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We report on time-resolved ultrafast optical spectroscopy study of the topological insulator (TI) Bi$_2$Se$_3$. We unravel that a net spin polarization can not only be generated using circularly polarized light via interband transitions between topological surface states (SSs), but also via transitions between SSs and bulk states. Our experiment demonstrates that tuning photon energy or temperature can essentially allow for photoexcitation of spin-polarized electrons to unoccupied topological SSs with two distinct spin relaxation times (∼25 fs and ∼300 fs), depending on the coupling between SSs and bulk states. The intrinsic mechanism leading to such distinctive spin dynamics is the scattering in SSs and bulk states which is dominated by $E^*_g$ and $A_{1u}^g$ phonon modes, respectively. These findings are suggestive of novel ways to manipulate the photoinduced coherent spins in TIs.

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Topological insulators (TIs), as a new quantum phase of matter, are characterized by an unusual electronic structure exhibiting both insulating bulk and robust metallic surface states (SSs) [1, 2]. This unique electronic structure combining external light excitation on TIs leads to many exotic physical phenomena [3–12], which hold TIs a great promise for opto-spintronics and ultrafast spintronics applications [13]. Therefore, it becomes crucial to study the out-of-equilibrium properties of TIs under photo-excitation. Among them, the charge and spin dynamics have attracted a lot of recent attention, and are explored effectively using the time- and angle-resolved photoemission spectroscopy (Tr-ARPES) [14–21] and time-resolved optical spectroscopy [22–32]. Investigation of the non-equilibrium charge dynamics enables deep understanding of the momentum scattering, which is a fundamental process determining the electronic transport in TIs [33]. On the other hand, comprehensive knowledge of the spin dynamics, including coherent spin generation and relaxation, is vital for actively manipulating spins in spintronics [34]. Most of previous time-resolved works focus on the charge dynamics in TIs, and have revealed that the electron-phonon (e-p) coupling plays a key role in momentum scattering of the non-equilibrium carriers. In contrast, very few works pay attention on the photoinduced coherent spin dynamics in TIs [24, 35, 36]. Specifically, several key questions are still open regarding the dynamical response of the spin properties to the incident light: (1) Can a surface net spin polarization be generated via interband transitions from bulk states to SSs using circularly polarized light? (2) Does the spin dynamics in topological SSs and bulk states behave in a similar manner? Or does the same mechanism dictate the spin dynamics in topological SSs and bulk states?

In this Letter, we employ time-resolved transient reflectivity and Kerr rotation measurements to investigate the photo-excited charge and coherent spin dynamics in the prototypical TI Bi$_2$Se$_3$. Two types of helicity-dependent photoinduced net spin polarization have been unveiled, and can be attributed to the interband transitions between SSs and bulk states. We show that two coherent spin dynamics with distinct spin relaxation times can be selectively excited by tuning the photon energy $h\nu$ or temperature $T$. We reveal that the e-p scattering in SSs and bulk states dominated by different phonon modes gives rise to the distinct spin relaxation.

Time-resolved transient reflectivity change $\Delta R(t)/R$ and time-resolved Kerr rotation (TRKR) $\Delta \Psi_K(t)$ were measured based on a pump-probe scheme using a Ti:sapphire laser with a time resolution of ∼35 fs. Experiments were performed on high quality Bi$_2$Se$_3$ samples with carrier density $n \simeq 4 \times 10^{18}$ cm$^{-3}$. The Fermi level $E_F$ resides inside the bulk band-gap, and is ∼0.1 eV above the Dirac point. Detailed description of the materials and the measurement setup can be found in Supplemental Material [37].

