Dirac Bands in the Topological Insulator Bi$_2$Se$_3$ Mapped by Time-Resolved Momentum Microscopy

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The energy dispersion of the unoccupied Dirac bands of the topological insulator Bi$_2$Se$_3$ has been studied up to large parallel momenta and intermediate state energies using a setup for laser-based time-resolved momentum microscopy with 6 eV probe-photons. A strongly momentum-dependent evolution of the topologically protected Dirac states into a conduction band resonance is observed, highlighting the anisotropy dictated by the symmetry of the surface. The results are in remarkable agreement with the theoretical surface spectrum obtained from a GW-corrected tight-binding model, suggesting the validity of the approach in the prediction of the quasiparticle excitation spectrum of large system with non-trivial topology. After photoexcitation with 0.97 eV photons, assigned to a bulk valence band-conduction band transition, the out-of-equilibrium population of the surface state evolves on a multi-picosecond timescale, in agreement with a simple thermodynamical model with a fixed number of particles, suggesting a significant decoupling between bulk and surface states.

1. Introduction

At the microscopic level, the functional properties of electronic and opto-electronic devices are determined by the out-of-equilibrium dynamics of their charge-carriers. In semiconductors, for example, generation, recombination, and, in general, manipulation of non-equilibrium electron and hole distributions by external fields enable the control of the macroscopic charge-flow, ultimately leading to the desired functionality of the device. However, the charge-flow is not solely determined by the action of the external stimuli but also critically depends on the electronic structure of the material, as well as the elastic and inelastic relaxation pathways available to the excited carriers. Time- and angle-resolved photoemission spectroscopy (trARPES) allows to probe both the out-of-equilibrium electronic structure and the relaxation dynamics of non-equilibrium carrier distributions by measuring the spectral function of a material after the excitation with a femtosecond laser pulse.[12] As a result, this technique has been extensively used to investigate the electron dynamics in semiconductors,[3–5] the exciton and electron dynamics in organic adsorbates,[6,7] the carrier dynamics in 2D materials[8,9] and their unoccupied electronic structure,[10] just to name a few.

In recent years, topological materials (TI) have been the subject of extensive investigations from both the theoretical and the experimental standpoint. Part of this interest is catalyzed by the peculiar properties that arise from their out of equilibrium carrier dynamics.

In fact, the properties of the out of equilibrium carriers in topologically protected states, together with their spin–momentum locking,[11,12] a hallmark of strong spin–orbit interaction, make this class of materials an ideal candidate for spintronic applications,[13] where the functionality of the device is achieved through the control and the manipulation of non-equilibrium spin-distributions together with, or even independently from, the charge carriers’ ones. Bismuth Selenide (Bi$_2$Se$_3$) is a prototypical representative of this family of compounds, and has been studied in respect of its electronic structure,[11,12] transport,[14–16] and optical properties,[17] as well as its non-equilibrium carrier dynamics.[18–24] Moreover, it has already been integrated into several experimental spintronic,[25,26] optoelectronic,[27] and opto-spintronic devices.[28]

Numerous trARPES investigation addressed the dynamics of the Dirac fermions finding unusually large population lifetimes. This dynamics, which is partially supported by the relaxation of the photoexcited electrons from the bulk bands to the surface state,[19] is consistent with the cooling of an hot-electron population via electron-phonon scattering processes,[19,20,22,23] but the relaxation rate and the magnitude of the phonon assisted surface-bulk coupling were found to vary due to a complex interplay between the level of doping and the temperature of the lattice.[20] Other trARPES measurements shed light on the
out-of-equilibrium electronic structure of the material, demonstrating the presence of a second, unoccupied, Dirac surface state directly accessible through an optical transition.\cite{21,23} The interest in the relaxation dynamics of the Dirac fermions, their coupling with the bulk bands, and their optical excitations stems mostly from the possibility they represent to drive spin-polarized currents in a contact-free manner through the circular photogalvanic effect.\cite{29} Evidences of this phenomenon were found in opto-electronic devices where the direction of the photocurrent is controlled by the helicity of the impinging light.\cite{28,30} and later in all-optical\cite{31} and trARPES experiments.\cite{32,33} The peculiar properties of the helical fermions have been also invoked to rationalize the optical properties of Bi$_2$Se$_3$, where the presence of chiral surface excitons has been suggested,\cite{34} while the topology and the linear dispersion of the Dirac state plays a major role in the efficient high-harmonic-generation observed in Bi$_2$Se$_3$ illuminated by strong mid-infrared fields.\cite{35}

