Ultrafast evolution of bulk, surface and surface resonance states in photoexcited Bi$_2$Te$_3$

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We use circular dichroism (CD) in time- and angle-resolved photoemission spectroscopy (trARPES) to measure the femtosecond charge dynamics in the topological insulator (TI) Bi$_2$Te$_3$. We detect clear CD signatures from topological surface states (TSS) and surface resonance (SR) states. In time-resolved measurements, independently from the pump polarization or intensity, the CD shows a dynamics which provides access to the unexplored electronic evolution in unoccupied states of Bi$_2$Te$_3$. In particular, we are able to disentangle the unpolarized electron dynamics in the bulk states from the spin-textured TSS and SR states on the femtosecond timescale. Our study demonstrates that photoexcitation mainly involves the bulk states and is followed by sub-picosecond transport to the surface. This provides essential details on intra- and interband scattering in the relaxation process of TSS and SR states. Our results reveal the significant role of SRs in the subtle ultrafast interaction between bulk and surface states of TIs.

The increasing quest of efficient ultrafast manipulation of spins for applications to spintronics and quantum information technology has pushed the investigation of sub-picosecond dynamics beyond traditional materials exhibiting spin order$^{1–4}$. Three dimensional topological insulators (TIs) have been the subject of such studies due to their conductive topological surface state (TSS), located within the bulk band gap, that hosts spin-polarized Dirac fermions$^{5,6}$. In TIs, the combination of spin-orbit coupling (SOC) and time reversal symmetry results in the helical spin-order of the TSS locked to the electron momentum, which leads to immunity against backscattering events$^{7–10}$. Recent observations have revealed that the spin-order of the TSS is not the only asset of TIs in spintronic engineering. The unoccupied part of the Dirac cone$^{11–13}$ and the induced Rashba splitting may open new routes for innovative devices$^{14–16}$. In addition, latest studies have reported the existence of surface resonance (SR) states with preferential spin character$^{17–20}$. SR could explain the complex interaction between TSS and bulk states, as already observed in the mixing of TSS with bulk bands (BBs)$^9,21,22$. However, previous studies did not distinguish between the sub-picosecond electron dynamics of SRs and of the nearby BBs$^{19,20,23,24}$. When a TI is optically perturbed, excited electrons decay through normally unoccupied states, including the portion of the Dirac cone above the Fermi level which is spin polarized. Thus, the electronic dynamics can be significantly affected by the spin constraints. This has motivated out-of-equilibrium experiments on TIs. In particular, time- and angle-resolved photoemission spectroscopy (trARPES) provides valuable information on electron interactions$^{23–25,30}$ as it gives access to the time evolution of excited carriers in the reciprocal space$^{26–33}$. However, detecting the spin degrees of freedom requires a more sophisticated method$^{18,34,35}$. One experimental approach is to employ circular dichroism (CD). CD-ARPES is based on the coupling between the helicity of the incident photons and the (total) angular momentum of the electrons. Here, we define CD as the normalized difference of the ARPES intensities measured with probe beams of opposite circular polarization, $\text{CD} = (I_{CR} - I_{CL})/(I_{CR} + I_{CL})$, where the subscripts CR and CL refer to right and left photon helicity, respectively. A proper interpretation of CD-ARPES can disclose relevant information on the spin of image potential states$^{36}$, magnetic doped TIs$^{37}$, Berry curvature in 2D materials$^{38}$ and on evolution of surface localization$^{39}$. The relation between CD and spin/orbital angular momentum (OAM) is highly complex$^{15,24,40–47}$ and a number of investigations reported that the CD signal might be affected by other factors, e.g. the final state effect and the experimental geometry$^{48–50}$. Therefore, the definitive link between spin-orbital texture and CD requires a comprehensive analysis and it is beyond the purpose of this work.