Figure 1(a) shows a typical transient reflectivity $\Delta R/R$ signal, where an initial fast-decaying component is followed by slow relaxation processes superimposed with an oscillatory behavior. An exponential fit to the initial fast decay (blue line) exhibits a time constant of $\tau_{ep} \simeq 300$ fs, consistent with the cooling time of electrons in the bulk bands for electronic temperature $T_e \gtrsim 600$ K [14, 15]. Since in such a state the optical phonon cooling
is expected to be the most effective channel, the fast-decaying component can be attributed to an electron-optical-phonon scattering process. The corresponding scattering rate $\tau_{ep}^{-1}$ as a function of $T$ ($T \gtrsim 10$ K) is shown in Fig. 1(b), where the linear-in-$T$ behavior is expected for a given Debye temperature of $\Theta_D \sim 180$ K [28]. The following slow relaxation has a time constant of $\sim 1$ ps, in well agreement with previous findings of the electron-phonon scattering time associated with the low energy phonons [14–17, 22–24, 32]. We can extract the oscillator component in $\Delta R/R$, whose Fourier transform (FT) reveals several frequencies [as shown in Fig. 1(c)], e.g., $\sim 2.2$ THz, 4 THz, and 5.2 THz at 293 K. These terahertz oscillations are due to coherent optical phonons, initiated via either coherent Raman scattering [38] or dispersive excitation [39]. The three peaks in Fig. 1(c) from left to right are attributed to $A_{1g}^1$, $E_g^2$, and $A_{2g}^2$ Raman-active optical phonon modes, respectively [40]. Another $E_g^1$ Raman-active mode can also be observed at low temperatures (see Supplemental Material [37]). Among all the optical phonon modes, $A_{1g}^1$ is the strongest, suggesting that electron-$A_{1g}^1$-optical-phonon coupling dominates the e-p scattering or the momentum scattering time ($\tau_p$) in the excited bulk states within the initial $\sim 500$ fs, as also indicated by the Tr-ARPES measurements [20].

In addition, one can unambiguously extract component $(\Delta R/R)_{E_g}$ from $\Delta R/R$ [Fig. 1(d)], which directly shows the electron-phonon scattering process coupled solely with the $E_g$ phonon modes ($E_g^2$ dominates, see Supplemental Material [37]). Fitting $(\Delta R/R)_{E_g}$ with an exponential decay, we obtain an electron-phonon scattering time of $\tau_{ep} \approx 30$ fs, which is nearly temperature independent. Clearly, $\tau_{ep}$ is about 10 times faster than $\tau_p$ associated with the dominant $A_{1g}^1$ phonon mode. Our experiment thus evidently demonstrates that scattering events coupled with different phonon modes will decay in distinct timescales.

Figure 2(a) shows typical measurements of $\Delta \theta_K$ at low and high temperatures. It can be seen that the circularly polarized light generates a non-equilibrium net spin polarization in Bi$_2$Se$_3$, as the Kerr rotation signals change sign with the helicity of the pump light. This observation is consistent with that reported in Ref. [24], where the spin relaxation time is $\sim 200$ fs (limited by time-resolution) for excited electrons in either SSs or bulk states of Bi$_2$Se$_3$ at room temperature. In this work, our TRKR signals evidently reveal novel complex structures.

Microscopically, generation of a net spin density in opaque materials by circularly polarized light involves direct optical transitions between different energy levels and the accompanying angular momentum transfer (spin selection rules), as in GaAs [41]. Therefore, in order to understand the nature of the related electronic states, we have carried out detailed measurements on the $h\nu$-dependent and $T$-dependent $\Delta \theta_K$. The results are shown in Figs. 2(a)-(c). Here, we first notice that the $h\nu$-dependence of $\Delta \theta_K$ resembles the $T$-dependence. This similarity can be understood by the band-gap shrinkage (BGS) with increasing $T$ via electron-phonon interactions commonly seen in most semiconductors [42]. Due to the BGS effect, the influence on optical transitions by increasing $T$ is analogous to that by increasing $h\nu$.

From Figs. 2(a)-(c), we also find that three dynamical processes contribute to $\Delta \theta_K(t)$. An ultrafast transient with positive sign (indicated by green arrows) dominates at large $h\nu$ or high $T$, but almost disappears at small $h\nu$ or low $T$. It lasts for about 100 fs. A relatively slow decay component with negative sign (indicated by blue arrows) is observed at all investigated $h\nu$ and $T$. It is persistent to $\sim 1.2$ ps. The third process with positive sign (indicated by magenta arrows) appears at small $h\nu$ and low $T$. It also lasts for about 1.2 ps. Quantitatively, these three processes can be fitted using three exponential decays with time constants $\tau_s$, $\tau_b$, and $\tau_{sh}$, respectively. The fitted results agree well with the experimental data [Fig. 2(c)]. Similar data have also been obtained in highly n-doped samples (see Supplemental Material [37]).