All these findings root their origin in the properties of the topologically protected Dirac states which in reality deviate from the ideal behavior at large parallel momenta\cite{36} and are eventually expected to resonate with the edge of the bulk conduction band,\cite{37} losing their spin-momentum locking character.\cite{38} Despite its importance, the dispersion of the surface state at large crystal momenta was never previously accessed with trARPES.

In this work, we close this gap by performing high-resolution photoemission spectroscopy measurements of the occupied and unoccupied surface state manifold of a Bi$_2$Se$_3$(0001) surface up to 1.1 eV from the Dirac point and ±0.5 Å⁻¹ in the parallel momenta. The experiments are performed with an apparatus that couples a widely tunable pump–probe beamline with an innovative ARPES detector: a photoemission electron microscope (PEEM) operating in momentum mode,\cite{39} also called momentum microscope (MM). With its ultimate angular acceptance, this instrument allows the acquisition of the entire photoemission horizon generated by a 6 eV femtosecond-laser probe, merging the high-resolution capabilities of low-photon-energy ARPES\cite{39} with the wide momentum field-of-view typical of higher energy photon sources. We compare our findings with state-of-the-art theoretical predictions of the Bi$_2$Se$_3$(0001) surface spectrum\cite{40} and find a remarkable agreement between the calculated and the experimental quasiparticle spectrum when the GW approximation is employed.

### 2. Results and Discussion

Before addressing the unoccupied electronic structure, we perform a conventional (static) ARPES characterization of the Bi$_2$Se$_3$(0001) surface. The results are reported in Figure 1.

The ARPES map in Figure 1a clearly shows the topologically protected surface state of Bi$_2$Se$_3$(0001) surface, which manifests as linearly dispersing bands in the energy gap between valence and conduction bulk states. In this sample the Dirac point (DP), where ideally the density of states of the surface state should vanish, falls 0.46 ± 0.05 eV below the Fermi edge ($E_F$) and is taken as the origin of the energy scale. The presence of occupied bulk conduction-band states, revealed as the parabolic feature encompassed by the linear dispersing bands between 0.26 and 0.46 eV, highlights the intrinsic, degenerate, n-doping of the sample, which is believed to stem from defects in the crystal structure, such as selenium vacancies.\cite{42} In this work, we refer the energy scale to the Dirac point to facilitate the comparison among the datasets and the theoretical predictions, as well as in view of the peculiar effect of the absorption of contaminants from the residual gas in the UHV environment drives on the Bi$_2$Se$_3$ surface. In fact, on this surface, the doping level is strongly influenced by the adsorption of contaminants, which induces band-bending and, eventually, leads to the emergence of a quantum-confined two-dimensional electron gas at the surface (2DEG).\cite{43,44} This phenomenon drives significant variations of work function and in the state-binding-energies as the adsorbates accumulate over the sample surface, making the Fermi energy a less than ideal reference for comparisons among different datasets. Moreover, in order to mitigate these drawbacks, we performed measurements on highly n-doped samples, where the effect of additional contamination is less severe.\cite{43} The momentum distribution maps (Figure 1b) collected above the Dirac point highlight the well-known hexagonal warping of the surface state: the ring-like feature of the unperturbed surface state ($E = E_{DP} = 0.21$ eV) evolves into a hexagonal pattern as it approaches the Fermi level of the doped semiconductor. This phenomenon, which is due to the threefold rotational symmetry of the surface,\cite{45} is more evident where the surface state branches become degenerate with the bulk states ($E = E_{DP} = 0.3$ and 0.46 eV). The corners of the hexagonal features fall along the $\Gamma - M$ directions and are exploited to determine the crystal orientation throughout the experiments. Our observation agrees with numerous precedent ARPES measurements.\cite{11,12,16} However, despite these results being established nowadays, the intriguing properties of topological matter are still driving the development of new approaches to rationalize the experimental evidences. One of such approaches, proposed by Aguilera et al.,\cite{40} makes use of many-body corrected tight-binding Hamiltonians generated for topological insulators of the Bi$_2$Se$_3$ family. The advantage of such an approach is the possibility of indirectly introducing the exchange and correlation effects, obtained with the GW method,\cite{46} to the electronic structure calculations of thick slabs and semi-infinite systems. Such an endeavor is beyond the current computational capabilities when DFT or GW methods are applied explicitly, while it can be tackled with a tight-binding approach employing the parameters obtained from DFT or GW calculations performed on a bulk system. Figure 1c,d shows the surface spectral function, i.e., the imaginary part of the surface Green function, of a Bi$_2$Se$_3$(0001) semi-infinite slab obtained through this approach. For a better comparison, it is displayed in the same energy and momentum regions probed by our experiment. The calculations are performed with the iterative Green-function method, as implemented in the software package WannierTools,\cite{47} starting from the real-valued Hamiltonian of Bi$_2$Se$_3$ published in ref.[40]. The excellent agreement between the experimental (Figure 1a) data and the theoretical prediction (Figure 1c) is evident in the dispersion of the surface state along the $M - \Gamma - K$ directions, while a small deviation in the onset of the conduction band edge is observed. This mismatch is in the order of 50 meV and is comparable to the energy resolution of the instrument (see Section S1, Supporting Information). Therefore, it can be related to the uncertainty in the Dirac point energy, which we extracted from the data. The presence of a 2DEG on the surface\cite{43} could also contribute to the observed deviation as the effects of the
electron-doping are not included in the calculations. The overall agreement between the experimental and theoretical predictions is also further highlighted by Figure 1d, that reports calculated momentum distribution maps to be compared to the experimental ones in Figure 1b.

