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Ultrafast dynamics and control of phase change in Ge-Sb-Te materials by the excitation of coherent phonons

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ABSTRACT

There is a possibility of manipulating the rapid phase change in chalcogenide in ultrafast time scale when using femtosecond laser pulses. Especially, coherent control of local vibrations, whose atomic motion plays an important role in the rapid phase change, has been considered to be one of the intriguing approaches. We demonstrate in Ge_2Te_2/Sb_2Te_3 superlattice that the phase change (or precursor of phase change) from crystalline into amorphous states can be observed within a few picoseconds by coherent phonon spectroscopy. Non-thermal nature of the phase change in superlattice and conventional $Ge_2Sb_2Te_5$ alloy will be discussed.

Key words: Coherent phonon, Femtosecond, Nonthermal phase change, GeSbTe.

1. INTRODUCTION

Multi-component chalcogenides, such as Ge-Sb-Te and Ag-In-Sb-Te, are potentially used in optical data storage media in the forms of rewritable CDs, DVDs, and Blu-ray discs. Especially, $Ge_2Sb_2Te_5$ (GST) has proven to be one of the highest-performance alloys among commercially available phase-change materials. A question arising from the dynamics of the phase change in $Ge_2Sb_2Te_5$ 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 [2-4].

Coherent phonon spectroscopy (CPS) is a powerful tool to study the ultrafast dynamics of 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 [5-7]. 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 [8]. We have so far used a weak femtosecond pump-pulse pair (< 100 μJ/cm^2) to demonstrate ultrafast switching of Ge_2Te_2/Sb_2Te_3 superlattice (SL) from the amorphous to crystalline states [9]. In addition, bond-selective excitation in Ge_2Te_2/Sb_2Te_3 superlattice has been examined by tuning the pump polarization of the single pump pulse with the similar weak fluence [10]. However, little is known by coherent phonon spectroscopy about the ultrafast process from the crystalline to amorphous states because one can imagine the potential in the crystalline states is more deep and stable than the amorphous [11] and therefore at least several mJ/cm^2 fluences will be needed to induce such reverse process.

Here we would discuss a possibility of observation of a metastable state in photoexcited crystalline GST-SL and coherent control of the phase change on the bases of coherent phonon excitation with more than 10 mJ/cm^2 fluences.

2. EXPERIMENTAL

The sample used were $Ge_2Sb_2Te_5$ (GST) alloy film and Ge_2Te_2/Sb_2Te_3 SL thin films, which consist of the GeTe and the Sb_2Te_3 layers on silicon (100) substrate by grown by using a helicon-wave RF magnetron–sputtering machine [9,12]. 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 (τ), 40 fs-width optical pulses (λ = 800 nm) from a Ti:sapphire regenerative amplifier system were utilized. A pair of the pump-pulses was generated through a Michelson-type interferometer, in which the separation time (Δt) of the double pump-pulses was precisely controlled by moving one arm of the mirror.

3. RESULTS & DISCUSSION

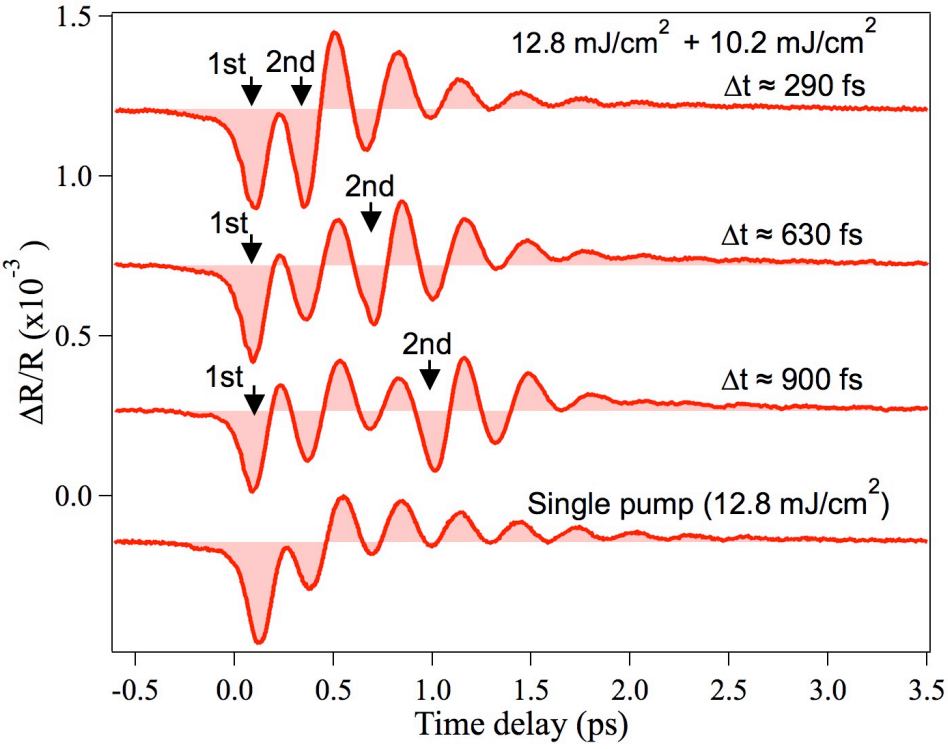

Figure 1. The $\Delta R/R$ signal observed in crystalline GST-SL film at three different separation times (Δt) of the double pump-pulse.

Figure 1 shows the time-resolved $\Delta R/R$ signal observed at room temperature in Ge_2Te_2/Sb_2Te_3 SL films in the crystalline (annealed) phase. After the transient electronic response due to the excitation of carriers ($\tau = 0$), coherent phonon oscillations with a few picoseconds relaxation time appear in the case of the single pump pulse excitation. When the second pump pulse is applied, the relaxation time of the coherent phonon significantly changes depending on the separation time (Δt). Fourier transformed (FT) spectra taken in crystalline state with single pump excitation (15 mJ/cm^2) exhibit one sharp peak at ≈ 3.5 THz, which slightly red-shifts from the peak position under lower photo-excitation [9]. Most importantly, the FT spectra taken only from the time-domain data (Δt = 290 fs) after the 2nd pump arrival explore the double peak structure peaking at ≈ 2.7 THz and ≈ 3.6 THz. The new peak at 2.7 THz is not due to the appearance of the amorphous phase, but would be a new local structure or bond softening being observed under high density photo-excitation. Strong lattice anharmonicity in the crystalline phase has been confirmed by thermal properties studies [11]. The main peak at 3.5 THz blue-shifts by 0.1 THz. This blue-shift may imply the amorphous phase appear under the strong photo-excitation. The separation time of Δt = 290 fs (3.4 THz) nearly corresponds to the time period of the coherent A_1 mode in the crystalline phase, so the excitation by such second pump pulse excite the lattice coherently. This case can be explained by the umbrella flip model of the phase change [13], where the local atomic arrangement accompanying phonon motion is crucial to switching the phase.

4. SUMMARY

We have observed a possibility of ultrafast phase change from crystalline into amorphous phases in Ge_2Te_2/Sb_2Te_3 SL films by the irradiation of a pair of femtosecond pump-pulses. To confirm such ultrafast phase change, time-resolved X-ray diffraction measurements have been carried out at SACLA in SPring-8 using 10-fs XFEL, although such measurements are still underway.

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