In spintronics, the two main approaches to actively control the electrons’ spin involve static magnetic or electric fields. An alternative avenue relies on the use of optical fields to generate spin currents, which can bolster spin-device performance, allowing for faster and more efficient logic. To date, research has mainly focused on the optical injection of spin currents through the photogalvanic effect, and little is known about the direct optical control of the intrinsic spin-splitting. To explore the optical manipulation of a material’s spin properties, we consider the Rashba effect. Using time- and angle-resolved photoemission spectroscopy (TR-ARPES), we demonstrate that an optical excitation can tune the Rashba-induced spin splitting of a two-dimensional electron gas at the surface of Bi2Se3. We establish that light-induced photovoltage and charge carrier redistribution - which in concert modulate the Rashba spin-orbit coupling strength on a sub-picosecond timescale - can offer an unprecedented platform for achieving optically-driven spin logic devices.
Spintronics has the potential to deliver computational devices that are less volatile, faster, and more energy efficient with respect to their electronic counterparts. However, the need to control the spin degree of freedom in a fast and efficient manner is challenging, as the field required to flip the electron’s spin in magnetic materials is often prohibitively high. Spin-orbit coupling (SOC) effects, such as the Rashba effect, the formation of spin-polarized electron states without a magnetic moment, thereby circumventing this limitation. In particular, the Rashba effect manifests as a broken spin degeneracy at semi-

where $ER$ is the strength of the Rashba SOC and $\Delta k_R = \alpha_k L$ is the momentum splitting. The system is then optically excited with a near-infrared pump pulse and its response is probed by photoemission spectroscopy (TR-ARPES) to track the evolution of the RSOC strength through the dispersion of the Rashba 2DEGs. By directly measuring $\Delta k_R$ and $\Delta E_R$ as a function of pump-probe delay, we unambiguously extract the evolution of $\alpha_k$.

**Results**

Among the materials that can host Rashba-split 2DEGs, bismuth-based topological insulators (TI) are an ideal platform: 2DEGs can be induced on the surface of TIs by applying a positive surface bias or chemical gating. The combination of the strong atomic SOC in TIs with surface gating generates a substantial Rashba effect in the 2DEGs, allowing one to finely resolve the spin splitting. In an ideal TI, only the topological surface state (TSS)—recognizable by its linear dispersion across the bandgap—crosses the Fermi level ($E_F$), and all charge carriers belong to the TSS. As represented in Fig. 2a, the application of a sufficient positive bias at the surface induces a strong band bending, leading to the creation of 2DEGs in the form of surface confined quantum well states (QWSs). While the TSS wavefunction extends only within a few layers from the surface and does not depend on the shape of the surface potential, the wavefunction of the QWS does, and extends comparatively deeper into the bulk. The difference in spatial extent between the TSS and QWS wavefunctions allows us to extract the behavior specific to QWSs, as opposed to the behavior of surface states in general.

Our experimental approach is depicted in Fig. 2b. We choose p-doped Bi$_2$Se$_3$ to host QWSs, as the hole doping provides a lower bulk conductivity in this material. The 2DEGs are prepared by depositing a controlled amount of alkali atoms on the surface, leading to a population of conduction band-derived states that are spin-split by the Rashba effect. Increasing the concentration of deposited atoms is analogous to raising the surface bias, which introduces a higher surface charge density and stronger band bending. The system is then optically excited with a near-infrared (1.55 eV) “pump” pulse and its response is probed by photoemission using a UV (6.2 eV) pulse at variable time delay $\Delta t$.
The result of such a TR-ARPES experiment is summarized in Fig. 2c over a long range of delays. The left and right panels show the ARPES spectra at negative delay (−100 ps), and 500 ps after the pump arrival, respectively. The central panel presents the evolution of the states at the Brillouin zone center (black dashed lines) is shown as a function of time. The dispersion, Rashba-splitting, and spatial extent of the 2DEGs depend on the detailed shape of the band bending. Here, the band bending pushes the two lowest QWSs (blue and green) below the Fermi energy. TR-ARPES experiment on p-type Bi₂Se₃; the cleaved sample is gated in situ by alkali atom deposition. A near-infrared (1.55 eV) “pump” pulse perturbs the system, and a UV (6.2 eV) pulse is used to probe the electronic structure by ARPES. The time delay (Δt) between pump and probe pulses is varied to resolve the electron dynamics. Temporal evolution of the QWSs in p-doped Bi₂Se₃ plotted relative to the electron quasi-Fermi level $E_{\text{Fq}}$. The left panel shows the ARPES spectra of all surface states before pump arrival (−100 ps); in the center panel photoemission intensity integrated around the Brillouin zone center (black dashed lines) is shown as a function of time (pump and probe are overlapped at time zero, red dashed line). We observe that a second QWS emerges after the pump excitation; the right panel shows the dispersion at 500 ps, characterized by two partially populated QWSs.

