Gigantic Surface Lifetime of an Intrinsic Topological Insulator

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The interaction between light and novel two-dimensional electronic states holds promise to realize new fundamental physics and optical devices. Here, we use pump-probe photoemission spectroscopy to study the optically excited Dirac surface states in the bulk-insulating topological insulator Bi_2Te_2Se and reveal optical properties that are in sharp contrast to those of bulk-metallic topological insulators. We observe a gigantic optical lifetime exceeding 4 μs (1 μs = 10^{-6} s) for the surface states in Bi_2Te_2Se, whereas the lifetime in most topological insulators, such as Bi_2Se_3, has been limited to a few picoseconds (1 ps = 10^{-12} s). Moreover, we discover a surface photovoltage, a shift of the chemical potential of the Dirac surface states, as large as 100 mV. Our results demonstrate a rare platform to study charge excitation and relaxation in energy and momentum space in a two-dimensional system.

Two-dimensional electrodynamics shows many interesting effects that do not arise in three dimensions [1–15]. Pump-probe photoemission spectroscopy may allow us to directly access many of these two-dimensional phenomena by studying the electronic band structure of a system in an excited state. However, there are currently few experimental systems available where electrons are well confined to move in a plane. Moreover, many of the best-studied two-dimensional electron systems arise at buried interfaces in semiconductor heterostructures and, consequently, are difficult to access by spectroscopy. Meanwhile, graphene has recently offered us a truly two-dimensional electron system [1], but the low-energy excitations are at the corners of the Brillouin zone, and this introduces many challenges in studying two-dimensional electrodynamics by photoemission spectroscopy. Topological insulators (TIs) give rise to surface states, and many known TIs have surface states with low-energy excitations at the center of the Brillouin zone. However, most experimentally studied materials suffer from residual bulk conductivity, which dominates transport phenomena and masks the properties of the surface states [2–7].

In order to measure the optical lifetime of an excited electron, time-resolved angle-resolved photoemission spectroscopy (TRARPES) is a powerful tool, where an intense pulse laser (the pump laser) is applied on the surface of a sample to excite electrons from below to above its chemical

potential, and then a separate laser beam (the probe laser) is used to study the transient electronic structure as a function of delay time with respect to the pump in a momentum-resolved way. Previously, TRARPES has been used to measure TIs, and a quite short optical lifetime of a few picoseconds (10^{-11}–10^{-12} s) for the Dirac surface states was reported [16–22]. Nevertheless, these experiments were performed with bulk metallic TI samples, which means both the surface states and the bulk conduction bands are present at the chemical potential. Therefore, as the pump light is shone, both surface states and the bulk bands were found to be excited and the lifetime for both was observed to be as short as 10^{-11}–10^{-12} s. Such fast dynamics renders the optical control of the surface states a major unsolved challenge in the field.

Here, we report a remarkably long lifetime exceeding 4 μs (1 μs = 10^{-6} s) for the optically excited Dirac surface states. This is achieved by using a truly bulk insulating TI sample Bi_2Te_2Se (BTS221) with bulk resistivity larger than 6 Ω cm [23–28], where only the surface states cross the Fermi level. Surprisingly, we show that the observed long optical lifetime in our BTS221 samples leads to a surface photovoltage (a shift of the chemical potential of the Dirac surface states) as large as ~100 mV, which is also observed to be robust on the time scale of microseconds. Although surface photovoltage has been observed in semiconductors [33,34], we note that our observation of such an effect in a

highly bulk insulating topological insulator is fundamentally different from a semiconductor. Topological insulators consist of spin-momentum locked surface states, whereas in a semiconductor such topological surface states are completely absent and fundamentally impossible because of the topologically trivial nature of the semiconductor system.

