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Temperature-dependent ultrafast carrier and phonon dynamics of topological insulator $\text{Bi}_{1.5}\text{Sb}_{0.5}\text{Te}_{1.8}\text{Se}_{1.2}$

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Using ultrafast optical pump-probe technique, we studied the temperature-dependent carrier and phonon dynamics of the topological insulator $\text{Bi}_{1.5}\text{Sb}_{0.5}\text{Te}_{1.8}\text{Se}_{1.2}$ single-crystal from 10 K to 300 K. Two relaxation processes of carriers and coherent optical/acoustic phonons have been observed. By using the two-temperature model, we are able to attribute the fast ($\sim$ ps) relaxation component to carrier-phonon coupling involving carriers in the conduction band. We also studied the temperature dependence of the dephasing time and frequency of optical phonon, and the optical penetration depth of $\text{Bi}_{1.5}\text{Sb}_{0.5}\text{Te}_{1.8}\text{Se}_{1.2}$. © 2014 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4879831>]

Topological insulators have attracted intense interest in recent years due to their importance in serving a test bed for fundamental physics, as well as for their prospective applications in spintronics and quantum computation.¹ Their insulating bulk but conducting surface is derived from the presence of time-reversal symmetry and spin-orbit interaction.² Recently, topological insulators $\text{Bi}_2\text{Te}_2\text{Se}$ and $\text{Bi}_{1.5}\text{Sb}_{0.5}\text{Te}_{1.7}\text{Se}_{1.3}$ (BSTS) have been found,³ where non-stoichiometry-induced donors and acceptors compensate each other, yielding high bulk resistivities of 1–10 Ω cm and high contribution ($\sim$ 70%) to the total conductance from the surface state, thus showing a greater contrast between the bulk and surface resistivities compared to Bi_2Se_3 , Bi_2Te_3 , and $\text{Bi}_2\text{Te}_2\text{Se}$ (Ref. 4) making BSTS a promising candidate for future applications.⁵ The insulating/semiconducting bulk ($\sim$ 0.25 eV) but metallic surface of BSTS makes it become an ideal material for studying new physics as well.

Ultrafast optical pump-probe spectroscopy has been widely used in studying the dynamics of non-equilibrium carriers and phonons in solids^{6–10} and proved to be suitable for studying topological insulators.^{11–17} Some pump-probe studies show that A_{1g} coherent optical phonon has been found in Bi_2Se_3 , Sb_2Te_3 , and Bi_2Te_3 .^{12–15} However, the dynamics of carriers in topological insulators has not been fully explored with optical pump-probe techniques. In this Letter, we report optical pump-probe results of BSTS in the temperature range of 10–300 K. We observe two relaxation processes of carriers and coherent optical/acoustic phonons. By making use of the two-temperature model (TTM), anharmonicity and strain pulse propagation model, we are able to explain the carrier dynamics, temperature dependence of dephasing time and optical phonon frequency, and the optical penetration depth of this topological insulator.

$\text{Bi}_{1.5}\text{Sb}_{0.5}\text{Te}_{1.8}\text{Se}_{1.2}$ single crystals were synthesized by melting high-purity (99.9999%) Bi, Sb, Te, and Se with molar ratio 1.5:0.5:1.8:1.2 at 850 °C in an evacuated quartz

tube.⁵ We took temperature- and fluence-dependent pump-probe data using two laser systems: (1) 250 kHz repetition rate Coherent RegA amplified laser (pump fluence $\sim$ 1 mJ/cm²), and (2) 80 MHz KMLabs Griffin oscillator (pump fluence $\sim$ μJ/cm²).¹⁸

Fig. 1 shows $\Delta R/R$ as a function of pump-probe time delay, at selected temperatures using the low-fluence Griffin laser. We observe a fast rise time of 200 fs following by a combination of non-oscillatory-decaying plus high-frequency-oscillatory signals. The initial fast rise is due to the excitation of carriers. The high-frequency oscillations are weak for low-fluence data ($\sim$ μJ/cm²), hence we will use only the high-fluence data (0.4 mJ/cm²) to study the oscillations later in Fig. 3. After the dip at $\sim$ 5 ps, $\Delta R/R$ rises again. A distinct peak appears at $\sim$ 15 ps (indicated by arrows in Fig. 1)—this is the crest of the low-frequency damped oscillatory signal that was also observed in Bi_2Se_3 and Sb_2Se_3 .^{14–16} Finally, $\Delta R/R$ decreases very slowly with a nanosecond timescale—the result of a long-lifetime decay process.

