Ultrafast Phonon Dynamics in Few-quintuple layer Topological Insulator Sb_2Te_3

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Abstract: We found few-quintuple layer Sb_2Te_3 presents the in-plane coherent oscillation (E_g^2 mode) using time-resolved transmission measurements, which is different from the results, out-of-plane oscillations (A_{1g}^1 and A_{1g}^2 modes) obtained in bulk.

OCIS codes: (320.7120) Ultrafast phenomena, (320.7150) Ultrafast spectroscopy

1. Introduction

Time-domain spectroscopy is powerful tool to observe the phonon properties in comparison with the conventional static methods such as ultraviolet-infrared and Raman spectroscopies. Coherent phonons are usually generated by irradiation with ultrafast laser pulses on the sample, which can be detected using the time-resolved methods as the modulation in transient reflectivity or transmission in electric susceptibility [1,2]. Here, we applied this time-resolved reflectivity and transmission methods to topological insulator Sb_2Te_3, which atomic arrangement can be considered as repeating units with each consisting of five atomic Te-Sb-Te-Sb-Te layers called quintuple layers. The binary compounds such as Sb_2Te_3, Bi_2Se_3 and Bi_2Te_3 have attracted much interest because of their unique properties as topological insulators. Topological insulator is a new quantum phase of matter, which has a band gap in bulk while conductivity on their edge and surface. The gapless surface states have confirmed using by angler-resolved photoelectron spectroscopy [3], scanning tunneling microscopy and spectroscopy [4]. However, there is little known about their phonon vibration dynamics [5]. In this context, what we observed is that the photo-excited coherent phonon modes on surface of Sb_2Te_3 are different from those in bulk. To investigate these phonon dynamics would be important for revealing the interesting properties of the topological insulators.

2. Experimental details

The femtosecond time-resolved optical pump-probe measurements were performed at room temperature and in the air. The optical pulses were generated from a Ti:sapphire mode-locked oscillator at the center wavelength of 810 nm (~1.5 eV), ~ 40 fs laser pulse duration and ~ 86 MHz repetition rate. The output of the laser was supplying an average power of 20 mW for the pump beam and 1.0 mW for the probe beam. The pump beam is circular polarized to spin-selectively excite the valence band electrons at the surface and the probe beam is linear polarized. The spot sizes of the pump and probe beams were ~86 μm diameter at the overlap of the two beams on the sample. The change of temporal evolution of the reflection or transmission change was measured by scanning the probe time delay (t) repetitively at 20 Hz with a fast-scan mechanical stage. The samples were single crystals grown by a modified Bridgeman technique [3]. The ultrathin-film is prepared to exfoliate the single crystal sample with an adhesive tape. The transmission of this ultrathin-film was measured to be 90% at wavelength of 810 nm. Using the reported refractive index (5.7–3.3i) at the wavelength of 810 nm [6], the thin-film thickness was estimated to be less than 3 nm, which corresponds to < 3 quintuple layers.

3. Results and discussion

The coherent phonon oscillations appeared in time-resolved reflectivity in bulk sample. Figure 1 (a) shows the phonon oscillations of Sb_2Te_3 with the inset of Fast-Fourier transformed (FFT) spectrum. These coherent phonon oscillations at the frequencies of 2.0 THz and 4.9 THz were identified as the out-of-plane A_{1g}^1 and A_{1g}^2 modes of Sb_2Te_3, respectively [7]. Therefore, the transient reflectivity change ($\Delta R(t)/R$) can be fit with following two damped oscillations:

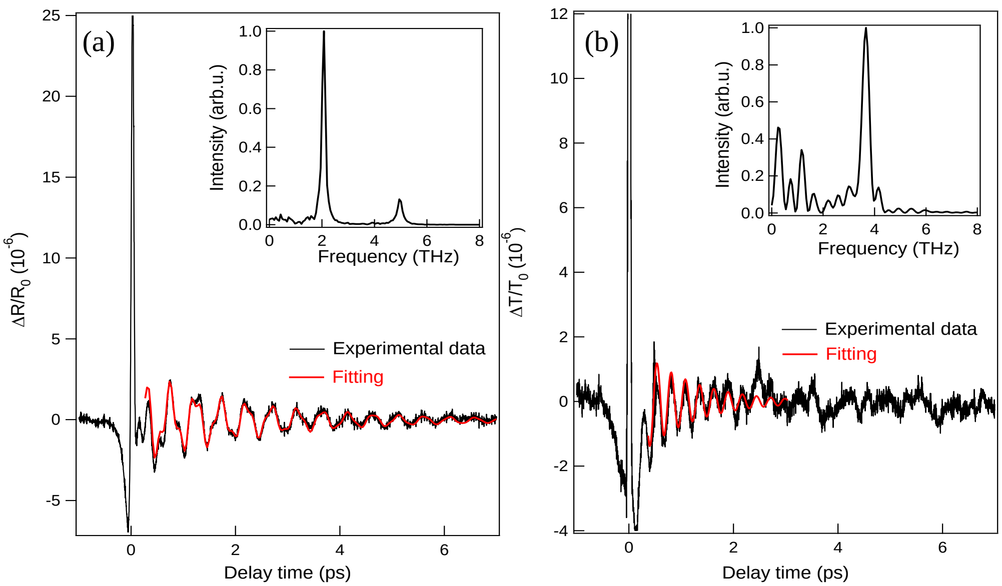

Fig .1 (a) The transient reflectivity measurement of Sb_2Te_3 bulk sample with inset of the Fast-Fourier transformed (FFT) spectrum, (b) shows those of the thin-film.

$$\frac{\Delta R(t)}{R} = \sum_{i=1,2} A_i \exp(-t/\alpha_i)\sin(\omega_i t - \theta_i) \tag{1}$$

where A_i is an amplitude, α_i is the lifetime, ω_i is the frequency, and θ_i is the initial phase. The observed frequencies from the transient reflectivity measurements were well agreed with those from Raman spectroscopy. The lifetime of A_{1g}^1 and A_{1g}^2 modes is estimated to be 2.5 ps and 1.6 ps, respectively. Figure 1 (b) shows the transient transmission from the Sb_2Te_3 ultrathin-film with inset of the FFT spectrum. The oscillation shown in Fig.1 (b) corresponds to in-plane vibrational mode (E_g^2 mode) at the frequency of 3.7 THz [7] with the lifetime of 1.5 ps. Generally it requires special method, i.e. electro-optic sampling technique [5] to observe the in-plane vibrational modes. However, we could observe the in-plane mode of coherent phonon by exfoliating topological insulator bulk to a few quintuple layered ultrathin-film. This result suggests the different physical properties between the surface and bulk of topological insulators.

4. References

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