Having characterized our sample in the static photoemission regime, we now address its unoccupied states manifold. To this end, we illuminate the surface with a 0.97 eV pump beam at a fluence of 310 μJ cm⁻² (see Experimental Section). Upon the arrival of the pump pulse, an electronic population is promoted to both the unoccupied part of the surface states and the bulk conduction band. We acquire an energy–momentum space snapshot of this out-of-equilibrium population 0.77 ps after the zero-delay condition between the pump and the probe pulses to ensure the complete thermalization of the electron distribution,¹⁹ to avoid any effect induced by the temporal overlap,⁴⁴ and to archive a statistical occupation of all the states within our measurement window (see Supporting Information).

Our findings, together with the theoretical predictions based on the approach introduced in the previous paragraphs, are
summarized in Figure 2. Figure 2a shows the ARPES map (along the $\Gamma - \Gamma - \Gamma$ directions) of the transiently occupied states located from 0.5 to 1.1 eV above the energy of the Dirac point. The linear-dispersing signature of the surface state continues, almost unaltered, along the $\Gamma - \Gamma - \Gamma$ direction and stays visible up to an energy $E - E_{DP}$ of 0.8 eV. Above this energy, the photoemission intensity drops, and only a diffused background extends up to the edge of the measurement window. Along the $\Gamma - \Gamma$ direction, the signature of the surface state is more evident but deviates from the linear dispersion and evolves in a local maximum at 0.37 Å$^{-1}$. At the limit of the momentum window, 0.49 Å$^{-1}$, a local minimum can also be discerned. The structure of the unoccupied state manifold is further highlighted in Figure 2b where the momentum distribution maps, collected at three selected energies from the Dirac point, are reported. The threefold rotational symmetry of the surface is evident in all the maps, and the distortion due to the hexagonal warping develops into a six-pointed-star shaped feature as the intermediate state energy increases. To our knowledge, this region of energy-momentum space was never probed in laser-based time-resolved ARPES experiments on Bi$_2$Se$_3$ surfaces. Moreover, although predicted by the theoretical calculations no direct evidences of such states were previously reported even in ARPES studies with femtosecond XUV radiation.\cite{22} Figures 2c and 3d display the theoretical ARPES map and the theoretical momentum distributions maps, respectively. A simple visual comparison reveals how the theoretical model reasonably grasps the experimental observations. In particular, we observe a remarkable agreement between the experimental data and the theoretical predictions along the $\Gamma - \Gamma$ direction, where the local maxima and minima sequences well match in both the energy and the momentum coordinates. The agreement is less obvious along the $\Gamma - \Gamma$ direction, as the intensity of the photoemission signal drops significantly for energies above $E - E_{DP} = 0.8$ eV. However, below this energy, two linearly dispersing features are discernible.

