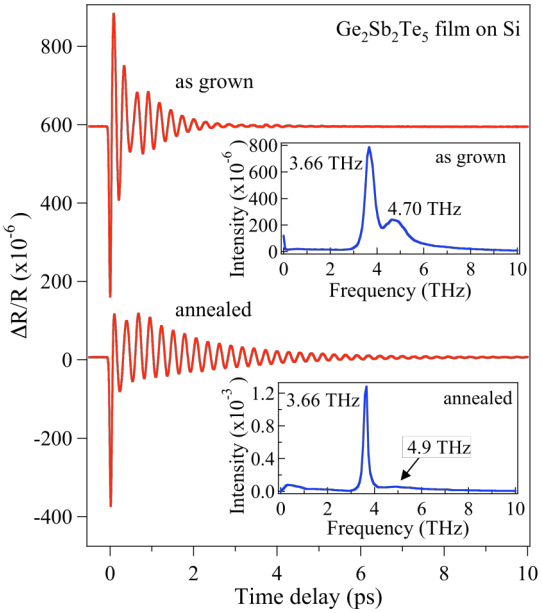

Figure 1. The $\Delta R/R$ signal observed in amorphous (as-grown) and crystalline (annealed) GST films at 295 K. The insets represent their FT spectra.

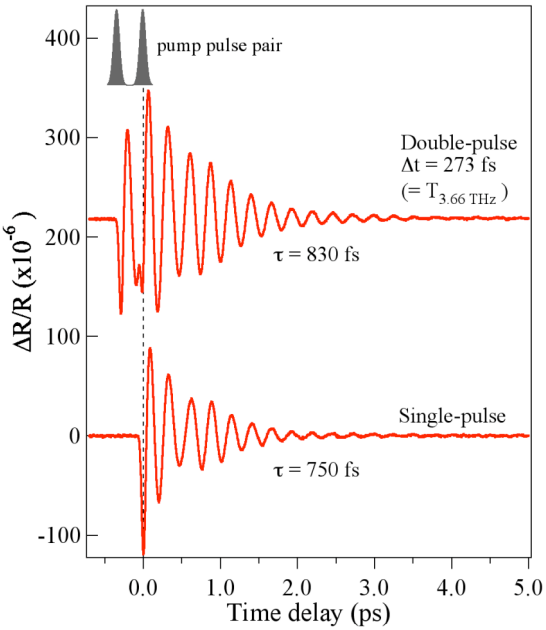

Figure 2. $\Delta R/R$ signal observed by the single and double pump excitation in amorphous GST SL.

4. CONCLUSION

We observed ultrafast phase change from amorphous into crystalline phases in $Ge_2Sb_2Te_5$ SL films by the irradiation of a pair of femtosecond pump-pulses.

REFERENCES

1. J. Hegedüs, S. R. Elliott, *Nature Mat.* **7**, 399 (2008).
2. M. Först, T. Dekorsy, C. Trappe, M. Laurenzis, H. Kurz, and B Bèchevet. *Appl. Phys. Lett.* **77**, 1964 (2000).
3. J. Tominaga, P. Fons, A. V. Kolobov, T. Shima, T. W. Chong, R. Zhao, H. K. Lee, and L. Shi, *Jpn. J. Appl. Phys.* **47**, 5763 (2008).
4. M. Hase, Y. Miyamoto, J. Tominaga, *Phys. Rev. B.* **79**, 174112 (2009).
5. A. V. Kolobov, P. Fons, A. I. Frenkel, A. L. Ankudinov, J. Tominaga, and T. Uruga, *Nature Mat.* **3**, 703 (2004).

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