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Here, we exploit CD-trARPES to disentangle bulk and TSS photocurrents. The separation of bulk, TSS and SR dynamics in the trARPES signal is required in order to explain the experimental observations. We present CD-trARPES of the nonequilibrium electron and spin states in Bi$_2$Te$_3$ where only the TSS crosses the Fermi level and the insulating bulk restricts the number of relaxation channels. Our results demonstrate femtosecond decay of unpolarized BBs to TSSs and SRs followed by an electronic accumulation in TSS lasting several ps. The same approach can be generalized to study the bulk and surface electron dynamics in a wide class of TIs and open a route towards their advanced engineering for innovative opto-electronic and spintronics applications.

**Results and discussion**

Figure 1a–c show the trARPES measurements of a freshly cleaved Bi$_2$Te$_3$ sample recorded along the ΓK direction of the Surface Brillouin Zone (SBZ) in equilibrium (a), 250 fs (b) and 1 ps (c) after excitation by linearly p-polarized pump pulses with 1.85 eV photon energy. The low energy electron diffraction (LEED) pattern of the sample, reported in the upper inset of Fig. 1a, confirmed the high quality of the cleaved surface and the absence of surface reconstruction. The Dirac-cone is clearly visible in all three panels. At 250 fs delay (Fig. 1b), two unoccupied bands (B1 and B2) can be seen: one is located 0.8 eV above the Fermi level, the other at lower energy has parabolic dispersion and is well-separated from the TSS. These unoccupied states have been widely investigated in TIs, and predicted theoretically$^{23,51}$. However, their nature is still under debate: while some studies considered them as bulk states$^{23,24}$, other investigations suggested that these bands are SRs$^{20}$. We will demonstrate that the photoemission (PE) signal is detected from both SRs and BBs, therefore, B1 and B2 bands in Fig. 1b,c are the superposition of BBs and SRs. At 1 ps delay, electrons in the higher energy B1 states have almost completely relaxed, while the two bands at lower energy (B2 and TSS) are still populated, in line with the electronic dynamics of Bi$_2$Te$_3$ previously reported$^{20,23}$. Figure 1d,e show the non-equilibrium CD measured at the same delays as in panels b and c obtained as the difference between the trARPES maps measured with CR and CL light. The dichroic signal is present in all three excited bands, revealing the following features: (i) a nearly perfect anti-symmetric CD with respect to the CR and CL light. The dichroic signal is present in all three excited bands, revealing the following features: (ii) the CD decreases approaching the center of the SBZ, consistent with the previous reports on the spin structure of the TSS$^{40,41,53}$; (iii) the opposite sign of CD in B1 and B2 with respect to the one in TSS: the signs of CD for different bands corresponds to the time dependent one-step PE model calculation and spin-resolved ARPES measurements of the Bi$_2$Te$_3$ unoccupied spin structure$^{55}$.