We fit the relaxation signal (after the first peak) with the following expression:

$$\frac{\Delta R}{R} = \sum_{i=1,2} A_i \exp\left(-\frac{t-t_0}{\tau_{Ai}}\right) + \sum_{j=1,2} B_j \exp\left(-\frac{t-t_0}{\tau_{Bj}}\right) \sin(2\pi f_j t + \phi_j) + C. \quad (1)$$

Here, A_i and τ_{Ai} are the amplitude and relaxation time of the i th non-oscillatory signal, respectively, that describes the carrier dynamics. B_j , τ_{Bj} , f_j , and ϕ_j are the amplitude, dephasing time, frequency, and initial phase of the j th oscillatory signal, respectively, that describes the coherent phonon dynamics. C is a constant offset.

First, we will discuss the relaxation dynamics of carriers. After the fast excitation, the excited carriers start to relax via two processes. The process with shorter relaxation time ($\tau_{A1} \sim$ 1 ps) is attributed to carrier-phonon interaction,

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FIG. 1. Photoinduced change in reflectivity ($\Delta R/R$) versus pump-probe time delay, at selected temperatures, using the low pump fluence Griffin laser ($\sim \mu\text{J}/\text{cm}^2$). Red lines are the fits to Eq. (1). The arrows indicate the first peak of the low-frequency damped oscillation signal at 15 ps.

while the process with longer relaxation time ($\tau_{A2} \sim 1$ ns) is attributed to the diffusion of carriers out of the excitation volume. The latter slow relaxation will not be discussed in this paper.

To understand the dynamics of photoexcited carriers, we examine the bulk and surface contributions to the absorption of the pump pulse by free carriers. Given the surface carrier density $n_s = 4.0 \times 10^{12} \text{ cm}^{-2}$ and bulk carrier density $n_b = 2.3 \times 10^{16} \text{ cm}^{-3}$ from transport measurements,^{3,5} within the excitation volume (50 μm diameter, 27 nm penetration depth derived later, and 2 nm surface layer¹⁹), the number of surface and bulk carriers of BSTS are 7.9×10^7 and 1.1×10^6 , respectively. On the other hand, after taking into account sample reflectance, the number of absorbed photons per pump pulse from the Griffin laser is 5.7×10^7 (for $1.3 \mu\text{J}/\text{cm}^2$ fluence). Hence, almost all the pump photons were absorbed by the free carriers on the surface and in the bulk. Comparing with time-and-angle-resolved photoemission spectroscopy (tr-ARPES) data,²⁰ we conclude that the shorter relaxation process corresponds to the relaxation of photoexcited carriers from higher energy level to the bottom in the conduction band, which is dominated by the electron-phonon coupling.

The timescale of τ_{A1} is consistent with that in metals, where carrier-phonon-interaction-mediated relaxation takes place.⁶ τ_{A1} increases with increasing pump fluence and temperature (Fig. 2). We explain these fluence and temperature dependences using TTM,^{6,21} which was previously used to describe the relaxation dynamics of free carriers in metals. At low-fluence excitation (with Griffin laser), most of the free carriers on the surface and inside the bulk will be excited like in metals. As in other topological insulators, BSTS contains metal-like surface and bulk in-gap impurity states that contribute to the total electrical conduction.³ Therefore, we anticipate the TTM to be applicable for our present scenario. In addition, we make the physically reasonable assumption that the carrier subsystem has already thermalized after 200 fs. This assumption is supported by the pump-probe works on Bi_2Se_3 .^{16,22}

The TTM fittings of τ_{A1} as a function of fluence and temperature are shown in Figs. 2(a) and 2(b), respectively. The

FIG. 2. Fast relaxation time τ_{A1} versus (a) fluence (at room temperature) and (b) temperature (at fluence $1.3 \mu\text{J}/\text{cm}^2$) measured by the low pump fluence Griffin laser. The solid curves are energy relaxation time τ_E from the TTM fitting.

fitting process is described in the supplementary material.¹⁸ From the fittings, we obtained the electronic specific heat coefficient (γ) and electron-phonon coupling constant (g_∞). Fitting fluence-dependent τ_{A1} data (at room temperature) to the TTM yields $\gamma = 6.4 \text{ J m}^{-3} \text{ K}^{-2}$ and $g_\infty = 1.7 \times 10^{15} \text{ W m}^{-3} \text{ K}^{-1}$, while fitting temperature-dependent τ_{A1} data (at fluence of $1.3 \mu\text{J}/\text{cm}^2$) to the TTM yields $\gamma = 3.9 \text{ J m}^{-3} \text{ K}^{-2}$ and $g_\infty = 1.1 \times 10^{15} \text{ W m}^{-3} \text{ K}^{-1}$. These values of γ agree with the γ_p extracted from the heat capacity measurement ($4.7 \pm 2.7 \text{ J m}^{-3} \text{ K}^{-2}$).¹⁸ These results confirm that TTM is able to describe the fast relaxation of carriers under the low-fluence optical excitation.