The effect of HC on the surface potential can be extremely complex, but it is likely that, the mobile hot carriers further screen the built-in electric field, causing the QWSs’ shift to higher energy. The second and more interesting process is a long-lasting shift of the QWSs to lower energy, that emerges as a consequence of an increase in the surface electron population and variation of the electrostatic environment. This process—as we will show in detail—is given by a photovoltage (PV) effect, and it is the central mechanism of this work. The effect of the PV arises within a few hundred femtoseconds and alters the energy and density of the QWSs over hundreds of picoseconds. As the PV alters the electrostatic environment at the surface, we expect it will have an impact on the Rashba effect as well. For a quantitative look at the momentum splitting, we plot in Fig. 3c the momentum distribution curves (MDCs) of QWS1 at $E_{\text{Fq}}$ in equilibrium (before the pump arrival) and 8 ps after the excitation. The MDCs span the two spin-polarized bands on the right-hand side of QWS1 (see red dashed line in Fig. 3a) and are referenced to the Fermi momentum of the inner branch ($k_{\text{F}}$). The MDC peak locations are indicated by dashed lines; in comparing the equilibrium (purple) and post-excitation (blue) MDCs, we observe that the momentum splitting of the carriers is reduced from $(26.0 \pm 0.5) \times 10^{-3}$ to $(22.5 \pm 0.5) \times 10^{-3}$ Å⁻¹. A similar result is observed for the energy splitting: in Fig. 3d, we plot the energy distribution curves (EDCs), for the same two delays, along the cut shown in Fig. 3a; we find that, whereas the outer branch of QWS1 maintains its position, the inner branch moves to lower energy, leading to a reduction of the energy splitting $\Delta E_R$ by
approximately 13 meV. The additional shoulder observed in the EDC at 8 ps arises from QWS2, which also moves to significantly lower energy. The simultaneous reduction of both $\Delta k_R$ and $\Delta E_R$ is a clear indication of an optically driven change of the Rashba spin-orbit coupling strength $\alpha_R$, and excludes modifications of the electron dispersion and effective mass as relevant contributions. The full temporal dynamics of the Rashba effect in QWS1 under optical excitation is given in Fig. 3e, where the splitting in momentum $\Delta k_R$ (in orange), as well as the RSOC strength $\alpha_R$ in QWS1 calculated from Eq. (1) and extracted by performing a posteriori analysis on the data. The momentum splitting $\Delta k_R$ is the distance between two peaks of the same MDC, and it is dynamically reduced with the pump excitation from $(26 \pm 0.5) \times 10^{-3}$ (at $\Delta t < 0$) to $(22.5 \pm 0.5) \times 10^{-3}$ Å$^{-1}$ (at $\Delta t = 8$ ps). The optical excitation also induces the reduction of the energy splitting. EDC and MDC profiles in the right branch of QWS1 (horizontal dashed line in (a)) at $E_F$ at equilibrium (purple) and after 8 ps (blue) relative to the Fermi wave-vector of the inner branch $k_F$; the solid lines are Voigt fits to the data. The momentum splitting $\Delta k_R$ is the distance between two peaks of the same MDC, and it is dynamically reduced with the pump excitation from $(26 \pm 0.5) \times 10^{-3}$ (at $\Delta t < 0$) to $(22.5 \pm 0.5) \times 10^{-3}$ Å$^{-1}$ (at $\Delta t = 8$ ps). The optical excitation also induces the reduction of the energy splitting. EDC and MDC profiles in (c and d) have been deconvolved by the energy resolution via Lucy–Richardson algorithm (Ref. 47) for better clarity. Both profiles are fitted using Voigt functions (solid lines). a Temporal evolution of the RSOC strength $\alpha_R$ in QWS1 calculated from Eq. (1) and extracted by fitting the momentum (orange) and energy (green) splitting at several time delay; $\alpha_R$ is reduced by about 0.1 eVÅ at 8 ps with respect to equilibrium. The values of $\Delta k_R$ and $E_F$ are explicitly plotted against the right axes. At $0 < \Delta t < 2$ the signal is too low to convey reliable physical significance. All the values of energy and momentum splitting are obtained by fitting the raw data, and error bars are evaluated from statistical distribution within 95% confidence.

