

Sign of the Rashba parameter in image-potential states

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Image-potential surface states are simple model systems, where spin-dependent effects can be studied in view of spintronic applications. The Rashba effect in surface states at high- Z materials is related to both the strong spin-orbit interaction in heavy atoms and the orbital angular momentum arising from the inversion-symmetry breaking at the surface. Our study on image-potential states at Bi_2Se_3 and Bi_2Te_3 showcases that the orbital angular momentum causes an intrinsically reversed Rashba parameter $\alpha_R \approx -100 \text{ meV \AA}$, i.e., a reversed spin splitting compared with the prototypical Rashba-split $\text{Au}(111)$ crystal-induced surface state. We present a consistent picture of spin-resolved experimental data from inverse photoemission and three-photon photoemission together with calculations from density-functional theory and many-body perturbation theory.

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I. INTRODUCTION

For spintronic devices, spin manipulation without external magnetic fields is highly desirable. At surfaces and two-dimensional materials, the spin degeneracy of electronic states can be lifted by the Rashba effect [1]. This phenomenon, originally discussed for a free electron gas under the influence of an electric field, requires in real materials two ingredients: spin-orbit interaction (SOI), mostly supplied by high- Z atoms, and nonzero orbital angular momentum (OAM) due to the inversion symmetry breaking at surfaces [2–5]. An empirical measure of this SOI-OAM effect in free-electron-like states with effective mass m^* is given by the Rashba parameter α_R in the dispersion relation

$$E_{\pm}(\mathbf{k}_{\parallel}) = \hbar^2 \mathbf{k}_{\parallel}^2 / 2m^* \pm \alpha_R |\mathbf{k}_{\parallel}|. \quad (1)$$

The sign of α_R is decisive for controlling the absolute spin directions in view of spintronic devices. For surface states with parabolic dispersion and m^* in the order of the free electron mass, the Rashba parameter has so far been found positive ($\alpha_R > 0$), e.g., for the prototypical crystal-induced Shockley surface state at $\text{Au}(111)$ with $\alpha_R = +330 \text{ meV \AA}$ [6–9].

Another class of surface states are image-potential induced states (IS), caused by the long-range Coulomb-like surface barrier potential, which results from electronic screening effects [10–12]. They form a Rydberg series of free-electron-like states pinned to the vacuum level (within $\approx 1 \text{ eV}$ for conductive surfaces). Spatially the states are very extended into the vacuum. A distinct overlap with surface atoms, however, can induce at high- Z materials a SOI-induced Rashba-type spin splitting. This was discussed in theoretical [13,14] as well as experimental studies [15–17]. The reported values for the Rashba parameter of IS are in the range of $+10$ to $+100 \text{ meV \AA}$, significantly smaller than for the $\text{Au}(111)$ Shockley state.

In all aforesaid cases, the Rashba effect is dominated by SOI and the spin texture is strictly the same: The electron spin is pinned within the surface plane and perpendicular to $\mathbf{k}_{\parallel}$. With $\alpha_R > 0$ the spin-down branch is always at higher energy compared to the spin-up branch for positive $\mathbf{k}_{\parallel}$. The $\mathbf{k}_{\parallel}$ vector, the spin-quantization axis, and the surface normal form a right-handed coordinate system. This observation is consistent with corresponding model calculations. However, a significant Rashba effect was also found in cases with strong OAM and only minor influence of SOI [2–5]. In addition, a negative Rashba parameter was reported for quantum-well states in $\text{Pb/Si}(111)$ [18]. It was ascribed to competing phase-shift effects at the two interfaces of the Pb film induced by details of the electronic and structural properties. A second example of a negative Rashba parameter was found in the surface alloy $\text{Bi/Cu}(111)$ [19]. This was explained “by the precise charge distribution asymmetry of a surface state close to the atomic nuclei”. Recently, it was calculated and discussed by

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Ishida [20], that the OAM influences the Rashba effect in IS differently for W(110) and Ta(110). In these calculations, the sequence of the spin branches differs, i.e., the sign of α_R is positive for W(110) and negative for Ta(110). Unfortunately, for the interesting case on Ta(110), experimental IPE studies were not able to detect the IS due to overlap with bulk states [21].