Single crystalline samples of topological insulators used in our measurements were grown using the Bridgman method and characterized by spectroscopic and transport methods, which is detailed elsewhere [23–27]. The TRARPES setup at the Institute for Solid State Physics (ISSP) at the University of Tokyo consists of an amplified Ti:sapphire laser system delivering $h\nu = 1.47$ eV pulses of 170-fs duration with 250-kHz repetition and a hemispherical analyzer [28,29]. We first describe the basic experimental setup for TRARPES [Figs. 1(a) and 1(b)] employed in our measurements. Our TRARPES uses a Ti:sapphire laser system, which delivers pulsed light of energy 1.47 eV. Each single pulse is as short as ~170 fs (1 fs = 10^{-15} s), and the pulse is repeated every 4 μs (the repetition rate is 250 kHz). The laser is split into two beams, a pump beam and a probe beam. While the pump laser is applied to the sample directly, the photon energy of the probe laser is quadrupled via two β-BaB$_2$O$_4$ nonlinear optical crystals from 1.47 to 5.9 eV. The delay time between the pump and the probe laser is tuned by a mechanical stage. Because of a length limit of the mechanical stage, the system can only be probed at a limited range of time delays. Specifically, for our experiment, we can probe the transient electronic structure at the delay time of 0 to 400 ps and

4 μs–50 ps to 4 μs. 0 to 400 ps means a time period that is immediately after a pump pulse hit the sample. On the other hand, 4 μs–50 ps corresponds to a time period that is long after a pump pulse hit the sample, which is immediately before the next pulse.

Figures 1(c)–1(f) show ARPES spectra on four different TI samples at representative delay time (DT) values. Specifically, the four samples are two pieces of BTS221, one (Bi$_{0.2}$Sb$_{0.8}$)$_2$Te$_3$, and one GeBi$_2$Te$_4$. Let us first focus on the ARPES spectra without the pump laser for these four samples. It can be seen from the left-hand panels of Figs. 1(c)–1(f) that the two BTS221 samples have their chemical potential within the bulk band gap, and thus only the Dirac surface states are observed to cross the Fermi level, which is consistent with the bulk insulating behavior found in a number of transport experiments [23,24,26]. On the other hand, the chemical potential of the (Bi$_{0.2}$Sb$_{0.8}$)$_2$Te$_3$ sample cuts the bulk valence band, which shows its bulk p-type metallic character, consistent with previous findings [3,7]. The GeBi$_2$Te$_4$ sample has chemical potential within the bulk conduction band. Next, we consider the transient ARPES spectra at two delay time values (DT = 1 ps and DT = 4 μs–3 ps). For the delay time of DT = 1 ps, which corresponds to the point immediately after the pump laser hits the sample, it can be seen that for all samples electrons are excited from below to above the chemical potential, and we can observe electronic states above the chemical potential using ARPES [see the middle panels of Figs. 1(c)–1(f)]. Next, we study the transient ARPES spectra at DT = 4 μs–3 ps.
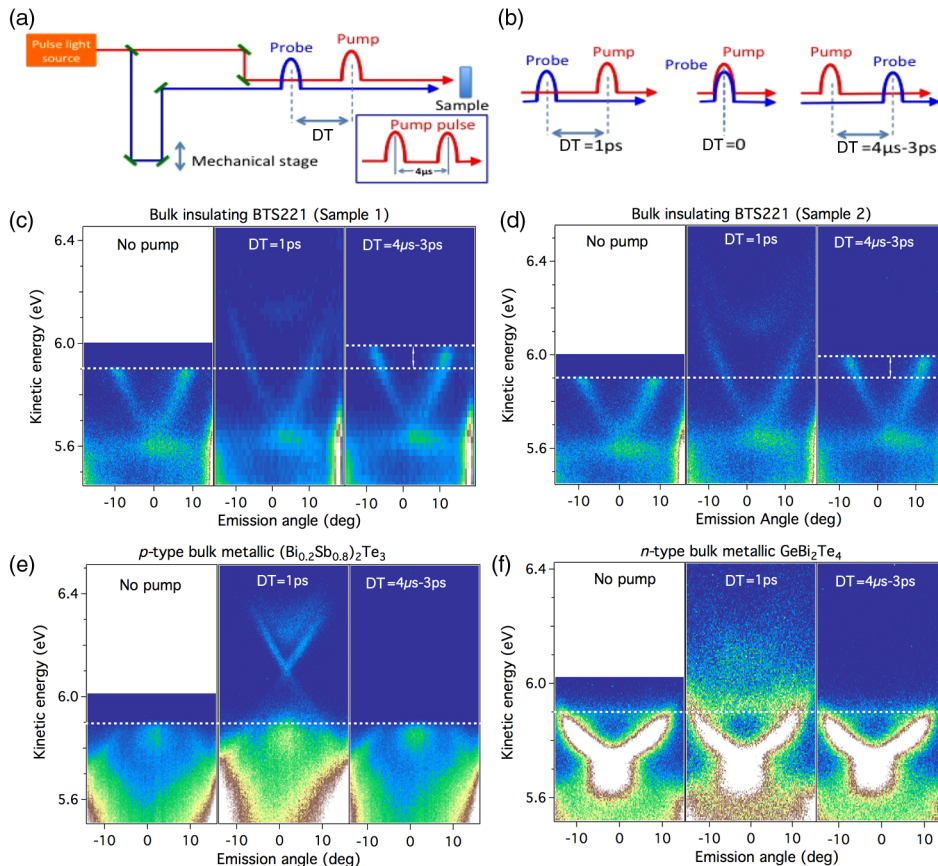

