Surface Optical and Bulk Acoustic Phonons in the Topological Insulator, Bi$_2$Se$_2$Te

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We explore the phonon dynamics of thin films of the topological insulator material Bi$_2$Se$_2$Te using ultrafast pump-probe spectroscopy. The time resolved differential reflectivity of the films exhibit fast and slow oscillations. We have given a careful analysis of variation of phonon frequency as a function of film thickness attributing this to existence of standing acoustic modes. However, no variation in the frequency of the optical phonon modes was found with film thickness. This indicates that the optical phonons intrinsically belong to the surface of the topological insulators. The fact that the acoustic phonons can be tuned by changing the film thickness has tremendous potential for room temperature low power spintronic devices and in topological quantum computation.

The newly discovered 3D topological insulators (TIs) represent a class of exotic quantum state of matter with an insulating bulk and conducting surface states [1],[2]. These conducting surface states exhibit a relativistic energy-momentum dispersion owing to the nontrivial Z$_2$ topology of the bulk electronic wave-function[3]. As a consequence, these surface states form a Dirac cone between bulk valence band and bulk conduction band which has been confirmed by angle resolved photo-emission spectroscopy (ARPES) studies [4],[5]. Additionally, the surface states are “topologically” protected from back-scattering. Helical spin texture due to high spin-orbit interaction, a $\pi$ Berry phase and the presence of odd number of Dirac cones are responsible for this topological protection. Furthermore, the constraint of time reversal symmetry ensures that the surface electrons are not scattered from non-magnetic impurities and defect states. Apart from these TI materials also exhibit spectacular magneto-optical effects which arise due to the presence of an axionic term [6]. These exciting properties have made TIs a subject of intense research both theoretically and experimentally over the past few years. TIs also promise important practical applications in quantum computation and low power spintronic device applications [7].

The family of Bismuth based compounds including Selenides and Tellurides are well established examples of TI materials[8]. Previously, ultrafast pump-probe spectroscopy has been employed to study the phonon and carrier dynamics of Bi$_2$Se$_3$ (BS) [9],[10] and Bi$_2$Te$_3$ (BT)[11]. The differential reflectivity measured by pump-probe spectroscopy exhibit fast and slow oscillations which have been attributed to surface optical and bulk acoustic phonons respectively [12]. However these claims have not been conclusively substantiated. In this letter we reinvestigate these claims through similar single coloured optical pump-probe spectroscopy in reflection geometry across TI samples of different thickness and using a range of laser fluence. Most of the existing research has been carried out on BS and BT systems which have dominant bulk carriers through defect states overwhelming the surface conduction [13]. In our work we have used Bi$_2$Se$_2$Te (BST) thin films which overcome bulk conduction owing to reduced defect states [14]. Moreover BST is also predicted to have a large spin texture giving rise to non-trivial Berry phase that prevents back-scattering [14],[15]. To support this claim variation of the resistance in temperature has been shown in the supplementary material clearly capturing bulk insulating feature. To the best of our knowledge this is the first time a full pump-probe signal with both fast and slow oscillation has been observed for a BST thin film.

For our experiments, BST thin films have been deposited on Silicon (100) substrates using pulsed laser deposition (PLD) technique. Samples of thickness 50 nm, 100 nm and 300 nm were prepared in identical conditions by varying the number of laser pulses. Further details of the BST sample preparation have been reported elsewhere [16]. More information on the characterisation of the thin films are provided in the supplementary material. An ultrafast femtosecond pulsed laser (MANTIS from Coherent Inc. USA) was used to generate Gaussian pump and probe pulses of 780 nm center wavelength and 100 fs pulse-width with a repetition rate of 80 MHz. The pump beam was modulated at 1.8 KHz using a mechanical chopper and the change in reflectivity of probe beam was measured using a lock-in amplifier as a function of pump-probe delay. Details of the experimental setup is reported in ref.[17]. Polarization of pump and probe pulse were made perpendicular to each other to reduce pump scattering and obtain a signal with low noise. All experiments reported in this letter were performed at room temperature.

The raw data for time resolved differential reflectivity for BST sample is plotted in solid line in fig.1 (inset). A fast rise time indicates rapid transition of electrons from valence to conduction band. The logarithmic plot of the...
signal shows a short lived rapid oscillation superimposed on a rather slow frequency oscillation that lasts for a longer time scale. The fast oscillations last for around 10 ps and show several complete cycles. However, the slower one, observed for around 100 ps barely completes 2 cycles. Measurements were carried up to much larger delay time (800 ps) but no significant modulation was observed after 100-120 ps.