In this work, we prove the relation of OAM and sign of α_R within a single class of surface states, namely free-electron-like IS. We obtained a negative α_R for IS at the examples of Bi_2Se_3 and Bi_2Te_3 in contrast to recent data of IS at $\text{Re}(0001)$ [17] or the Shockley state at $\text{Au}(111)$ both with a positive α_R . We present spin-resolved measurements with inverse photoemission (IPE) and multiphoton photoemission. The results are explained along state-of-the-art density-functional theory (DFT) and many-body perturbation theory calculations.

II. METHODS

The spin- and angle-resolved IPE experiments on Bi_2Se_3 were performed in Münster [22,23]. Spin-polarized electrons ($P = 29\%$) used for excitation were extracted from a GaAs photocathode. The transversal spin polarization of the electron beam was oriented within the sample surface plane and perpendicular to $\mathbf{k}_\parallel$, i.e., sensitive to the Rashba component. All data have been normalized to 100% electron spin polarization [24]. The electron beam impinged on the sample with defined energy [see left part of Fig. 1(a)] and angle θ relative to the surface normal with a FWHM divergence $< 3^\circ$ [23,25]. The emitted photons were detected with a band-pass detector at $\hbar\omega = 9.9$ eV. The overall FWHM energy resolution of the IPE experiment was ≈ 400 meV [26]. The IPE measurements were carried out at room temperature.

The spin- and angle-resolved three-photon photoemission (3PPE) experiments on Bi_2Se_3 and Bi_2Te_3 were performed in Tokyo using 2.38 eV photons. The work functions of Bi_2Se_3 and Bi_2Te_3 are 5.44 eV and 5.33 eV, respectively. Consequently, the states can be populated via two-photon absorption of the 2.38 eV light. A subsequent excitation with the same photon energy enables their probing, thereby allowing photoemission spectroscopy of the IS [see right part of Fig. 1(a)]. The light source was an Yb-doped fiber laser, and the second harmonic (2.38 eV) of its fundamental (1.19 eV), generated in a BBO crystal, was used for the 3PPE process (pulse duration time 300 fs, fluence of the pulse laser 0.5 mJ/cm^2 , p polarization). The FWHM energy resolution was about 40 meV and the FWHM angular resolution was 2.4° . The spin-resolved measurements were carried out using a hemispherical analyzer combined with a VLEED spin detector, the spectra have been normalized to the respective Sherman function (for details see Refs. [27,28]). All 3PPE measurements were carried out at a sample temperature of 35 K.

We used an n -type Bi_2Se_3 sample already described elsewhere [29,30] and an n -type Bi_2Te_3 crystal fabricated by using the Bridgman method. The samples were prepared by exfoliation at ultrahigh vacuum conditions. All IPE and 3PPE measurements were performed along the $\bar{\Gamma}\bar{M}$ direction.

The electronic structure has been calculated within the local-density approximation (LDA) [31] of DFT. A basis of