According to the previous Tr-ARPES studies [18, 19], the instantaneous populated states from the optical transitions with $h\nu \sim 1.61$ eV should involve the second topological SS (SS$_2$) above the bulk conduction band (BCB$_1$), as shown in Fig. 3. In this situation, the allowable direct optical transitions are from the first topological SS (SS$_1$) to SS$_2$, from the first bulk valence band (BVB$_1$) to the second high-lying bulk valence band (BVB$_2$), and from SS$_1$ to BVB$_2$ [Fig. 3(a)]. These transitions are symbolically...
ized by SS$\rightarrow$SS$_2$, BVB$_1$→BVB$_2$, and SS$_1$→BVB$_2$, respectively. However, when $h\nu$ or $T$ is relatively small, $h\nu$ becomes insufficient for populating SS$_2$ via SS$_1$→SS$_2$, although BVB$_1$→BVB$_2$ and SS$_1$→BVB$_2$ can still occur. Apparently, if the process characterized by $\tau_s$ is attributed to the relaxation of the spin-polarized electrons in SS$_2$ via SS$_1$→SS$_2$, the corresponding TRKR signal should vanish with decreasing $h\nu$ or $T$, in well consistent with our experimental observations [Fig. 2]. This scenario is also consistent with the theoretical works of Refs. [9, 10], that is, the spin-polarized electrons can be generated by SS-to-SS interband transitions with particular spin selection rules using circularly polarized light.

The spin dynamics characterized by $\tau_0$ occurs at all $h\nu$ and $T$ investigated, where both transitions BVB$_1$→BVB$_2$ and SS$_1$→BVB$_2$ are always allowed [Fig. 3]. However, the electrons excited by $\sigma^\pm$ photons via interband transitions between bulk states should be essentially unpolarized, due to the bulk inversion symmetry [35]. Even if for some other reasons, a net spin polarization is generated from the transition between bulk states BVB$_1$→BVB$_2$, it is expected to be $h\nu$- and $T$-independent, as the total number of the populated bulk states remains nearly intact in the energy range investigated [Figs. 3(a) and (b)]. But, experimentally, we find that the amplitude of $\tau_0$ process exhibits a strong $h\nu$ and $T$-dependence: it decreases with increasing $h\nu$ and $T$, as shown in Figs. 2(b) and (c). Such dependence is consistent with the population change associated with SS$_1$→BVB$_2$. Therefore, one can naturally attribute $\tau_0$ to the spin relaxation of excited electrons in BVB$_2$, as illustrated in Figs. 3(c) and (d). It is worth noting that, due to angular momentum conservation, the spin-polarized electrons in bulk states excited from SS$_1$ should have an opposite polarization direction to that in SS$_2$ excited from bulk states. This phenomenon is indeed observed in highly $n$-doped samples, where additional transition BCB$_1$→SS$_2$ induces a surface net spin polarization in SS$_2$ opposite to the spin state in BVB$_2$ via SS$_1$→BVB$_2$ (BVB$_2$ process) (see Supplemental Material [37]).

The spin dynamics characterized by $\tau_{sb}$ only appears at small $h\nu$ and $T$. It has the same sign as the dynamics characterized by $\tau_s$ (SS$_1$→SS$_2$), whereas with a similar relaxation timescale as the dynamics characterized by $\tau_b$ (SS$_1$→BVB$_2$). Such observations suggest that this dynamics might be associated with both excited SS$_2$s and bulk states, i.e., SS$_3$ (same sign) and BVB$_2$ (similar timescale). In fact, as $h\nu$ or $T$ decreases, the non-equilibrium spin-polarized electrons excited from SS$_1$ to SS$_2$ will be at the edge of BVB$_2$ [Fig. 3(d)], where strong coupling between SS$_2$ and BVB$_2$ exists [43]. The energetic spin-polarized electrons can then efficiently transfer from SS$_2$ to available bulk states in BVB$_2$, while maintaining the polarization direction as in the dynamics characterized by $\tau_s$. This interpretation also leads to $\tau_{sb} \approx \tau_b$, as they both describe the spin relaxation of the spin-polarized electrons in BVB$_2$. It naturally explains the unusual slow rise time ($\sim 150$ fs) in the $\tau_{sb}$ process, which is needed for the charge transfer.