According to the theory of the thermal relaxation of electrons,²³ the electron-phonon relaxation time of electrons in metals is given by

$$1/\tau_E = 3\hbar\lambda\langle\omega^2\rangle/\pi k_B T_e, \quad (2)$$

where λ is the dimensionless electron-phonon coupling constant and $\langle\omega^2\rangle$ is the second moment of the phonon spectrum. From the measured relaxation time at 300 K, $\tau_{A1} = (1.4 \pm 0.2) \text{ ps}$, we obtain $\lambda\langle\omega^2\rangle = (4.2 \pm 0.6) \times 10^{25} \text{ Hz}^2 = (18.3 \pm 2.6) \text{ meV}^2$. By use of $\langle\omega^2\rangle = 1.4 \times 10^4 \text{ K}^2$ obtained from Debye model,¹⁸ we calculate $\lambda = (0.17 \pm 0.03)$. The value of λ is similar to that of metals such as gold (0.13 ± 0.02) and chromium (0.13 ± 0.02)²⁴ and is consistent with the reported values for Bi_2Se_3 (0.08 – 0.43).^{25–27}

Although TTM provides a good description of electron-phonon relaxation in the low-fluence regime, it fails to explain the short relaxation process for the high-fluence regime ($\sim 1 \text{ mJ}/\text{cm}^2$, measured by RegA laser). First, τ_{A1} decreases with increasing fluence—at odds with TTM, although the values of τ_{A1} in both fluence sets of data are similar (~ 1 ps). Second, the rise time of ~ 700 fs is long compared with that from low-fluence data and of other metals. If we perform a fit to τ_{A1} using TTM, the fitted $\gamma = 7.8 \times 10^2 \text{ J m}^{-3} \text{ K}^{-2}$ is much larger than γ_p . We attribute this disagreement to the electron-hole pair generation and large optical phonon population at high

FIG. 3. Photoinduced change in reflectivity ($\Delta R/R$) versus pump-probe time delay, at selected temperatures, using the high pump fluence RegA laser (~ 1 mJ/cm²). The inset shows the signal up to 10 ps. Red lines are the fits to Eq. (1). The arrows indicate the first peak of the low-frequency damped oscillation signal at 15 ps.

fluence. At high fluence, the majority of the pump photons will be used to excite electrons in the bulk valence band across the band gap (~ 0.25 eV) into the conduction band—this interband transition will generate a large number of electron-hole pairs. The interaction between electrons and holes cannot be accounted for by the TTM. Moreover, the longer rise time could be suggesting that in the high-fluence condition, the population of a large number of electronic excitations may have changed the underlying crystal structure, which influences transiently both the lattice dynamics and the strength of electron-phonon coupling. The stronger the electron-phonon coupling is, the more electronic energy is transferred. This quick energy transfer effectively slows down the energy accumulation of electronic degrees of freedom, giving rise to a longer rise time in the pump probe signal. Furthermore, the emission and reabsorption of optical phonons may also cause a slow buildup of photoexcited carriers and subsequent long rise time in the pump probe signal. Such long rise times have been seen in pump-probe data of bilayer graphene intercalated with FeCl₃,¹⁰ as well as in sulfate-covered gold nanoparticles.²⁸

Therefore, the interesting observation in high-fluence data has opened a regime of non-equilibrium problem, which calls for a development of excited-state electronic structure theory for real materials take into account (1) contributions from existing free carriers in the sample, (2) carriers (electrons and holes) generated by the pump pulse from interband transitions, (3) electron-hole interactions, and (4) emission and reabsorption of optical phonons. Experimentally, advanced techniques such as tr-ARPES are also needed to disentangle the surface and bulk contributions to the pump-probe signal, and ultrafast relaxation of electrons and holes in different bands. These are unfortunately beyond the scope of our paper.