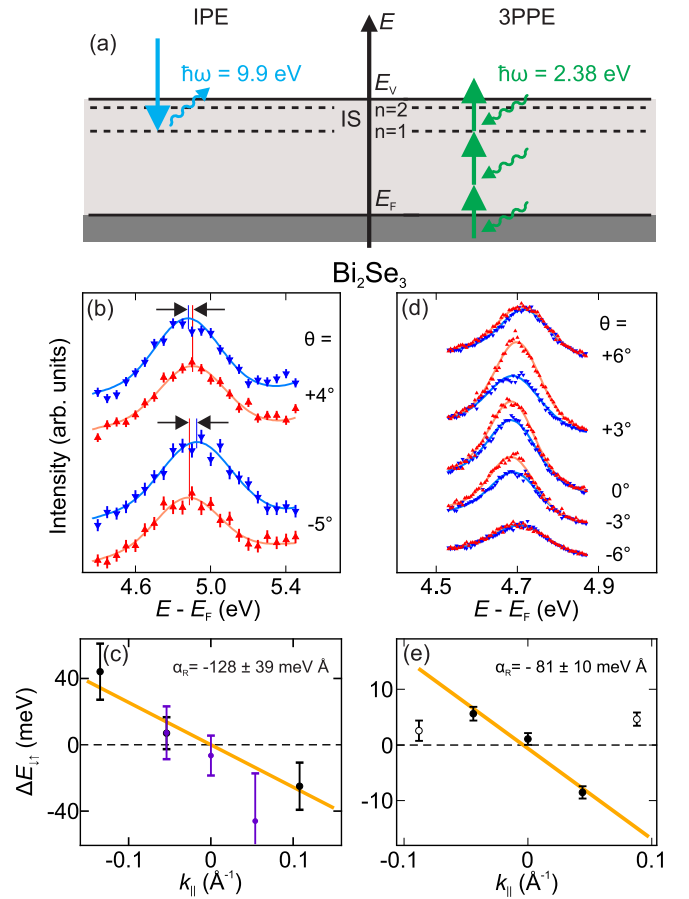


FIG. 1. Experimental sketches and data of the $n = 1$ IS at Bi_2Se_3 . (a) Energy sketches of the IPE and 3PPE processes. (b) Spin- and angle-resolved IPE spectra taken from dataset 1. (c) Energy splitting $\Delta E_{\downarrow\uparrow}(k_\parallel)$ from IPE with linear fit (orange line) resulting in $\alpha_R = -128 \pm 39 \text{ meV \AA}$ (dataset 1: black dots, dataset 2: purple dots). (d) Spin- and angle-resolved 3PPE spectra. (e) $\Delta E_{\downarrow\uparrow}(k_\parallel)$ from 3PPE with linear fit (orange line) resulting in $\alpha_R = -81 \pm 10 \text{ meV \AA}$. (Please note the different scales of the energy and momentum axes.)

atom-centered Gaussian orbitals is employed together with pseudopotentials that include scalar relativistic corrections and spin-orbit coupling [32,33]. To represent the wave functions in the vacuum, Gaussian orbitals located at the virtual atomic positions of up to three quintuple layers above one surface are incorporated. The surfaces of Bi_2Se_3 and Bi_2Te_3 are treated within a supercell approach using slabs with six quintuple layers (QL). Details for the calculation of the $\text{Re}(0001)$ surface are given in Ref. [34].

Since the LDA does not lead to an image-potential-like surface barrier, we replace the LDA potential in the vacuum region by a potential containing the correct $1/z$ asymptotic tail (see Appendix A). Alternatively, we have employed for Bi_2Se_3 and Bi_2Te_3 a second set of calculations using *ab initio* many-body perturbation theory. The exchange-correlation effects are described by the electron self-energy operator and the corresponding electron-hole interaction. This is evaluated within the GW approximation [35,36], which allows for a more reliable description natively including image states [37], and has the correct $1/z$ asymptotic by construction. We find

good agreement of both approaches for each of the two materials (details for the case of Bi_2Te_3 , e.g., are given in the Appendix B).

III. RESULTS

Figure 1 presents experimental results of the previously reported $n = 1$ IS at Bi_2Se_3 [38–42] obtained with both, spin- and angle-resolved IPE [Figs. 1(b) and 1(c)] and 3PPE [Figs. 1(d) and 1(e)]. Figure 1(b) displays a closeup of two IPE spectra for almost opposite electron incidence angles θ . Red and blue triangles denote spin-up and spin-down components, respectively. Note that the spectra are shifted along the intensity axis for clarity. The fine solid curves represent results from a fitting procedure (for details see Supplemental Material [43]). Black arrows and fine vertical lines highlight the small, but significant spin splittings $\Delta E_{\downarrow\uparrow}$. The spin sequence appears reversed compared to the Shockley surface state at Au(111) [44] and also to the IS at $\text{Re}(0001)$ [17] measured in the same apparatus under identical experimental conditions.

Figure 1(c) summarizes the results from 1(b) (dataset 1: black dots) and additional measurements (dataset 2: purple dots) in a $\Delta E_{\downarrow\uparrow}(k_{\parallel})$ plot. The two datasets originate from different sample exfoliations to check the reproducibility of the results. For each exfoliation, the IS appeared slightly shifted in energy, mostly due to different doping at the surface and/or slight work function changes. While a lower signal-to-background ratio in dataset 2 (purple) results in larger uncertainty bars, the energy splittings match the ones from dataset 1. A linear fit to both datasets (orange line) yields a Rashba parameter of $\alpha_R = -128 \pm 39 \text{ meV \AA}$. A simplified fitting approach to approximate the peak maxima by parabolic functions results in $\alpha_R = -100 \pm 33 \text{ meV \AA}$. The peak positions obtained by the two fitting procedures agree within their uncertainty intervals.

