Selective Excitation of Ge-Te Bond by Linearly-Polarized Femtosecond Pulse in GeTe/Sb$_2$Te$_3$ Superlattice

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Improvement of operation speed of phase change memory technology is the present-day important research subjects. In order to realize ultrafast phase change in Ge$_2$Sb$_2$Te$_5$ (GST), so far, we have demonstrated the optical manipulation of atomic arrangements in GST superlattice (SL) [1] by selectively exciting the phonon mode of the GeTe$_4$ local structure using femtosecond pump-pulse pair [2]. This result indicates that non-thermal coherent manipulation of a specific phonon mode can provide fast phase change without melting and nucleation process. During the phase change from disordered (amorphous) to ordered (crystalline) state, in our phase change model of GST-SL, Ge atom displaces along [111] direction from tetrahedral-like to octahedral-like sites accompanied by breaking of one of the four Ge-Te bonds which is weaker than other three bonds. From this point of view, selective Ge-Te bond weakening induced by bonding–antibonding carrier excitation may become a potential approach for the realization of ultrafast phase change with lower power consumption.

We utilized a 20-fs near-infrared optical pulse (1.46 eV) to excite and observe coherent local phonons at room temperature. The reflectivity change was recorded as a function of the time delay between the pump and probe pulses. The incident angles of the pump and probe beams from the sample normal were set to 35° and 30°, respectively. The polarization angle of the pump beam was varied from 0° (s-polarization) to 180° (s-polarization) via 90° (p-polarization), while that of the probe was fixed at 90°, as shown in Fig. 1. A sample investigated was as-deposit thin film (20 nm thick) of GST-SL alternatively deposited 0.5-nm-thick GeTe and 0.5-nm-thick Sb$_2$Te$_3$ sub-layers on a Si (100) substrate. In our sample, weaker Ge-Te bond primarily has [111] directional component. The [111] direction is corresponding to the sample normal direction, i.e., p-polarized pulse would excite the weaker Ge-Te bond preferentially.

Fig. 1. The experimental configuration of our bond-selective coherent phonon measurement.
Fig. 2 shows the phonon frequency of the coherent $A_1$ mode of the GeTe$_4$ local structure as a function of pump polarization angle. We found that the p-polarized pump pulse (90°) induce the frequency down-shift relative to the s-polarized (0° and 180°) pulses. Note that the magnitude of the down-shift (0.05 THz) is smaller than the previously reported values in the crystallization by both thermal [3] and nonthermal [2] process. We would interpret the down-shift observed by the p-polarized pump pulse to the displacement of Ge atoms toward tetrahedral position as we intended.

For a better understanding of our results, we would like to argue the excitation of carriers from bonding into antibonding states. Under our experimental condition of above gap excitation, the p-polarized pulse would introduce transient anisotropic carrier distribution and weaken the weaker Ge-Te bond preferentially [4]. Then the equilibrium position of Ge atom shifts toward octahedral site, which is thought to be a precursor of the phase change.

REFERENCES


BIOGRAPHY

Kotaro Makino is a doctoral student of institute of applied physics, University of Tsukuba and a Research Fellow of The Japan Society of the Promotion of Science. He received the B.S. and M.S. degrees in applied physics from University of Tsukuba. His current research field is ultrafast coherent phonon spectroscopy and optical coherent manipulation of phase change recording media.