FIG. 1 (color online). Generation of photovoltage in a bulk insulating TI. (a),(b) Schematic view of the pump-probe ARPES experimental setup. The delay time is defined in (b). The inset in (a) shows the frequency of the pump pulse. (c) ARPES band dispersion of bulk insulating Bi$_2$Te$_2$Se (sample 1) near $\bar{\Gamma}$ point measured with no pump (left-hand panel), with pump (middle and right-hand panel). The delay time is noted on the spectra. (d) Similar measurement in (c) for Bi$_2$Te$_2$Se (sample 2). From (c) and (d) the generation of photovoltage (~100 mV) is evident in the bulk insulating sample. Similar measurement as in (d) for (e) p-type bulk metallic (Bi$_{0.2}$Sb$_{0.8}$)$_2$Te$_3$ TI and (f) n-type bulk metallic GeBi$_2$Te$_4$ TI. No generation of the photovoltage is obtained for the bulk metallic topological insulators [see (e) and (f)]. The native Fermi level (Fermi level with the absence of pump pulse) is marked by the white dashed line in the spectra.

As shown in the right-hand panels of Figs. 1(c)–1(f), for the two metallic samples [the right-hand panels of Figs. 1(e) and 1(f)], the transient ARPES spectra at DT = 4 μs–3 ps is nearly identical with the spectra without the pump within our experimental resolution. This is reasonable because we are measuring the sample nearly 4 μs after the pump excitation, so we expect that the sample has already returned to the ground state. However, strikingly, for the two bulk insulating BTS samples [the right-hand panels of Figs. 1(c) and 1(d)], at DT = 4 μs–3 ps the transient ARPES spectra are clearly different from the spectra without pump [the left-hand panels of Figs. 1(c) and 1(d) and Ref. [28]]. More specifically, we found that the chemical potential of the samples is higher by $\simeq$100 mV. We refer to this shift of the chemical potential due to the pump pulse as a surface photovoltage. We observe this effect in multiple pieces of BTS221, suggesting that it is not due to systematic errors or artifacts.

In order to systematically study how the excited electronic states relax, we present the transient ARPES spectra as a function of DT in Fig. 2. For the two metallic samples $(Bi_{0.2}Sb_{0.8})_2Te_3$ and $GeBi_2Te_4$, as seen in Figs. 2(b) and 2(c), all excitations relax within 5 ps. On the other hand, for the bulk insulating BTS221 sample, the relaxation process is longer and more complex. (1) Immediately after the pump ($0 \leq$ DT ≤ 2 ps), the electrons are excited to unoccupied states that extend to energies up to 0.8 eV above the Fermi level. At these short time delays we observe not only the Dirac surface states but also a parabolic bulk conduction band. At these short DT values immediately after the pump, the states are quite spread out along the energy axis and it is hard to define a chemical potential [18]. (2) For DT ≥ 2 ps, the excited intensities far from the Fermi level exhibit a very fast decay. Furthermore, a quasistatic chemical potential appears after 2 ps. And again, strikingly, the quasistatic chemical potential is clearly different from the ground state chemical potential without the pump, so we observe a surface photovoltage. (3) As the delay time increases, the quasistatic chemical potential keeps shifting down. Thus, the surface state photovoltage value decreases. (4) At