To confirm our IPE results independently, we carried out additional spin- and angle-resolved 3PPE measurements on the same sample. Figures 1(d) and 1(e) present the corresponding 3PPE results. Surprisingly, the spectral intensity and spin polarization of the IS changes in the spectra for $+6^\circ$ and -6° . We interpret this as a result of the excitation process with intermediate states in 3PPE being influenced by bulk bands at this momentum (see Fig. 6 left) and/or the overlap of bulk bands and IS at this particular sample preparation. Therefore, we evaluate spectra with $|\theta| < 6^\circ$ only. Fits to the spectra consist of one Voigt peak and a Shirley-type background. For the Voigt function the Gaussian-to-Lorentz ratio was optimized and fixed. The obtained Lorentzian width is about the same as the apparatus function and is compatible with reported lifetime broadening in image-potential states [45]. The Shirley background shows negligible spin polarization. The resulting peak positions from Fig. 1(d) are displayed as $\Delta E_{\downarrow\uparrow}(k_{\parallel})$ plot in Fig. 1(e). The linear fit (orange line) includes only the filled dots and results in $\alpha_R = -81 \pm 10 \text{ meV \AA}$.

The most remarkable result of our study on the Bi_2Se_3 IS with different spin-resolved experimental methods and at several sample preparations is a Rashba parameter with reversed sign compared with the Au(111) Shockley state [44]. Concerning the size of the Rashba parameter, different sample preparations, different experimental techniques, and different

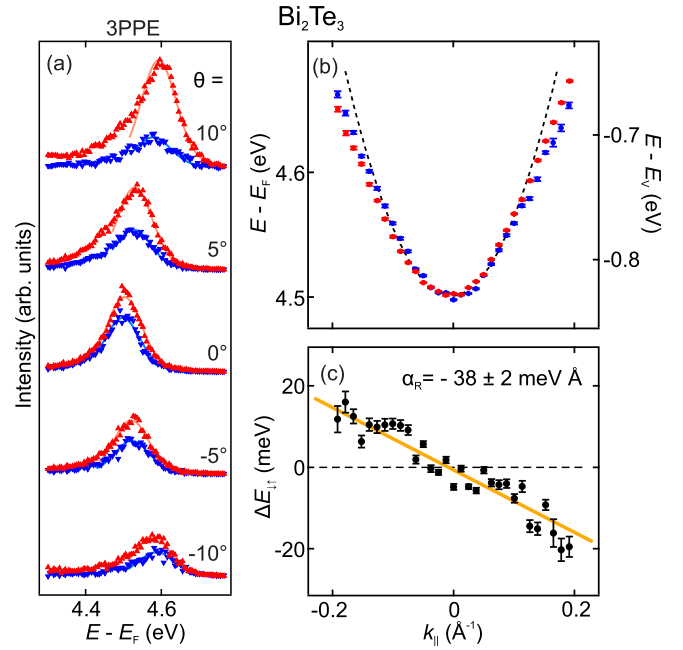


FIG. 2. (a) Selection of spin- and angle-resolved 3PPE spectra of the $n = 1$ IS at Bi_2Te_3 (see Fig. S1 in Supplemental Material for full series [43]). (b) Spin-dependent energy vs $k_{\parallel}$ dispersion of the IS. Black dashed curve: parabolic dispersion fitted to $|k_{\parallel}| \leq 0.1 \text{ \AA}^{-1}$. (c) $\Delta E_{\downarrow\uparrow}(k_{\parallel})$ with linear fit (orange line) resulting in $\alpha_R = -38 \pm 2 \text{ meV \AA}$.

data analysis procedures yield results compatible within the uncertainty intervals. The given uncertainty values are mostly due to the statistical uncertainties of the fitting procedures. In addition, each new sample exfoliation may have altered the surface roughness and intrinsic doping at the surface slightly [30,46]. This causes small differences in the work function and/or position of the Fermi level. The consequence is a small energy shift of the IS relative to the bulk bands and therefore slightly deviating Rashba parameters can be expected. Nevertheless, the negative sign of α_R at Bi_2Se_3 is consistently found with at least three σ significance.

