Ultrafast Nonequilibrium Carrier Dynamics of 2D Materials Measured by Time Resolved THz Spectroscopy

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***Abstract*—In this paper, we will discuss the nonequilibrium ultrafast carrier dynamics in 2 dimensional phase change material $\{Sb(3)Te(9)\}_n$ thin film. The evolution of the optical properties is measured by THz-TDS and OPTP during the phase change from amorphous to crystalline states.**

I. INTRODUCTION

Chalcogenide based compound, especially Sb_2Te_3 and related compounds, exhibit pronounced structural and optical contrast with rapid phase transition from amorphous to crystalline phase. This makes them suitable candidates for rewritable optical storage media and phase change random access memory. Simultaneously, Sb_2Te_3 is reported to be one of the best p-type thermoelectric materials at room temperature, and has topological insulator property. Since electrons and phonons play a crucial role in determining the performances of any real devices, a better understanding of the transition mechanism from amorphous phase to crystal phase is imperative. Terahertz time-domain spectroscopy and optical pump-THz probe (OPTP) spectroscopy would give valuable information on the characteristics of chalcogenide compound.

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II. RESULTS

The multilayered $\{Sb(3)Te(9)\}_n$ thin film was deposited on Si (100) substrate in an ultrahigh vacuum thermal evaporator system. The crystallinity and orientation of the antimony telluride thin films were investigated by XRD and TEM measurement. While increasing the annealing temperature, the structure of the as-grown sample was totally changed, which means that the amorphous phase was changed to rhombohedral Sb_2Te_3 crystal phase. All observed peaks show that the grown films have a single phase of Sb_2Te_3 rhombohedral crystal structure with lattice parameters a=4.26 Å, c=29.9 Å. XRD intensity of the thin films is enhanced as the annealing temperatures increased, indicating that the crystallinity of the thin films becomes higher. To further probe the phase transitions of the as-grown $\{(Sb(3)Te(9)\}_n$ thin films during the annealing process, resistivity and mobility as a function of annealing temperature were acquired by Hall coefficient measurement.

A Ti:sapphire regenerative amplifier system (Micra-Legend Elite, Coherent Inc.) produces a 1 kHz pulse train of 2.5 mJ and 800 nm pulses with a pulse duration of 120 fs (FWHM). The second harmonic (400nm, 3.1 eV) of an amplified laser through a type-II β-Barium Borate (BBO) crystal was used as the pump beam. THz probe pulse generation occurs via optical rectification of the fundamental pulse in a 10x10 mm <110> ZnTe crystal with a thickness of 1 mm. The transmitted THz radiation was detected by means of an electro-optic (EO) sampling method in another 3 mm thick <110> ZnTe nonlinear crystal. The signal was collected with a lock-in amplifier (Stanford Research System, SR830) phase-locked to an optical chopper which modulates either the THz generation line for time domain spectroscopy (TDS) or the pump beam for time resolved THz spectroscopy (TRTS). After ultrafast optical pulses excite the sample, the transient change of the sample in the 0.2-2.6 THz frequency range was probed by THz pulses via EO sampling. The time evolution of the pump-probe signal was collected by scanning the time delay of the pump pulses with respect to the THz pulse. (Fig. 1) All pump-probe experiments carried out in this study were performed at room temperature and the entire THz beam path was enclosed and purged with dry air. Sample integrity was checked before and after the pump-probe experiments by measuring the THz-TDS signal to verify negligible sample degradation.

We investigated the relation between structural phase transition and the evolution of the optical properties in THz range by THz-TDS and OPTP and studied the ultrafast carrier dynamics in $\{Sb(3)Te(9)\}_n$ thin film. Only crystal phase shows the rise time component. The origin of the rise time, fast/slow decay time at each annealing temperature will be discussed. (Fig. 2)

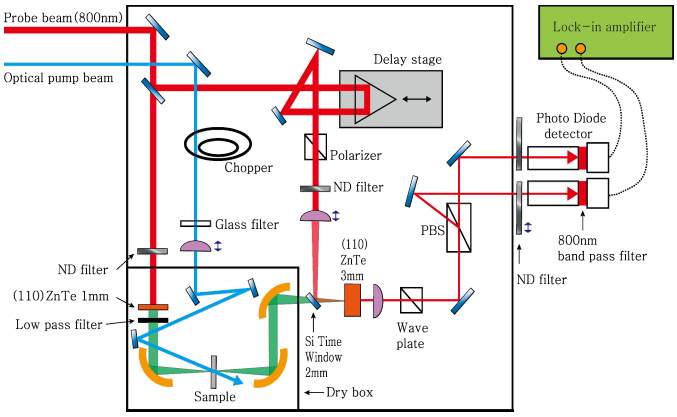

Fig.1. THz Time Resolved Spectroscopy set up

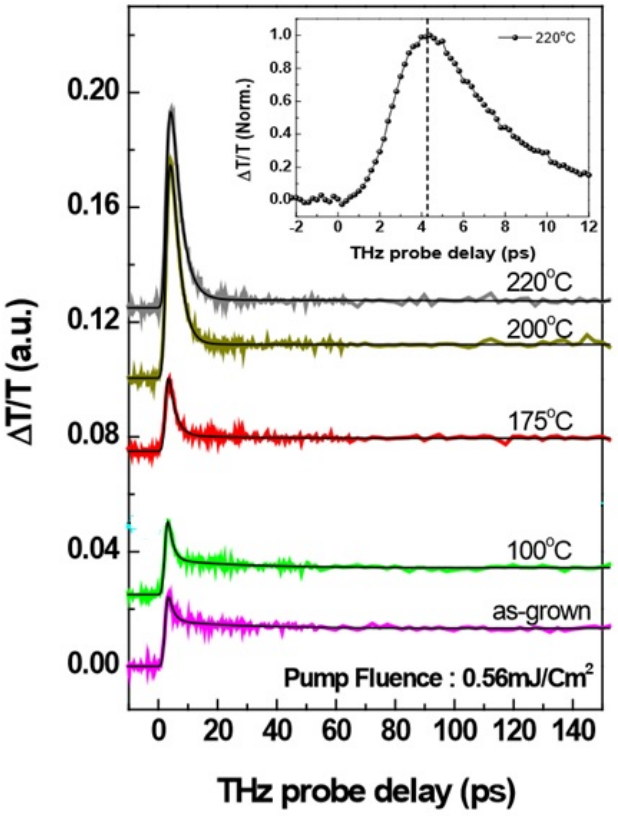

Fig.2. Time-resolved transmittance change from the amorphous phase and crystalline phase sample at a fluence of 0.56 mJ/cm^2.

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