DT = 98 ps, the bulk conduction band disappears from the spectrum, and a surface photovoltage of ~100 mV is found. (5) From DT = 98 ps to DT = 4 μs–3 ps, the surface chemical potential only further shifts down by a small energy value (~18 mV) [28]. Thus, the spectrum changes much more quickly for the period $0 \leq$ DT ≤ 98 ps as compared to the period of 98 ps $\leq$ DT ≤ 4 μs–3 ps. (6) A minor but interesting observation is that the energy position of the bottom of the bulk conduction band is found to move down as a function of DT. This is consistent with a surface band-bending effect [35] as a number of surface state electrons are piled up, which is evident from the observed surface photovoltage. (7) Moreover, we also find the light-induced change in the surface-state filling, thanks to the detection in the angle-resolved manner with high energy resolution. What we find is not only the long duration of the surface photovoltage, but the direct observation of the change in the filling of the surface states that persists for times greater than microsecond time scale. Although the effect of band bending after photoexcitation has been previously discussed in topological insulator materials using the TRARPES technique [16,22], the observation of such high surface photovoltage has not been reported in a highly bulk insulating topological insulator material.

In order to quantitatively analyze the rate of change of the transient spectra and obtain the lifetime of the excited states, we show the ARPES intensity of the excited states as a function of delay time (see Fig. 3). To distinguish the lifetime between the surface states and the bulk bands, we do this in a momentum-resolved manner. The white boxes in Fig. 3 show the momenta chosen to represent the surface states and the bulk bands, respectively. For the two metallic samples $(Bi_{0.2}Sb_{0.8})_2Te_3$ and $GeBi_2Te_4$, as seen in Figs. 3(c)–3(f), the lifetime for both the surface states and the bulk bands is found to be below 5 ps, which is consistent with the lifetime reported in previous TRARPES experiments on similar metallic TI samples [16–18,20,21]. The fact that the lifetime for the surface and the bulk is very similar is also intuitive because there are both surface and
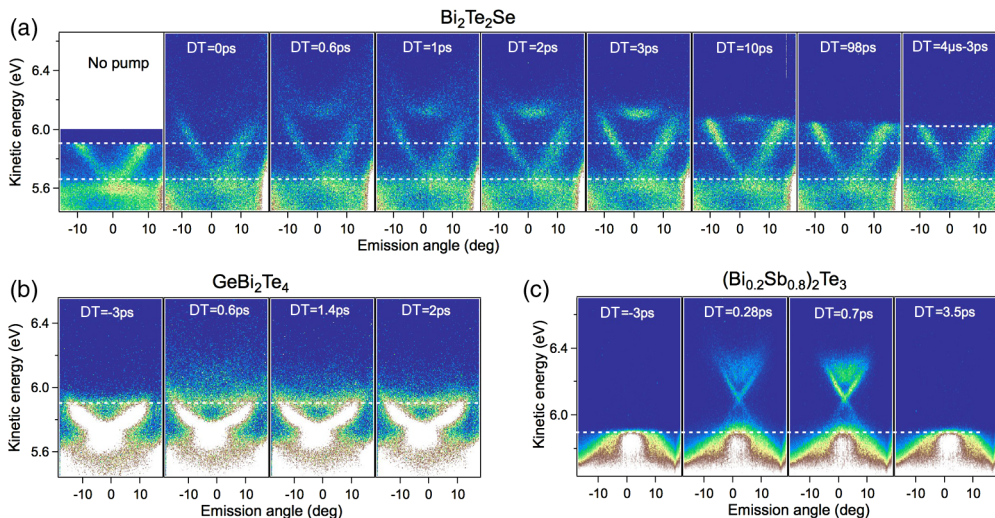