We expanded our study to the sister material Bi_2Te_3 . A selection of spin- and angle-resolved 3PPE spectra of the $n = 1$ IS at Bi_2Te_3 is shown in Fig. 2(a) (see Fig. S1 in the Supplemental Material for full series [43]). The high signal-to-background ratio is particularly advantageous for a precise analysis. The overall stronger spin-up signal is ascribed to the excitation process [47]. An energy-dependent spin splitting is clearly visible, especially for $\theta = \pm 10^\circ$. Peak energies were determined by a fitting procedure shown by the solid lines. The analyzed part of the spectrum is approximated by a Lorentzian (FWHM fixed to 20 meV) added to a constant background. This sum was convoluted with a Gaussian function to account for experimental broadenings. The resulting spin-up and spin-down peak positions from the full spectra series ($-15^\circ \leq \theta \leq +15^\circ$ in 1° steps) are shown in Fig. 2(b). The dashed black parabola is a fit to the spin-integrated energy positions for $|k_{\parallel}| \leq 0.1 \text{ \AA}^{-1}$. The IS dispersion deviates from a free-electron-like parabola for higher $k_{\parallel}$. This indicates the influence from bulk states to the IS. Finally, Fig. 2(c) displays the energy splitting between the two spin components of the

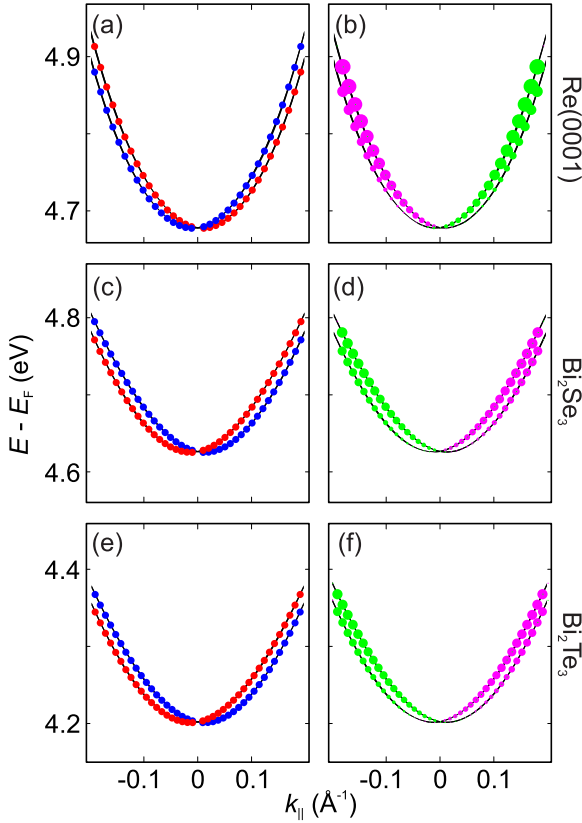


FIG. 3. Results of LDA-DFT calculations for the IS at [(a), (b)] Re(0001), [(c), (d)] Bi₂Se₃, and [(e), (f)] Bi₂Te₃. In [(a), (c), (e)] the red (blue) dots symbolize the spin up (down) expectation value. In [(b), (d), (f)] the green (magenta) dots are proportional to the clockwise (counterclockwise) OAM. The dot size in (d) and (f) is scaled by a factor 1.6 for better visibility.

IS. A linear fit (orange line) over the full $k_{\parallel}$ range yields again a negative Rashba parameter of $\alpha_R = -38 \pm 2$ meV Å. We note that alternative approaches for the fit functions were evaluated and yield α_R values, which are compatible with the uncertainty interval given above (see Fig. S2 in the Supplemental Material [43]).

To explain the negative sign of the Rashba parameter, we calculated the IS electronic structure of Bi₂Se₃ and Bi₂Te₃. The results are compared to calculations of the IS with a positive Rashba parameter at Re(0001). Figure 3 displays the theoretical results for Re(0001) in Figs. 3(a) and 3(b), Bi₂Se₃ in Figs. 3(c) and 3(d), and Bi₂Te₃ in Figs. 3(e) and 3(f). The red (blue) dots in Figs. 3(a), 3(c), and 3(e) are proportional to the expectation value of the up (down) Rashba spin polarization. The green (magenta) dots in Figs. 3(b), 3(d), and 3(f) are proportional to the clockwise (counterclockwise) OAM.