In ref. \cite{40}, a tight-binding simulation performed on a 100 quintuple-layers slab of Bi$_2$Se$_3$ (0001), employing the same many-body corrected Hamiltonian, is reported. Not surprisingly, the prediction of the of the surface spectrum calculations and the results of such a large slab model show a remarkable agreement. We thus rely on the results of the slab model to classify the features found in the photoemission data with respect to their...
degree of localization in the first quintuple layer. Following the results of ref. [40], we assign the feature labeled as 1 in Figure 2c as the true surface state, which is predicted to retain a strong localization on the first quintuple layer up to energies above 1 eV from the Dirac point. Feature 2 (Figure 2c) has a bulk-conduction-band nature with some minor contributions from surface resonances. Along the $\Gamma - M$ direction, the situation is dramatically different as the surface state quickly evolves into a surface resonance (feature 3 Figure 3c) and merges into an edge of the bulk conduction band (feature 4 Figure 3c) already at 0.7 eV from the Dirac point, losing its surface localization. Some of the differences observed between the data and the theoretical predictions, such as the drop in the intensity of the surface state along the $\Gamma - K$ direction, may be rationalized in terms of matrix-element or final-state effects.

The latter are, in general, important in low-photon-energy photoemission as the final state cannot be approximated by the nearly free-electron model.

A good agreement between GW predictions and ARPES data is not necessarily surprising. However, here, we observe a correspondence between the theoretical surface spectrum and the ARPES distribution of an out-of-equilibrium system, whose spectral function was perturbed by a femtosecond optical excitation prior to the electron-removal step. This correspondence implies a small electron–hole interaction, i.e., negligible excitonic effects, that might be screened by the background electron population of the degenerate, n-doped, surface. Moreover, the results of Figure 2c,d are not obtained through an explicit GW calculation of the surface but indirectly introduced as correction in a parametrized tight-binding Hamiltonian on a Wannier functions basis.[40] Therefore, our results testify to the validity of this computationally less expensive approach in approximating the effects of many-body interactions on the Bi$_2$Se$_3$ surface.

Before concluding, we briefly discuss the picosecond dynamics of the photoexcited population. The ultrafast electron dynamics of Bi$_2$Se$_3$ has already been the subject of numerous tr-ARPES investigations.[18–24,32] A coherent picture of the basic underlying processes emerges from these studies: depending on the doping of the sample, and on the photon energy, upon the arrival of the pump pulse interband and intraband transitions populate both the surface state and the conduction band states above their equilibrium chemical potential. Irrespective on the transition pathway the population thermalizes, via electron–electron scattering, to a Fermi–Dirac distribution with a high temperature and a new effective chemical potential within 40 fs[19,32,49] from the initial excitation. The excess energy is subsequently dissipated to the lattice, on a picosecond time scale, through electron-phonon scattering and eventually diffuses away from the photo-excited region restoring the original equilibrium condition. In the present study, the 0.97 eV optical excitation is expected to couple valence to conduction band states[30,51] and we address the cooling dynamics of the hot electron distribution in the whole energy–momentum region accessed by our setup. The experimental data and the details of the analysis are reported in Section S1 (Supporting Information). In particular, by following a procedure similar to the one introduced in ref. [20], we quantify the temperature and the transient chemical potential of the electron population in the topologically protected surface state and in the conduction band resonances. Upon the optical excitation the electronic temperature increases to about 900 K and is accompanied by a transient reduction of the chemical potential in the order of 30 meV. The electronic temperature relaxes with an exponential time constant of $\tau = 3.57 \pm 0.09$ ps, revealing an electron–phonon scattering rate of $0.285 \pm 0.007$ THz, in good agreement with previous investigations of highly n-doped samples.[20] The negative variation of the chemical potential suggests that the interband transition at 0.97 eV is unable to significatively alter the total population in the Dirac cone, despite the surface–resonance nature of the states along the $\Gamma - M$ direction. Moreover, despite their large excess energy with respect to the equilibrium chemical potential the surface resonances present long population dynamics, in the order of 500 fs.