As an intermediate conclusion, our experiments unveil that generation of net spin polarization in three-dimensional TIs using $\sigma^\pm$ photons requires the participation of the topological SS$_s$, e.g., SS$_1$ and/or SS$_2$ in Bi$_2$Se$_3$. Remarkably, we find that by properly tuning $h\nu$ or $T$, the spin-polarized electrons excited to unoccupied SS$_s$ (e.g., SS$_2$ in Bi$_2$Se$_3$) can have distinct spin relaxation times, depending on whether they are strongly coupled to bulk states or not. Such tunability is essential for the
development of future ultrafast spintronic devices [34]. Therefore, the underlying mechanism behind it requires to be further explored.

In a TI system such as Bi$_2$Se$_3$, the electron spin precession around an effective magnetic field with corresponding frequency $\Omega$ during a correlation time $\tau_c$ (average timescale of spin precession along one direction) may experience two different spin relaxation mechanisms. The first is the D’yakonov Perel’ (DP) mechanism for SSs with spin-momentum locking in absence of the inversion symmetry [34]. In this case, the strong spin-orbit coupling (SOC) in SSs induces the effective magnetic field with the corresponding $\Omega$ given by the energy splitting between $E_{\mathbf{k}_+}$ and $E_{\mathbf{k}_-}$: $\hbar \Omega \sim 2 \hbar v_f k$ [1, 2, 34], where $\mathbf{k}$ is the electron wave vector, $\hbar$ is the reduced Planck’s constant, $v_f$ is the Fermi velocity, and $\uparrow\downarrow$ stand for spins with opposite directions. Taking $\hbar \Omega \sim 2 E_f \simeq 0.2$ eV and $\tau_c \simeq \tau_p \sim 50$ fs from infrared transmission measurements [33], we thus can obtain $\Omega \tau_c \gg 1$. The second spin relaxation mechanism is the Elliot-Yafet (EY) mechanism for bulk states with inversion symmetry [34]. Here, the spin-orbit interaction, altered by phonons, causes the coupling of different spin states and leads to the spin flips via momentum scattering. The correlation time $\tau_c$ is then determined by the momentum scattering time $\tau_p$ and the inverse of the frequency $f^{-1}$ of relevant thermal phonons [34], both of which give $\tau_c \sim 500$ fs, taking $\tau_p \sim \tau_{ep}$ and $f^{-1} \sim f^{-1}(A_{ig}^1)$. In addition, $\hbar \Omega$ can be estimated by the strength of SOC in Bi$_2$Se$_3$, with a typical value of $\hbar \Omega \simeq \lambda_{SO} \sim 0.1$ eV [2, 44]. In fact, the coupling strength between bulk states BCB$_3$ and BVB$_j$ ($j = 1, 2$) can be as large as the energy gap ($\sim 0.3$ eV) [44]. $\Omega \tau_c \gg 1$ then can also be obtained for the bulk states in EY mechanism.

The relation $\Omega \tau_c \gg 1$ implies that the electron spin precesses many full cycles during $\tau_c$ around the effective magnetic field. The spin polarization decays irreversibly after $\tau_c$ [34]. Therefore, the spin relaxation time $\tau_{spin}$ is given by $\tau_{spin} \sim \tau_c$, applied to both excited SSs and bulk states in Bi$_2$Se$_3$, which is consistent with recent theoretical works [45–47].