The calculations for the spin polarization confirm the experimental results of a sign reversal of α_R between Re(0001) on the one hand, and Bi₂Se₃ and Bi₂Te₃ on the other hand. The spin structure with a positive $\alpha_R = +99$ meV Å at Re(0001) in Fig. 3(a) is consistent with earlier calculations and experimental results [17]. Also, the Rashba parameter of the Re(0001) IS has the same sign as α_R of the prototypical Rashba-split Au(111) Shockley surface state. The corresponding spin structures for Bi₂Se₃ and Bi₂Te₃ in Figs. 3(c) and 3(e)

are reversed and yield negative values $\alpha_R = -128$ meV Å and $\alpha_R = -120$ meV Å, respectively. Additional GW calculations yield Rashba parameters of $\alpha_R = -115$ meV Å (Bi₂Se₃) and $\alpha_R = -110$ meV Å (Bi₂Te₃). The quantitative differences to the experimental results can be expected for this type of sample. While theory models use perfect Bi₂Se₃ (Bi₂Te₃), the corresponding experimental samples exhibit varying doping levels and surface structures for each new exfoliation (see above). Yet the qualitative result of a negative α_R is consistently reproduced.

The OAM calculations in Figs. 3(b), 3(d), and 3(f) show an apparent correlation between spin structure and OAM. While the OAM structure is similar for Bi₂Se₃ and Bi₂Te₃, the overall sign is reversed compared to Re(0001). A detailed analysis for the orbital contributions to the IS wave function reveal manifold orbital influences to the OAM. We cannot identify a distinct orbital contribution being responsible for the OAM change in IS. Nevertheless, we interpret the reversal of the OAM structure as the origin of the sign change in the Rashba parameter.

Surprisingly, the calculations give similar α_R values for both, Bi₂Se₃ and Bi₂Te₃, even tending to higher values for Bi₂Se₃. This result is counterintuitive in view of the lower Z of Se. We investigated the spatial distribution of the IS by calculations of the charge density. In Figs. 4(a) and 4(b) filled (open) green markers show the positions of Bi atoms within (outside) the drawing plane, the same applies for black markers symbolizing Se/Te atoms. Typical for free-electron-like IS, the charge is mostly delocalized outside the sample surface. Importantly, the maxima of the distribution appear close to the topmost Se/Te atoms, which form the surface, rather than at the much heavier Bi atoms. Additional calculations with artificially separated influence of SOI by either Bi or the chalcogen atoms reveal about equal contributions. Figure 4(c) shows a comparison of the charge densities along the red dashed lines in Figs. 4(a) and 4(b) for Bi₂Se₃ (cyan) and Bi₂Te₃ (brown), respectively. The yellow area highlights the essential z range at the topmost atom. In this range with strongest SOI, the IS charge density at Bi₂Se₃ extends the IS charge density at Bi₂Te₃. This overcompensates the lower Z value of Se and, therefore, a higher α_R for the IS at Bi₂Se₃ can be justified.

Comparing the two charge density maxima around the topmost atom ($-3 \text{ Å} \leq z \leq 1.5 \text{ Å}$) in Fig. 4(c), more charge is located at the sample side than the vacuum side. This spatial asymmetry was also discussed by Bentmann *et al.* [19] for a surface alloy state in Bi/Cu(111). The extended charge distribution of IS at Bi₂Se₃ and Bi₂Te₃ is in contrast to the surface alloy state at Bi/Cu(111), which is localized within $\approx 1 \text{ Å}$ around the Bi atoms. In both cases, however, a charge localization towards the sample side is observed close to the topmost atoms. As in our calculations similar distributions can be found for other surface states with positive Rashba parameter, we are not able to assign this as an exclusive indicator.

IV. CONCLUSION

We presented multiple proof for a negative sign of α_R in the intrinsic spin structure of Rashba-split free-electron-like