3. Conclusion

We have mapped the band dispersion of the unoccupied states in a prototypical topological insulator up to 1.1 eV from the Dirac point and 0.5 Å$^{-1}$ in parallel momenta, close to the boundary of the photoemission horizon from the 6 eV probe. Our measurements highlight the momentum–dependence evolution of the Dirac cone into surface resonances at the edge of the conduction
band, as predicted by a state-of-the-art theoretical model of the surface's electronic structure. The agreement between the theoretical model and the experimental data suggests that in the highly n-doped Bi$_2$Se$_3$, the effects of the electron–hole interaction in the photoexcited state are of small magnitude. This result is surprising considering the difference in the values of the band gap found among ARPES$^{[52]}$ and optical studies,$^{[53–55]}$ which is in the order of 100 meV, and hints to a stronger electron–hole interaction. Our results may also be beneficial in the interpretation and the exploitation of high harmonic generation processes in Bi$_2$Se$_3$ and in the optical pumping of spin polarized current, as these phenomena are known to critically depend on the band structure and on the helical nature of the Dirac fermions.

In general, the necessity to probe wide energy–momentum volumes in time-resolved experiments is one of the driving forces behind the rapid diffusion of trARPES setups based on femtosecond extreme ultraviolet light sources, which can illuminate energy-momentum regions inaccessible to low-photon-energy ARPES. Despite the advantages of these last-generation systems, we argue that combining a momentum microscopy apparatus with a conventional laser-based source is a viable approach to probe the out-of-equilibrium electronic structure in a wide momentum field of view, covering a large portion of the surface Brillouin zone of many crystalline materials.

4. Experimental Section

The experimental setup was developed upon two commercially available sub-systems: a computer-controlled, turn-key, femtosecond laser source from Light Conversion and a KREIOS PEEM manufactured by Specs GmbH. A simplified diagram is reported in Figure 3.

The laser system was coupled to the photoemission end-station in a conventional pump–probe configuration. The primary light source was a 20 W, diode-pumped Ytterbium-based amplified laser emitting nearly Fourier-transform-limited femtosecond pulses with a central wavelength of 1028 nm and a time duration of 300 fs FWHM (Pharos). This laser source allowed on-demand selection of the pulse repetition rate up to 1 MHz. A substantial fraction of the output beam was fed to an optical parametric amplifier (OPA) (Orpheus-F) to generate the optical pump employed in the time-resolved experiments. An ancillary second harmonic generation unit (Lyra) extended the tuning range of the system to the near-UV wavelength allowing an almost seamless selection of the pump photon energy between 0.5 and 3.5 eV with micro-joule-level pulse energies at 208 kHz.$^{[54]}$

A fifth harmonic generation stage (Hiro) converted a fraction of the fundamental beam into the 6 eV probe. Thanks to its high conversion efficiency (≥1% at 208 kHz), the harmonic stage accepted a wide range of pulse energies at its input and can operate at high repetition rates. This feature in static photoemission experiments to increase the photoelectron count rate without exacerbating the space charge effects was exploited.$^{[57,58]}$ In fact, in the static mode, the constraint imposed by the OPA operation was lifted, and by setting a 1 MHz repetition rate, an almost fivefold increase in the photoemission intensity was achieved without variations in the photoelectron-cloud density. The repetition rate can be switched through a computer-controlled interface without requiring changes in the optical system.

In the PEEM instrument, an immersion lens column generates an image of the lateral distribution of the photoelectrons (x) and an additional Fourier lens transforms it into an image of the photoelectron emission angles. The latter contains the momentum distribution of the photoelectrons I(Κ, Κ) with high angular resolution.$^{[59]}$ The photoelectrons were filtered by a hemispherical-analyzer section of the instrument, which thus acquires momentum-resolved two-dimensional (2D) maps at a fixed, and selectable, kinetic energy (E,Κ,Κ).