Experimentally, we reveal two distinct relaxation timescales for the coherent spins excited in SSs and bulk states. The fast dynamics is characterized by $\tau_s \sim 25$ fs, which is nearly $T$-independent and related to the excited spin-polarized electrons in SS$_2$. Since $\tau_s \sim \tau_{ep}$, one can conclude that the electron-$E_g^2$-optical-phonon scattering dominates the scattering process in excited SS, and hence $\tau_s$. This result is in contrast to that in the excited bulk states, where two relatively slow spin relaxation $\tau_b$ and $\tau_{sh}$ associated with BVB$_2$ are found to be $\sim 300$ fs at low $T$, similar to $\tau_{ep}$ ($\tau_b \simeq \tau_{sh} \sim \tau_{ep}$) [Figs. 1(b) and 2(d)]. In addition, a linear-in-$T$ dependence is observed for $\tau_{ep}^{-1}$, which exactly follows the $T$ dependence of $\tau_{ep}^{-1}$. These observations thus suggest that the scattering/spin relaxation in excited bulk states is dominated by the $A_{ig}^1$ phonon mode. Since the $E_g^2$ and $A_{ig}^1$ phonon modes have distinct energies, the spin dynamics in topological SSs and bulk states exhibit different behavior. This finding, along with previous works [20, 48–50], may inspire further investigation on dynamical properties in topological SSs.

![Schematic illustration of the resonant photo-excitation processes in Bi$_2$Se$_3$ based on the Tr-ARPES results in Refs. [14, 19]. (a) and (b) show the electrons resonantly populated by the direct optical transitions SS$_1 \rightarrow$ SS$_2$, SS$_1 \rightarrow$ BVB$_2$, and BVB$_1 \rightarrow$ BVB$_2$ (indicated by red arrows) using linearly polarized light at $\sim 1.61$ eV and $\sim 1.53$ eV, respectively. (c) and (d) show the spin-polarized electrons resonantly excited via SS$_1 \rightarrow$ SS$_2$ (red shading) and SS$_1 \rightarrow$ BVB$_2$ (green shading) using circularly polarized light at $\sim 1.61$ eV and $\sim 1.53$ eV, respectively. Excited electrons generated via SS$_1 \rightarrow$ SS$_2$ in (a) and (c) are above the top of BVB$_2$, and thus weakly coupled to BVB$_2$. Excited electrons generated via SS$_1 \rightarrow$ SS$_2$ in (b) and (d) are at the edge of BVB$_2$, where SS$_2$ and BVB$_2$ are strongly coupled. The spin-polarized electrons generated via SS$_1 \rightarrow$ SS$_2$ have an opposite polarization direction to that via SS$_1 \rightarrow$ BVB$_2$ (indicated by up and down yellow arrows). In (d), the spin-polarized electrons are expected to transfer from SS$_2$ into BVB$_2$, and thus exhibit a spin relaxation $\tau_{sh} \simeq \tau_b$, being distinct from $\tau_s$. In all panels, black dashed lines represent the Fermi level $E_f$. The upper dashed lines are guides to the eye for projection of the initial states to $\sim 1.61$ eV or $\sim 1.53$ eV higher in energy. Because the electrons generated via BVB$_1 \rightarrow$ BVB$_2$ in (a) and (b) are essentially unpolarized, they are not shown in (c) and (d).]
In summary, we performed ultrafast optical spectroscopy study of spin dynamics in TI Bi$_2$Se$_3$. We unravel that non-equilibrium net spin polarization induced by $\sigma^\pm$ photons requires the participation of topological surface states (SSs) in the interband transitions. For the first time, we demonstrate that besides manipulating non-equilibrium electron spins in TI Bi$_2$Se$_3$ by switching circular polarization, one can selectively excite spin-polarized electrons to unoccupied SS$_2$ with two distinct spin relaxation times by only tuning the photon energy or temperature. We reveal that the distinct spin relaxation arise from the scattering in SSs and bulk states which is dominated by $E^g_2$ and $A^1_g$ phonon modes, respectively. Our measurements thus pave a way to manipulate the photoinduced coherent spins in TIs, which may have profound implications in future TI-based ultrafast spintronic devices.

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