Fig. 3 Ultrafast response of Rashba QWSs to optical perturbation. a ARPES dispersion at negative time delay (before pump arrival), at time zero (pump and probe fully overlapped), and at 8 ps. The latter two are accompanied by differential spectra obtained by subtracting the spectrum acquired at around $-0.5$ ps; the blue (red) color is indicative of a pump-induced decrease (increase) of photoemission intensity. b Temporal evolution of the energy minimum for QWS1 and QWS2 extracted from fitting the ARPES data at $k_F = 0$. The fit to the experimental data (solid black line) stems from two contributions, each consisting of a finite rise-time step function and an exponential decay. The positive contribution (purple) is defined as a hot-carriers (HC) driven process, which is short-lived, the negative contribution (cyan) is the result of a photovoltage (PV) effect. c Momentum distribution curves (MDC) profiles across the right branch of QWS1 (horizontal dashed line in (a)) at $E_F$ at equilibrium (purple) and after 8 ps (blue) relative to the Fermi wave-vector of the inner branch $k_F$; the solid lines are Voigt fits to the data. The momentum splitting $\Delta k_R$ is the distance between two peaks of the same MDC, and it is dynamically reduced with the pump excitation from $(26 \pm 0.5) \times 10^{-3}$ (at $\Delta t < 0$) to $(22.5 \pm 0.5) \times 10^{-3}$ Å$^{-1}$ (at $\Delta t = 8$ ps). d Energy distribution curves (EDC) profiles across the inner and outer branch of QWS1 at $k_F = 0.05$ Å$^{-1}$ (vertical dashed line in a) before pump arrival (purple) and after 8 ps (blue). The optical excitation also induces the reduction of the energy splitting. EDC and MDC profiles in (c and d) have been deconvolved by the energy resolution via Lucy–Richardson algorithm (Ref. 47) for better clarity. Both profiles are fitted using Voigt functions (solid lines). e Temporal evolution of the RSOC strength $\alpha_R$ in QWS1 calculated from Eq. (1) and extracted by fitting the momentum (orange) and energy (green) splitting at several time delay; $\alpha_R$ is reduced by about 0.1 eVÅ at 8 ps with respect to equilibrium. The values of $\Delta k_R$ and $E_F$ are explicitly plotted against the right axes. At $0 < \Delta t < 2$ the signal is too low to convey reliable physical significance. All the values of energy and momentum splitting are obtained by fitting the raw data, and error bars are evaluated from statistical distribution within 95% confidence.

excitation, requires one to consider the detailed variation of the surface electric field in relation to the spatial electron distribution. To this end, we build a model to capture the salient aspects of the experimental results. We begin by calculating the band bending of the system at equilibrium, which can be described by a one-dimensional model in the out of plane direction $x$. The potential profile $V(x)$ is calculated by solving the Poisson equation within a modified Thomas-Fermi approximation$^{29,30}$. The binding energy and wavefunction of the QWSs is computed $a posteriori$ by solving the Schrödinger equation within the calculated $\text{scr}$ parameters for the calculation are taken from Refs. 31–34, and the surface potential $V_0$ is determined empirically by the shift of the TSS Dirac point induced by chemical gating (see Supplementary Information).

We present the calculated equilibrium potential and QWSs’ energy minima in Fig. 4a. The space charge region (SCR) spans more than 30 nm, and QWS1 and QWS2 are partially populated, replicating the experimental observations of Fig. 3. Following an optical excitation across the band gap, the generated electron-hole pairs within the SCR are swept apart by the electric field $E_{\text{SCR}}$, which pushes the negative charges towards the surface and the positive charges towards the bulk. The electrons and holes become spatially separated over tens of nanometers, giving rise to a long-lasting photovoltage field $E_{PV}$ of the opposite sign to $E_{\text{SCR}}$. This effectively softens the band bending, pushing the surface...
potential $V_0$ to less negative values, as shown in Fig. 4b. The new shallower surface potential drives both the QWSs and the E$_{Fn}$ to higher energy. To accommodate the surplus of surface electric charge, the E$_{Fn}$ shifts further upwards, resulting in the QWSs moving to more negative energies when plotted with respect to E$_{Fn}$ as seen in the TR-ARPES data (details in Supplementary Information). It must be noted that, as small surface state-induced band bending is a common feature in semiconductors, surface PV has previously been observed in pristine TIs.  