Next, we will discuss the dynamics of coherent phonons. With a higher pump fluence (~ 1 mJ/cm²), the high-frequency oscillations are more clearly visible (Fig. 3). Since the BSTS

sample is opaque at 800 nm, dispersive excitation of coherent phonons¹⁵ should be the mechanism that causes the coherent phonons in our case. We observe both coherent optical and acoustic phonons from our data, which is consistent with the previous pump-probe data on Bi₂Se₃.^{15,16}

In this experiment, the high-frequency damped oscillations before 10 ps have frequency $f_{B1} \sim 1.9$ THz and are attributed to the A_{1g} symmetric coherent optical phonon, commonly seen in other topological insulators.^{11,13,14,16,17} The temperature dependence of dephasing time and frequency are shown in Figs. 4(a) and 4(b) suggesting a phonon-softening with increasing temperature. Such phonon-softening can be explained by anharmonic effects in phonons, which comes from anharmonic terms in vibrational potential energy.^{29,30} With three-phonon corrections in the phonon self-energy, the frequency and dephasing time can be written as^{29,30}

$$\frac{1}{\tau_{B1}} = \gamma_0 + C_1 \left[1 + \frac{2}{\exp\left(\frac{hf_0}{2k_B T}\right) - 1} \right], \quad (3)$$

and

$$f_{B1} = f_0 + C_2 \left[1 + \frac{2}{\exp\left(\frac{hf_0}{2k_B T}\right) - 1} \right], \quad (4)$$

where γ_0 is a disorder-induced damping term, f_0 is the optical phonon frequency at 0 K, and C_1 and C_2 are constants. For the A_{1g} coherent optical phonon, good fits to the data were obtained, with fitting parameters $f_0 = (1.992 \pm 0.002)$ THz and $\gamma_0 = (0.16 \pm 0.01)$ THz, which are similar to the values for Bi₂Te₃ ($f_0 = 1.908$ THz and $\gamma_0 = 0.04$ THz).³⁰

We also show in Figs. 4(c) and 4(d) the temperature dependence of the dephasing time and frequency for the low-frequency oscillations. These low-frequency oscillations in the pump-probe data are commonly caused by coherent acoustic phonons (CAP), often seen in semiconductors and metals, and explained with the strain pulse propagation model.³¹ The incidence of probe beam on the sample generates a layer of strain with different dielectric functions from the unprobed area, which can propagate within the crystal to

FIG. 4. (a) The dephasing time τ_{B1} and (b) frequency f_{B1} of the coherent optical phonon. Solid curves are the fits to anharmonic model described by Eqs. (3) and (4). (c) The dephasing time τ_{B2} and (d) frequency f_{B2} of the coherent optical phonon.

modulate the $\Delta R/R$ signal. According to this model,³¹ the frequency of CAP is given by $f_{B2} = 2nv_s \cos(\theta)/\lambda_{probe}$, where n is the refraction index of BSTS, v_s is the sound speed in BSTS, θ is the angle of incidence of the probe beam ($\sim \pi/12$), and λ_{probe} is the probe wavelength. Using the value of n (~ 5.4) obtained for Bi_2Se_3 ,³² we obtain an average sound speed of $v_s = 3.22 \times 10^3$ m/s in the 10–300 K temperature range. Then, the penetration depth of probe beam can be estimated via the relation $\zeta = v_s \tau_{B2}$ to be 27 nm.³¹ These parameters are consistent with the sound speed of Bi_2Te_3 (2.95×10^3 m/s)³³ and the penetration depth of Bi_2Se_3 (24 nm for $\lambda_{probe} = 800$ nm).³²

In conclusion, we studied the photoinduced change in reflectivity of the BSTS topological insulator to reveal the dynamics of carriers and coherent optical/acoustic phonons, with the information of Debye temperature and electronic specific heat coefficient obtained from thermodynamical measurements. We applied a two-temperature model to explain the fast relaxation process of carriers in the conduction band. The obtained dimensionless electron-phonon coupling constant (λ) is consistent with those of conventional metals and Bi_2Se_3 topological insulator. Anharmonic effects of the A_{1g} coherent optical phonon were used to explain the temperature dependence of the frequency and dephasing time of the high-frequency oscillatory signal. Finally, applying the strain pulse propagation model to the coherent acoustic phonon yields a penetration depth of 27 nm in BSTS for 800 nm laser light.

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¹⁸See supplementary material at <http://dx.doi.org/10.1063/1.4879831> for details about parameters of laser pulses, specific heat measurement of BSTS, the two-temperature-model fitting of relaxation time, and the calculation of phonon spectrum.

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