FIG. 2 (color online). Time-resolved ARPES spectra. (a) TRARPES spectra near the $\bar{\Gamma}$ point for various pump-probe delays for BTS221. The populated surface states and bulk conduction band are observed. White dashed lines represent the native chemical potential and the kinetic energy position of the Dirac point. Similar measurement as in (a) for (b) $GeBi_2Te_4$ and (c) $(Bi_{0.2}Sb_{0.8})_2Te_3$. The white dashed line in (b) and (c) represents the native Fermi level.

bulk bands at the Fermi level. As a result, their relaxation processes are coupled. On the other hand, for the bulk insulating BTS221 sample, first of all, the decay is much slower than that of the metallic samples [see Figs. 3(a) and 3(b)]. Second, we note that a relatively fast decay (fast with respect to 4 μs, but still much slower than the ~5 ps lifetime in metallic samples) is found in the first 100 ps ($0 \leq DT \leq 98$ ps) for the surface states. After 100 ps, the surface photovoltage of ~100 mV decays slowly. In particular, the surface photovoltage decreases by only ~18 mV from DT = 98 ps to DT = 4 μs–3 ps [28]. Thus, the total lifetime for the transient electrons exceeds 4 μs.

Now we turn to a discussion of the possible nature of the observed phenomena. First, we note that the existence of a surface photovoltage in our BTS221 samples necessarily

(a) Bi$_2$Te$_2$Se DT = 1ps

(b) Bi$_2$Te$_2$Se
● Bulk State
● Surface State

(c) GeBi$_2$Te$_4$ DT = 1ps

(d) GeBi$_2$Te$_4$
● Bulk State
● Surface State

(e) (Bi$_{0.2}$Sb$_{0.8}$)$_2$Te$_3$ DT = 1ps

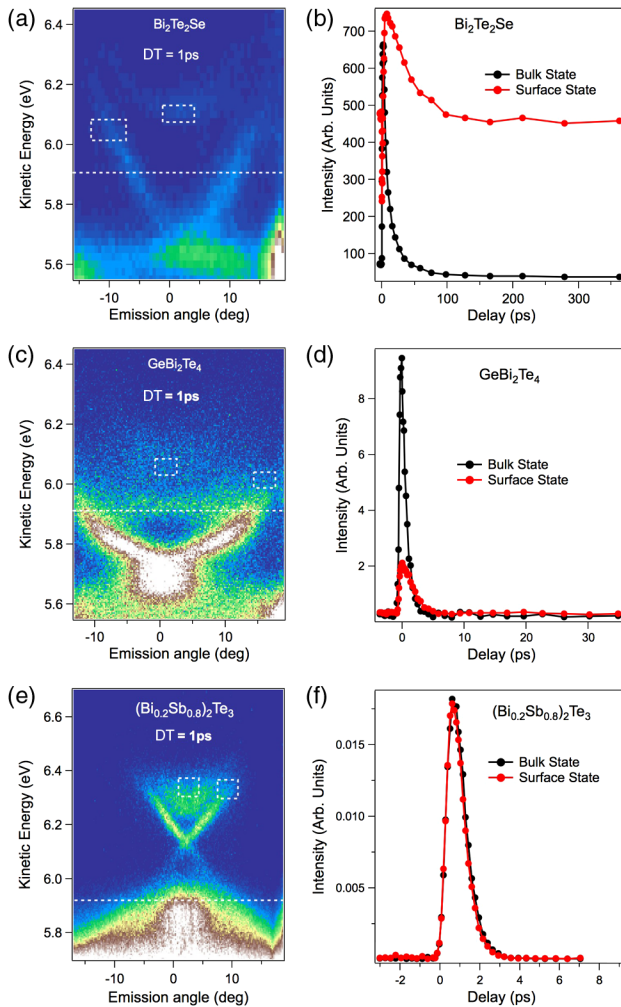(f) (Bi$_{0.2}$Sb$_{0.8}$)$_2$Te$_3$
● Bulk State
● Surface State

