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Citation: Appl. Phys. Lett. 110, 212103 (2017); doi: 10.1063/1.4984141
View online: http://dx.doi.org/10.1063/1.4984141
View Table of Contents: http://aip.scitation.org/toc/apl/110/21
Published by the American Institute of Physics
Linear-optical access to topological insulator surface states

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(Received 9 February 2017; accepted 13 May 2017; published online 25 May 2017)

We demonstrate efficient linear-optical access to surface-state spin dynamics in Bi2Se3 by probing transitions between two surface-state Dirac cones, providing a practical technique for spin-current dynamics studies in topological-insulator devices. Using broadband transient-reflectivity pump-probe measurements, we distinguish bulk and surface state-responses, by controlling photon energy and circular polarization at oblique incidence. For pump-photon energies corresponding to bulk-state transitions, the probe polarized co-circularly with the pump shows stronger reflectivity change, compared to the anti-circularly polarized probe. However, pump photon energies corresponding to surface-state transitions result in an opposite effect, with the anti-circularly polarized probe exhibiting stronger reflectivity change. This surprising behavior stems from the surface-state in-plane spin orientation near the Dirac point, and the surface-state spin population remains at the injected energy for several ps. These results enable an efficient approach for studying spin current dynamics in topological-insulator based technologies. Published by AIP Publishing.

Topological insulators (TIs) are becoming a topic of great interest in both fundamental physics and in technological applications, due to their remarkable properties, with an insulating bulk and topologically protected metallic surface states. These surface states exhibit massless Dirac dispersion and locking of the spin orientation to the momentum vector, resulting in dissipationless spin currents, enabling spintronic devices. Unconventional phases of matter and quasiparticles have been predicted based on combinations of topological insulators with superconductors inspiring various experimental directions, including TI combinations with unconventional superconductors, paving the way for practical realizations of topological quantum computing. Electronic characterization has been able to determine much about the spin transport, however, it does not allow noninvasive probing of the surface states. Moreover, the investigation of the ultrafast spin dynamics is virtually impossible by electrical transport techniques. All-optical access on the other hand, provides a very powerful method for noninvasive investigation of ultrafast processes in TIs while separating the surface response from that of the bulk is still extremely challenging. Recently, a nonlinear-optical approach was employed to study the TI surface. However, such nonlinear-optical techniques are limited significantly by the inherently weak high-order effects.

Here, we demonstrate efficient linear-optical access to TI Bi2Se3 ultrafast spin dynamics by broadband time-resolved transient reflectivity measurements. This enables a practical technique to access spin-current behavior in TI based devices. We exploit the interplay between co- and anti-circular polarizations of the pump and the probe photons at oblique incidence—to distinguish between bulk and surface state responses in optically excited transitions between two Dirac cones in Bi2Se3 separated by approximately 1.5 eV [Fig. 1(b)]. Optical transitions between these two cones have been recently employed for coherent control based techniques of current injection in TIs and for the investigation of charge-carrier dynamics. We take advantage of the two-cone transitions under circularly polarized excitation in order to investigate spin dynamics and to distinguish the response of the surface states from those of the bulk. We study two characteristic cases of pump photon energies—1.72 eV and 2 eV, accounting for surface-to-surface and bulk-to-bulk state transitions between the two cones, probed by a white-light supercontinuum.

In the bulk transition case, the strongest change in differential reflectivity is for the co-circularly polarized pump and probe, resulting mainly from components of the photon and electron spins oriented at small angles to the normal.
whereas for surface states, the strongest change is, surprisingly, for opposite pump and probe circular polarization. We explain this behavior by the fact that in surface states, the electrons interact mainly with the parallel-to-surface components of the photon spin. We find that the effect of the polarized pump on the surface states is stronger than in the bulk case. Furthermore, carriers injected into bulk states result in significant change in differential reflectivity at a different energy from that of the pump—close to the bulk band edge. Carriers injected into surface states cause the strongest change in differential reflectivity at the injected energy, maintained for several ps. This is attributed to reduced scattering in surface states maintaining the injected carrier energy and the corresponding spin current.

We studied experimentally different spin-polarized transitions in off-stoichiometric Bi$_2$Se$_3$ by controlling photon energy and circular polarization. In principle, there are three possible transitions: bulk-bulk, surface-bulk, and surface-surface [Figs. 1(a) and 1(b)]. Our approach provides a technique to distinguish between surface and bulk transitions. For our experiments, we used a bulk single crystal of off-stoichiometric Bi$_2$Se$_3$ grown in the modified Bridgman method. Based on angle-resolved photoemission spectroscopy (ARPES) and electrical transport measurements, we estimate its carrier concentration to be $\sim 10^{18} \text{ cm}^{-3}$, corresponding to the relatively low Fermi level position. The Fermi level position is specified relative to the bottom edge of the conduction band such that a low Fermi level corresponds to the low bulk conduction band carrier concentration. Unlike mid-infrared absorption within a Dirac cone, the near-infrared photons in our experiments can only probe transitions between the two Dirac cones with some contribution of bulk absorption. However, due to the low Fermi level in our off-stoichiometric sample, these transitions between the first and second Dirac cones (cone-to-cone) are less obscured by contributions of bulk-to-surface and bulk-to-bulk transitions.

The transient polarized differential reflectivity $\Delta R/R$ pump-probe measurement is used here to observe spin dynamics of the surface states.

The equilibrium population of the sample band structure was mapped using ARPES at a temperature of 23 K with a photon energy of 20 eV. The ARPES measurement on our sample shows very low electron population in the conduction band—0.2 eV above the Dirac point [Fig. 2(a)]. This significantly reduces the contribution of several transitions involving bulk states, compared to those involving surface states—in addition to the angular momentum projection selection.