The differential reflectivity in fig.1 is fitted with double decay function to capture the overall carrier relaxation dynamics. In addition, two damped oscillation terms have been added to extract the characteristic frequencies along with the appropriate decay times [18]. The final fit equation is thus given by,

$$\frac{\Delta R}{R} = A_1 \exp \left\{ -(t - t_0)/t_1 \right\} + A_2 \exp \left\{ -(t - t_0)/t_2 \right\} + B_1 \exp \left\{ -(t - t_0)/t_3 \right\} \sin(2\pi f_1 + \phi_1) + B_2 \exp \left\{ -(t - t_0)/t_4 \right\} \sin(2\pi f_2 + \phi_2) + C \quad (1)$$

From the fit the frequencies of fast and slow oscillation are obtained to be 1.93 THz and 0.03 THz respectively. These fast and slow oscillations are attributed to surface optical and bulk acoustic phonon modes respectively [12]. To confirm, Raman scattering spectra of these samples were taken using blue laser (488 nm) of 6 mW power at 50× magnification and peaks are observed at 65.67 cm⁻¹, 109.67 cm⁻¹ and 158.74 cm⁻¹ (see fig.2). Previous literature [19] attributes these peaks to the A₁g Raman mode is 1.97 THz which is in close agreement to the surface optical phonon mode observed in our pump probe measurements. Temporary charge separation and vibration for A₁g Raman mode is shown in inset of fig.2. As the experiment has only been carried out in room temperature, oscillations corresponding to other Raman modes which are expected to be very short lived (reference) are hence not observed.

For a single sample the pump fluence is varied from 15 μJ/cm² to 50 μJ/cm² while maintaining a constant probe fluence of 1.25 μJ/cm². However, neither the fast nor the slow oscillation frequency show any significant variation (see fig.3). This indirectly supports the hypotheses that the two oscillations represent the optical and acoustic phonon modes as change in the pump fluence will only change the carrier density in the conduction band and is not expected to have any effect on either of the phonon modes.

The same experiment was then repeated on identically prepared BST films of thicknesses 50 nm, 100 nm and 300 nm. Any change in the thickness is expected to alter the boundary conditions for the acoustic phonon oscillations. In fig.4 the pump-probe signal for the three different samples are shown along with their fitting as per eqn.(1). The magnitude of differential reflectivity is prominently reduced with increasing thickness.

To illustrate this further, the fast and the slow oscillations...
TABLE I. Observed frequency and damping of the fast and slow oscillations

| Sample Thickness (nm) | Fast Oscillation | Slow Oscillation |
|-----------------------|------------------|------------------|
|                       | Frequency (THz)  | Damping (ps)     | Frequency (THz) | Damping (ps) |
| 50                    | 1.960 ± 0.008    | 1.41 ± 0.03      | 0.0330 ± 0.0008 | 4.79 ± 0.21  |
| 100                   | 1.914 ± 0.022    | 1.40 ± 0.05      | 0.0299 ± 0.0010 | 8.42 ± 0.59  |
| 300                   | 1.917 ± 0.049    | 1.33 ± 0.03      | 0.0149 ± 0.0010 | 25.34 ± 1.03 |

Thickness of the sample also shows a pronounced effect on the damping of the slow oscillation. The damping can be physically attributed to the strain owing to the lattice mismatch between the sample and the substrate. Whereas hexagonal quintuple BST layers have a lattice constant of 4.218 Å[20], Si(100) has a diamond cubic lattice structure with lattice spacing of 5.431 Å. Because of this lattice mismatch, there is a mechanical strain on the BST layers deposited. However, with increasing thickness, the BST films are allowed more degrees of freedom which helps in relaxing this strain, resulting in reduced damping in the slow oscillation. From the fitting of fig. 4, the fast and slow oscillations are extracted and plotted separately in fig. 5. The slow oscillations clearly demonstrate this reduction in damping with increasing thickness. Amplitude of the slow oscillation also decrease with increasing thickness. As mentioned above, the damping of the faster oscillation remains constant for different samples indicating that these are indeed signatures of a surface phenomenon without any bulk contribution. This indicates that the optical phonons corresponding
FIG. 5. Fast and Slow oscillation for different samples as extracted from the fitting of fig.4. (a)-(c): Fast oscillation for 50 nm, 100 nm and 300 nm sample respectively. (d)-(f): Slow oscillation for 50 nm, 100 nm and 300 nm sample respectively.

The ability to tune the frequency of acoustic phonons with thickness can be of advantage. In addition to this the fact that we have managed to separate the contribution of bulk acoustic phonons and surface optical phonons has a bearing on improving the low power spintroninc device application in certain cases without having to cool the system.

In conclusion, the experiment captures the optical and acoustic phonon modes through time resolved differential reflectivity measurements of BST thin films. Short lived fast oscillations, which are superimposed on the pump-probe signal are attributed to the $A_{1g}$ optical phonon mode. The remaining slow oscillations correspond to modulation due to interference of bulk acoustic phonon modes. By studying films of different thickness, we have been able to conclusively establish that optical phonon modes are indeed a surface phenomenon with no contribution form the bulk. The slow oscillation corresponding to the acoustic phonon modes on the other hand is significantly affected by thickness and this has been qualitatively explained in the light of dynamics of mechanical standing waves in elastic media. This ability to tune the frequency of acoustic phonons with thickness has a bearing on improving the room temperature low power spintroninc device applications and in topological quantum computation.

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