FIG. 3 (color online). Lifetime comparison between bulk insulating and metallic TIs. (a) ARPES band dispersion of BTS221 with positive time delay. (b) Ultrafast evolution of the population of surface states (red curve) and bulk states (black curve) for BTS221. Similar measurement as in (a) and (b) for GeBi$_2$Te$_4$ shown in (c) and (d) and (Bi$_{0.2}$Sb$_{0.8}$)$_2$Te$_3$ shown in (e) and (f). The white rectangles represent the integration window of transient photoemission intensity for surface and bulk states in (a), (c), and (e).

implies a charge redistribution upon illumination [36]. The most likely scenario for this to occur is a charge drift caused by the electric field of the photovoltage. We point out some reasons that the excited electrons occupying Dirac surface states in bulk insulating samples have exceedingly long lifetimes. Upon application of the pump pulse, photoexited electrons and holes are created. Their energy gain can be up to the photon energy of 1.47 eV. No electrons escape to the vacuum level because the pump pulse energy is lower than the BTS221 work function ≈4–5 eV. One straightforward relaxation process for the electron hole pairs is the direct recombination across the bulk energy gap. Since our pump pulse is very intense, it generates many electron-hole pairs per pulse (the pump power is 20 mW, which means ~10$^{17} \times A$ electron-hole pairs per second, where A is the optical absorption coefficient of BTS221), and the direct interband recombination process is likely to be inefficient. These allow some of the excited electron on BTS221 to drift towards the surface, giving rise to a surface photovoltage. This process is almost complete at about DT = 10 ps, as can be seen Fig. 2(a).

For DT > 10 ps, a quite stable shift in the surface chemical potential is observed and a slow decay in the ARPES intensity of the excess charge states sets in. This can be understood at a qualitative level by a drift of charge moving out of the beam spot via a two-dimensional relaxation process [37], where excess charge relaxes as $\rho(r,t) = (1/2\pi)v/(r^2 + v^2 t^2)^{3/2}$, where v is the drift velocity. This power-law decay in two dimensions is in sharp contrast to fast exponential decay in three-dimensional metals. We estimate the time it takes for the charge to drift out of the pump laser spot (with the length scale $D = 1$ mm) under the observed photovoltage. The drift velocity of the excited electrons is given by $v = \mu E$, where μ is the mobility of the Dirac surface states ($\mu \approx 3000$ cm^2/V s, see Ref. [24]) and E is the electric field from the location of the pump beam spot to other locations on the sample's surface not affected by pumping. Since the quasistatic surface photovoltage is about 0.1 eV, we set the voltage drop V to be 0.1 V. So the electric field $E = V/D = 100$ V/m, and the drift velocity is $v = \mu E = 30$ m/s. And finally, the drift time scale is estimated to be $t = D/v \sim 30$ μs. To understand the absence of surface photovoltage in the bulk conducting samples GeBi$_2$Te$_4$ and (Bi$_{0.2}$Sb$_{0.8}$)$_2$Te$_3$, we consider the 3D relaxation, which is exponential $\rho(t) = \rho(0)e^{-t/\tau_{3D}}$, where $\tau_{3D} = \epsilon/\sigma$. An estimate of τ_{3D} for our GeBi$_2$Te$_4$ and (Bi$_{0.2}$Sb$_{0.8}$)$_2$Te$_3$ samples (at the same density) gives τ_{3D} on the order of a picosecond, which is indeed much shorter than for BTS221, in agreement with our observations (see also Ref. [28], which includes Ref. [30–32]).

In conclusion, we observe a gigantic lifetime for excited surface states in a bulk insulating topological insulator, providing compelling evidence for a power-law charge relaxation, which is unique to two-dimensional electrodynamics, and, at the same time, offering a direct optical signature of low bulk conductivity in a topological

insulator. Our observation potentially suggests a way to develop optical devices, such as an optical *p-n* junction (see Ref. [28]) and a solar cell [38], based on the optically excited Dirac surface states. Our results also provide a paradigm to compare with possible surface photovoltage in other bulk insulating topological insulators [39–42] as well as to engineer them for future optical devices.

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