In our setup [Fig. 2(b)], a 1 kHz Ti:sapphire amplifier delivering 35 fs pulses at 800 nm served to generate a white-light supercontinuum using a sapphire plate. The generated white light was used as a probe to measure the differential reflectivity of the Bi$_2$Se$_3$ sample over a broad energy range using an optical spectrum analyzer. An optical parametric amplifier (OPA) provided tunable-wavelength $\sim 120$ fs pulses, which served as a pump. A quarter wave plate (QWP) on the white-light probe path allows co-circular and anti-circular polarization settings between the pump and the probe, and the delay line sets the relative time delay $\Delta t$. The spot size of the pump is $\sim 100 \mu$m in diameter with the average pump power of $\sim 3 \text{ mW}$. The probe white light spot diameter is $\sim 50 \mu$m (smaller than that of the pump), with the probe average power of $\sim 100 \mu\text{W}$. The reflectivity of the sample is $\sim 50\%$, and about 50% of the incident pump power is absorbed in the sample.

The measurements were performed at room temperature at two characteristic pump-photon energies of 1.72 eV and 2 eV, accounting for surface-to-surface and surface-to-bulk transitions between lower and upper Dirac cones, respectively, as shown in Fig. 1(b).

Since the pump and the probe beams are incident at small angles, in the case of pump-photon energy corresponding to surface-to-bulk transition, the spin orientation excited by the pump pulse is at an angle close to the normal. A probe pulse, co-circularly polarized with the pump, addresses the excited spin polarization along a projection onto the axis.
defined by the angular momentum orientation of the pump photons. This results in large projection of the spin and a resulting strong effect in differential reflectivity [Fig. 3(b)]. In contrast, a probe pulse, anti-circularly polarized with the pump, addresses the spin polarization with negligible projection onto the spin excited by the pump—resulting in a vanishing effect on differential reflectivity [Fig. 3(a)].

However, in the case of pump-photon energy corresponding to the surface-to-surface transition between the two Dirac cones, the response is, in fact, opposite (Fig. 4). Here, for the co-circularly polarized pump and probe [Fig. 4(b)], the change in differential reflectivity is much less pronounced with comparison to the anti-circular polarization case [Fig. 4(a)]. This behavior is caused by the fact that near the Dirac point, surface-state spins are oriented in-plane of the surface. Therefore, only the in-plane component of the photon spin is coupled to the surface-state electron spin ($s_x$).

In this case, when the pump and probe are anti-circularly polarized, the spin population induced by the pump on the surface is probed [Fig. 4(a) inset]. As a consequence, significant changes in the differential reflectivity are observed. On the other hand, for the co-circularly polarized pump and probe, the surface spin population induced by the pump is opposite to the probe polarization, and no significant reflectivity changes are observed [Fig. 4(b)]. Therefore, by changing pump-photon energy and polarization, we can distinguish between transitions from the surface-to-bulk and surface-to-surface of the lower Dirac cone to the upper Dirac cone. Another evidence for the surface-bulk distinction in our approach is manifested in the fact that in the case of the surface-to-bulk transition corresponding to 2 eV pump photon energy, the strongest response is observed around $\approx 1.82$ eV, which can be explained by the relaxation of injected electrons in the bulk to the minimum of the upper conduction band.

However, in the case of the surface-to-surface transition, this relaxation does not occur on such short timescales in the surface states, and the strongest response is observed near the energy, at which the carriers were injected by the pump.

In the case of surface-to-bulk transition, the material response was found to be opposite to the surface-to-surface case and less pronounced. This is explained by the fact that bulk bands are spin-degenerate with no spin-momentum...
FIG. 5. Time dependence of the normalized differential reflectivity $\Delta R/R$ minimum for bulk (solid red line) and surface (solid blue line) responses. The dashed lines represent exponential fits with the corresponding time constants $\tau_b = 1.6$ ps for the bulk and $\tau_s = 2$ ps for the surface. The insets show schematic drawings of pump and probe photon angular momenta and the corresponding spin orientations.

correlation. This clear distinction between two opposite behaviors of the bulk and of the surface states in TIs enables us to specifically address surface-state spin dynamics. The time constants for the signal decay (Fig. 5) are $\tau_b = 1.6$ ps for the bulk and $\tau_s = 2$ ps for the surface. These values are comparable to the decay times reported in $\text{Bi}_2\text{Se}_3$ previously but slightly shorter. We attribute this shortening of decay times to the significantly higher carrier densities in our experiments due to the higher pump intensities ($>100\text{GW/cm}^2$) compared to the previous reports. Under such high excitations, carrier-carrier scattering can affect the decay, which is dominated by phonon scattering at lower densities. This contribution of carrier-carrier scattering at high carrier density also results in slightly shorter decay time in the bulk compared to that of the surface states.

In conclusion, we have shown linear-optical access to topological insulator ultrafast spin dynamics using broadband transient-differential reflectivity measurements. This method enables direct access to spin current dynamics in TI based spintronic devices, providing important information about the spin-current decay times in practical TIs. We have separated bulk and surface responses by changing excitation energy and incident light circular polarization in the specially prepared off-stoichiometric $\text{Bi}_2\text{Se}_3$ sample and have selectively observed spin-polarized cone-to-cone transitions. For the surface-to-bulk transition, a spin response is pronounced when pump and probe polarizations are co-circular, whereas for the surface-to-surface transition, the response is significant for anti-circular polarization. Moreover, in contrast to the faster-decaying bulk-state population, the surface-state spin population and the corresponding spin current remain near the injected energy for several ps. Our results enable a practical approach for the study of topological materials and provide an efficient method for controlling and probing spin-polarized dynamics in topological insulators for a wide application range including spintronics and quantum computation.

This research was supported by the Israel science foundation (Grant No. 2220/15).