We also measured CD-ARPES at several azimuthal angles α (see Fig. 2a and Supplementary Information$^{54}$). Previous static spin-resolved ARPES and CD-ARPES measurements reported a strong hexagonal warping of the Dirac-cone in Bi$_2$Te$_3$.$^{56}$ The distortion of the Dirac cone is amplified at larger k$_f$ and eventually creates a snowflake-shape. The warped Dirac cone leads to an out-of-plane spin component which follows a three-fold symmetry and is maximum along ΓK$^{43}$, as schematically reported in Fig. 2a. The sign-reversal of CD following a sin(3α) law was observed experimentally$^{40,41}$ and supported theoretically$^{53}$. We rotated the sample by α = 60° (see Fig. 2a), checked the orientation by LEED and repeated the CD-trARPES measurements along the direction D2 (azimuthal rotation of 60° relative to D1). Figure 2c shows the results at 250 fs delay and should be compared side-by-side with the D1 map (Fig. 2b): the sign reversal is clear. Additional measurements along other directions show consistent results according to the sin(3α) factor and the absence of CD along ΓM or when the mirror plane of the crystal matches the incidence plane (see Supplementary Information$^{55}$). We observed a similar
We first repeated the experiment with s-polarized pump beam and the results confirmed similar CD dynamics time-dependent CD induced by a linearly polarized pump pulse in order to determine the most plausible one. photon energy remain constant during the time evolution. We explored various factors which can influence the number. Note that the experimental geometry, the symmetry of initial and final states, the probe polarization and the proper spin orientation can occupy these states, resulting in a constant dichroic signal, regardless of their A stationary CD is consistent with the electronic dynamics of a spin-polarized band, since only electrons with electronic dynamics of B1 and decay convoluted with a Gaussian instrumental response function) show a fast sub 200 fs rise-time for all structures shown in Fig. 2a marked by squares, circles and triangles, respectively. The symbols "+" and "-" indicate the dynamics in the upper and lower parts of B2 (B2[0.45 eV] and B2[0.05 eV]). Since all bands are symmetric with respect to the Fermi point, we extract the electronic dynamics of each band by referring to the energy of states, i.e. B1[0.8 eV] is the B1 electronic states at 0.8 eV above the Fermi level (see Supplementary Information 54). Figure 2d reports the corresponding normalized electronic dynamics. The pump pulse fills states at higher energies first. Hot electrons relax to lower states by scattering processes. The phenomenological fittings (exponential growth and decay convoluted with a Gaussian instrumental response function) show a fast sub 200 fs rise-time for all curves with binding energies > 0.05 eV. To fit the rise-time of TSS[0.05 eV] and B2[0.05 eV], we use an additional exponential component with larger time constants of 0.72 ± 0.1 ps and 0.9 ± 0.2 ps, respectively. This behaviour can be explained by intra-band scattering of electrons towards states with lower energy, resulting in a build-up at the bottom of the bands. This accumulation of electrons is attributed to the modest electron-phonon coupling56–58 and to the lack of states/scattering channels for electron and hole recombination in an intrinsic TI 20,59–61. Despite the similar rise-times of TSS[0.05 eV] and B2[0.05 eV], the decay occurs on significantly different time scales of 7.4 ± 0.6 ps and 3.10 ± 0.26 ps, respectively. We note that B2[0.05 eV] is spin unpolarized and consequently has more scattering channels for relaxation with respect to TSS[0.05 eV]. Figure 2e shows the time evolution of CD for B1[0.8 eV], B2[0.25 eV] and TSS[0.05 eV]. While the CD signals of B1[0.8 eV] and TSS[0.05 eV] is essentially unaffected by the pump, the CD of B2[0.25 eV] shows a clear rise-time. A stationary CD is consistent with the electronic dynamics of a spin-polarized band, since only electrons with the proper spin orientation can occupy these states, resulting in a constant dichroic signal, regardless of their number. Note that the experimental geometry, the symmetry of initial and final states, the probe polarization and photon energy remain constant during the time evolution. We explored various factors which can influence the time-dependent CD induced by a linearly polarized pump pulse in order to determine the most plausible one. We first repeated the experiment with s-polarized pump beam and the results confirmed similar CD dynamics of B2[0.25 eV] (see Supplementary Information 54). Therefore, the pump induced modification of matrix elements is excluded. In addition, the effect cannot be attributed to any change of the electronic population in B2[0.25 eV]. This can be ruled out considering that, after reaching its maximum value within 600 fs past photoexcitation, the CD signal remains constant despite the almost complete loss of spectral weight (red circles in Fig. 2d,e at
The dynamics of spin population in the unpolarized B2 is mainly related to the spin-polarized SR2 and TSS. The 50 fs delay between S (B2 electrons in the PE signal) and TSS at higher energies (i.e. TSS) is improbable. The last hypothesis is that in the probed region, we detect the superposition of unpolarized B2 electrons in the PE signal and polarized SR2 electrons giving rise to the variations of CD.