Clean Bi$_2$Se$_3$(0001) surfaces were prepared in situ by cleaving a commercially available crystal (2D Semiconductors) under high vacuum conditions (base pressure ≈10$^{-8}$ mbar) and were transferred to the analysis chamber (base pressure <1×10$^{-9}$ mbar) without additional treatments. The sample was cooled down with liquid helium throughout all the measurements, reaching a temperature of 8 ± 1 K at the position of the sample holder, and monitored with a temperature controller (Lake Shore model 325).

Both the ARPES and trARPES spectra were collected with a pass energy of 50 eV and the narrowest slit (0.2 mm) at the entrance of the hemispherical analyzer. The choice of these parameters was dictated by the necessity to find the best compromise between the energy resolution and the transmission function of the analyzer. A high transmittance was necessary in the trARPES experiments to acquire the transients within the lifetime of the surface in the UHV environment and with low-illumination levels to suppress space-charge effects. In the instrument, setting a pass energy of 50 eV was given the best results; and can be rationalized considering the linear scaling of the energy resolution with the pass energy versus the power-law behavior of the transmission function with this parameter.$^{[60]}$

A field aperture was introduced in the intermediate image plane of the microscope column to restrict the area of the sample contributing to the signal (diameter: 30 μm) and select a uniform region of the surface. Due to the low photon energy employed for the pump pulse, which restricted the range of accessible parallel momenta, and to the limited extent of the surface state in the reciprocal space, the instrument was operated with high momentum–magnification factors, which corresponded to a k field-of-view of either ±0.5 Å$^{-1}$ (MAG.4) or ±0.2 Å$^{-1}$ (MAG.5). The momentum coordinates were calibrated by fitting the boundaries of the photoemission horizon with the free-electron dispersion relation. For all the measurements reported in this work, the energy resolution of the apparatus obtained by fitting the fermi edge of a Bi$_2$Se$_3$ sample, was found to be in the order of 50 meV (see Supporting Information) while the momentum resolution was estimated in 0.02 Å$^{-1}$ FWHM from the linewidth of the Dirac state features in the momentum distribution maps.

The mapping of the occupied states band structure was performed by illuminating the sample with the p-polarized 6 eV probe only and at a repetition rate of 1 MHz. In the measurements of the unoccupied states band structure and of the photoexcited carrier dynamics instead, the repetition rate of the laser system was set to 208 kHz. The pump–photon energy was tuned to 0.97 eV and its fluence set to 310 μJ cm$^{-2}$. Both the pump and the probe beam were p-polarized. Being interested in the unoccupied electronic structure at the edges of the conduction band the photon energy was chosen to be resonant with a weak optical absorption band in Bi$_2$Se$_3$,$^{[51]}$ and assigned to the valence to conduction band transition.$^{[50,51]}$ Due to the large work function of the sample, which was found to be in the order of 5.2 eV, spurious multiphoton photoemission from the irradiation with low-energy photons was suppressed. As a result, the nonlinear-photoemission background from the pump beam alone was below the dark-count rate of the photoelectron detector throughout the experiments. The pump–probe crosscorrelation was obtained by fitting a fast transient attributed to the photoemission from an image potential state of the Bi$_2$Se$_3$ surface with a Gaussian envelope, and resulted in 580 fs FWHM (see Supporting Information).

The theoretical calculation reported in Figures 1c,d and 2c,d was obtained starting from the real-valued many-body corrected tight-binding Hamiltonian published by Aguiler et al.$^{[40]}$ The Hamiltonian was used as an input file for the post-processing program WanierTools$^{[47]}$ together with the lattice vectors, the atomic positions and the value of the Fermi energy were published in ref. [40]. This post-processing software is capable to calculate the imaginary part of the surface Green’s function for surfaces of arbitrary mirror indices using the iterative Green’s function method.$^{[61]}$ This quantity can be regarded as probability to remove an electron with a defined energy and momentum from the many-body system and approximates the photoemission current as long as the matrix elements of the electric dipole transition between the initial and the final (photoemitted) state are neglected.$^{[62]}$ The number of principal layers$^{[61]}$ was set to 2 throughout the calculations.
Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The raw data that support the findings of this study are available in the ZENODO database, at https://zenodo.org/communities/interfast-fetopen.

Keywords

bismuth selenide, momentum microscopy, quantum materials, time-resolved ARPES, topologically protected surface states

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