However, while this is technologically relevant for TIs—because it leads to spin-polarized diffusion currents—the TSS only undergoes a rigid shift in energy under the surface PV. The 2DEGs, on the other hand, are much more sensitive to the shape and magnitude of the confining potential $V(x)$, and by extension, the PV. The full temporal dynamics of the QWSs has been simulated by introducing a pair of effective photo-charges in the system, which approximates the collective charge motion via a center of mass approach. At each time step, the charge distribution, electric field and E$_{Fn}$ are reevaluated. The time evolution of the QWSs’ energy with respect to the E$_{Fn}$—shown in Fig. 4c—is in good qualitative agreement with experimental data. Both QWS1 and QWS2 fall to more negative values almost instantaneously at positive delays. Consistent with experimental observations, the energy shift of QWS2 is larger than that of QWS1. This is a manifestation of the shallower confining potential, which allows for a smaller energy difference between consecutive QWSs. Lastly, the RSOC strength and, by extension, Rashba-splitting for QWS1 are calculated at each step from Eq. (1). The evolution of $\Delta k_R$ is also consistent with experimental findings (Fig. 4d), confirming that the reduction in the Rashba strength is due to the PV-induced softening of the surface potential.

With the exception of the fluctuation at early delays given by the presence of hot carriers, our simple model succeeds in reproducing all salient features of the experimental data, proving that a photovoltage is responsible for the observed behavior. The small quantitative deviations between the simulated and measured PV effect can be attributed to the model simplicity and approximations, such as the omission of changes in the screening and dielectric properties of the material induced by the PV. More complex simulations of the PV effect such as those recently developed in ref. might further improve quantitative accuracy. Nevertheless, the model captures the fundamental observations of the experiment and provides a clear explanation of the underlying mechanisms. The simulated spectral function in the inset of Fig. 4d showcases the calculated dispersion of QWS1 at the two representative time delays, highlighting the faithful reproduction of the TR-ARPES data. Finally, the PV model can also account for the long timescale (950 ps) needed to recover equilibrium conditions (Fig. 2c), as the spatial separation of electrons and holes in the SCR drastically reduces the recombination rate. The same timescale is expected for the Rashba splitting to recover its initial value as it is modified by the same effect. Since the return to equilibrium is ultimately determined by the charge carriers’ diffusion from the illuminated area, the lifetime of the PV effect could in principle be tuned by varying the size of the pump beam.
Methods

Samples of Ca-doped Bi$_2$Se$_3$ are synthesized as described in Ref. 41. Here, Ca acts as acceptor atom, positively doping Bi$_2$Se$_3$ which is normally found to be n-doped due to Se vacancies. The samples are cleaved in vacuum at pressures lower than 7·10$^{-11}$ mbar, and kept at a temperature of 15 K during evaporation and measurements. 2DEGs are induced by evaporating K (Fig. 2) or Li (Fig. 3) in situ on the cleaved sample surface. The TR-ARPES experiments are performed at QM’s UBC-Moore Center for Ultrafast Quantum Matter$^{42,43}$, with 1.55 and 6.2 eV photons for pump and probe, respectively. Both pump and probe have linear horizontal polarization (parallel to the analyzer slit direction). The pump (probe) beam radius is 150 μm (100 μm), and the pump fluence is 40 and 80 μJ/cm$^2$ for experiments represented in Figs. 2 and 3, respectively. Pump and probe were collinear with an incidence angle of 45 degrees with respect to the sample normal. Energy and temporal resolution are 17 meV and 250 fs, respectively, as determined by the width of the gold Fermi edge and of the combined pump-probe dynamics of the pump induced direct population peak in Bi$_2$Se$_3$. For the band bending model, the Poisson equation was solved numerically employing a modified Thomas-Fermi approximation, which intrinsically accounts for modulation of the charge density due to confinement-induced quantization, without the need for numerically heavy self-consistent calculations$^{40,44}$. The Schrödinger equation was solved numerically with the Numerov algorithm$^{45}$. The code is available at Ref. 46.

Data availability

The authors declare that the main data supporting the findings of this study are available within the paper and its Supplementary Information files. The raw ARPES data generated in this study have been deposited in the Zenodo database under the digital object identifier https://doi.org/10.5281/zenodo.6471678. Extra data are available from the corresponding authors upon request.

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Author contributions

M.M., F.B., H.-H.K., M.-X.N. performed the measurements and data analysis, S.K.Y.D. contributed to the measurements, A.C. contributed to data analysis, G.L., S.Z., A.K.M., D.J.J. provided technical support and instrumentation, J.L.M., B.B.I., and P.H. provided the samples, P.H. and A.D. were responsible for the overall direction, planning, and management of the project. All authors contributed to the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

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