In order to confirm the last hypothesis, we analyzed CD at two specific photoemission angles $\theta_1 \approx 9^\circ$ and $\theta_2 \approx 16^\circ$ as depicted in Fig. 3a (see Fig. 2a for geometry of $\theta$ rotation). Note that B2 at $\theta_1$ and TSS at $\theta_2$ have the same binding energy 0.3 eV (see the black dashed window $W$ in Fig. 3a). Figure 3b shows the energy distribution curves (EDCs) at +250 fs delay, measured at emission angles $\theta_1$ and $\theta_2$ with both CR and CL probe polarization. We emphasize that, due to energy and overlap, the PE signal from the localized SR2 states is mixed with the PE signal of B2 states. The unpolarized background (i.e. the helicity-independent spectral weight) is the contribution of B2: at angle $\theta_1$ it is mostly overlapped with SR2 and at $\theta_2$ it overlaps with TSS. TSS is in contrast with TSS at lower energies and are well separated from B2. Figure 3b shows the TSS at 0.3 eV and SR2 at 0.3 eV which display opposite CD (see the different intensities of CR and CL for each EDC in the black dashed window $W$). Figure 3c shows the dynamics of CD signal for B2 at 0.3 eV and TSS at 0.3 eV. Interestingly, the CD dynamics of TSS is different with respect to the dynamics of TSS reported in Fig. 2e. This is because TSS at higher energies (i.e. TSS) is strongly superimposed to B2 at 0.3 eV. Presumably, the presence of B2-electrons in the PE signal causes the initial variations of the CD of TSS. To clarify this point, we compare the dynamics of CDs of B2 at 0.3 eV and TSS at 0.3 eV. The B2 electrons are present in both cases and we must detect a common dynamics. We fit CDs from B2 at 0.3 eV and TSS at 0.3 eV by an exponential function obtaining similar rise-times of 93 ± 4 fs, but opposite signs, the signature of a common underlying dynamics. In the energy region marked by the box $W$, we attribute the common time-dependent signal in CDs to B2 unpolarized electrons since surface and bulk bands overlap. In fact, the constant CDs after about 600 fs show that the predominant PE signal comes from TSS and SR2 at longer delays. Therefore, the relaxation dynamics is mostly a surface mechanism in the picosecond regime. Figure 3d compares the dynamics of B2 and TSS when taking the sum of opposite helicities, $S = I_{CR} + I_{CL}$, and the difference between opposite helicities $D = I_{CR} - I_{CL}$. $S$ contains information about the dynamics of B2 at 0.3 eV (i.e. the unpolarized background). Figure 3d shows that the dynamics of S is similar for B2 at 0.3 eV and TSS at 0.3 eV (see solid symbols). This evidence suggests the presence of B2 electrons, a common spin-degenerate dynamics in both cases. Instead, D is mainly related to the spin-polarized SR2 and TSS. The 50 fs delay between S (B2 at 0.3 eV) and D (SR2 at 0.3 eV) or TSS at 0.3 eV indicates that the photoexcitation predominantly involves bulk states. Furthermore, we find exact match between the dynamics of SR2 at 0.3 eV and TSS at 0.3 eV (open symbols). Figure 3c,d show that the unpolarized
B2 states can be disentangled from SRs and TSSs on the femtosecond regime. Indeed, when electrons relax from bulk bands into the surface states, the effect of B2 in the probed region becomes less detectable. Our results suggest that in the measured energy-momentum window W, the spin-polarized surface electrons of TSS and SR2 appear with a delay after perturbation, then dwell at the surface for the entire relaxation process. These findings allow us to differentiate the complex dynamics of TSSs, SRs and BBs. Although some recent studies focused on the spin decay behavior in TIs, a complete dynamical view of the electronic redistribution upon photoexcitation cannot be achieved without considering the role of TSSs, SRs and BBs. Experimentally, we observed a much faster decay of B2 states with respect to SRs and TSSs which is attributed to the 3D unpolarized nature of bulk bands with larger number of available decay channels. This agrees with previous studies in which the electron-electron scattering rate of the bulk bands of TIs has been suggested to be an order of magnitude larger than that of the TSSs\(^2\). The electronic transition between states with opposite spins requires a spin flip event. As a result, some transitions are hindered. This is in line with our observation that the B2 and TSS remain almost isolated from each other apart from the zone center, where the decay channel from the bottom of B2 becomes effective. Thus, our analysis provide a more comprehensive picture of the spin-based relaxation mechanisms. In this context, Fig. 4 sketches the time-dependent electronic population and relaxation dynamics of Bi\(_2\)Te\(_3\). At zero delay (Fig. 4a) the pump pulse promotes electrons from the occupied valence bands to empty bulk states. Then, electrons of unpolarized bulk bands migrate to surface (-resonant) bands according to the available spin states of each one. The opposite spin texture of the TSS and SR favors inter-band scattering for one spin direction and forbids it for the opposite direction. Consequently, intra-band electron decay is enhanced with respect to inter-band relaxation and explains the lack of interaction between SR2 and TSS during the relaxation time (Fig. 4c). Our results demonstrate that in an intrinsic TI, Bi\(_2\)Te\(_3\), the picosecond relaxation process is mainly due to surface (resonant) states with strongly limited scattering channels. The complete relaxation occurring at long delays is explained by the energy transfer from the bottom of the TSS to low energy phonons\(^5\) and electron-hole recombination\(^\) at the surface (Fig. 4d).

**Conclusions**

In summary, we have investigated the ultrafast electronic dynamics of Bi\(_2\)Te\(_3\) combining circular dichroism with trARPES. The results show that the excitation with 1.85 eV pump photon energy takes place in bulk states, with a consequent ultrafast transport and redistribution of electrons at the surface. SR states act as a reservoir to accommodate the electrons with spins opposite to those in the TSS and play a key role in re-establishing equilibrium. Distinguishing between bulk and surface dynamics has a fundamental importance for TI-based spintronic devices. In this context, our technique directly maps the unoccupied band structure and extracts the femtosecond light-induced dynamics resolving different unoccupied bands. Our study opens a new route to study the time-dependent electronic behavior in the bulk and surface of other TIs. We believe our results will

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**Figure 4.** The schematic representation of the electronic distribution after excitation of Bi\(_2\)Te\(_3\) by a linearly polarized pulse. Each panel shows the dominant process. (a) First, upon the photoexcitation, the 1.85 eV pump promotes electrons above the Fermi level, hot electrons fill mainly the unoccupied BBs. (b) BB electrons scatter and move to the surface. (c) Intraband scattering process in TSSs and SRs. (d) Bottom of B2 and TSS decay by electron-hole recombination and energy transfer to phonons.
trigger future theoretical and experimental studies on SRs and their contribution to the electronic relaxation of a wide class of TI compounds.

Methods

Single crystals of Bi$_2$Te$_3$ were grown using the self-flux method. The stoichiometric mixture of high purity elements was heated to 1000°C for 12 hours and then gradually cooled down to 500°C over 100 hours before reaching room temperature. The samples were cleaved in situ and measured in ultrahigh vacuum conditions at pressure < $5 \times 10^{-10}$ mbar and at room temperature.

TrARPES experiments were conducted using a Yb-based regeneratively amplified laser system with repetition rate of 100 kHz. The pump (1.85 eV photon energy, 30 fs pulse duration and p-polarization) and probe (6.05 eV, 65 fs) pulses, impinging on the sample at an incidence angle of about 45°, were focused to spot sizes of about 100 and 50 μm, respectively. The time resolution of the experiment (temporal width of the instrumental response function) is about 80 fs, and the pump fluence $\sim 300 \mu J/cm^2$.

Photoemission (PE) spectra were recorded using a time-of-flight (ToF) analyzer with an energy resolution of about 50 meV and angular acceptance of $\sim 0.8°$. The angle-resolved maps were acquired by rotating the sample's normal with respect to the analyzer axis by 3° steps. CD data were obtained using a $\lambda/4$ waveplate on the probe beam to generate circularly polarized light. The sample orientation was checked in situ by LEED.

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Author contributions
E. C., G. C. and C. D. conceived and coordinated the project. H. H. and D. B. performed the CD and trARPES experiments. H. Y., C. C. and X. J. Z. synthesized Bi$_2$Te$_3$ single crystals. H. H. and E. C. analyzed the data and wrote the manuscript with contribution from all authors. All authors reviewed the manuscript and discussed the results.

Competing interests
The authors declare no competing interests.

Additional information
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