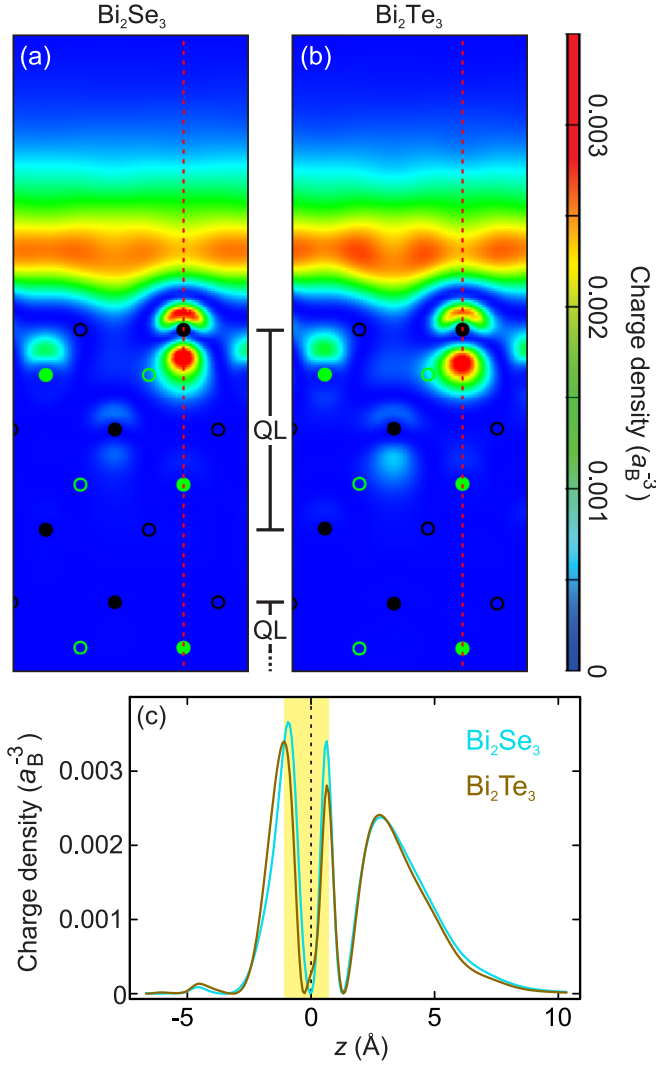


FIG. 4. Charge density distributions of the IS close to $\bar{\Gamma}$. (a), (b) Filled (open) green markers show the positions of Bi atoms within (outside) the drawing plane. The same is valid for black markers symbolizing Se/Te atoms. The charge density for Bi₂Te₃ in (b), (c) is scaled by the volume of the unit cells $V_{\text{Bi}_2\text{Te}_3}/V_{\text{Bi}_2\text{Se}_3}$. (c) Comparison of the charge densities along the dashed red lines in (a), (b). The z axis of Bi₂Te₃ is scaled by the lattice constants $a_{\text{Bi}_2\text{Te}_3}/a_{\text{Bi}_2\text{Se}_3}$. The dashed black line marks the position of the topmost Se/Te atom.

states, namely image-potential states at Bi₂Se₃ and Bi₂Te₃. Two different spin- and angle-resolved experimental approaches yield a negative Rashba parameter α_R : IPE at Bi₂Se₃, 3PPE at Bi₂Te₃, and also 3PPE at Bi₂Se₃ as a cross-check experiment. Theoretical calculations confirm the experimental results and relate the negative sign of α_R to a sign change in the OAM of the IS wave function. Controlling the sign of the Rashba parameter opens the way of spin manipulation without external fields for spintronic devices. Since this principle is very general, more materials and states with reversed sign of the Rashba parameter are expected to be found in the future.

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DATA AVAILABILITY

The data that support the findings of this article are available from the authors upon reasonable request.

APPENDIX A: MODELING THE SURFACE BARRIER

To incorporate the existence of image states into our DFT calculation, we replace the self-consistently calculated DFT potential $V_{\text{LDA}}(\mathbf{r})$ for $z > 0$ by

$$V(\mathbf{r}) = V_{\text{LDA}}(\mathbf{r})W(z, z_0) + V_M(z, z_0)[1 - W(z, z_0)], \quad (\text{A1})$$

with the model potential (in Rydberg energies)

$$V_M(z, z_0) = \left(\Phi - \frac{1}{2} \frac{1}{z - z_0} \right) (1 - e^{\lambda(z_0 - z)}). \quad (\text{A2})$$

Φ is the work function resulting from the DFT calculation (5.56 eV for Bi₂Se₃ and 5.14 eV for Bi₂Te₃). $W(z, z_0)$ is a weight function that rapidly goes to 1 for $z < z_0$ and goes to 0 for $z > z_0$. For a given value of λ , z_0 is determined by the requirement that $V_M(z, z_0)$ touches the averaged DFT potential

$$\bar{V}_{\text{DFT}}(z) = \frac{1}{\Omega} \int_{\Omega} V_{\text{DFT}}(\mathbf{r}) d^2\mathbf{r}_{\parallel}. \quad (\text{A3})$$

Employing V_M to calculate the image states of several materials, we find that $\lambda = 1/a_B$ describes the energy of the band minima in fair agreement with the respective measured positions. We use the same λ value in the actual calculations for Re(0001), Bi₂Te₃, and Bi₂Se₃.

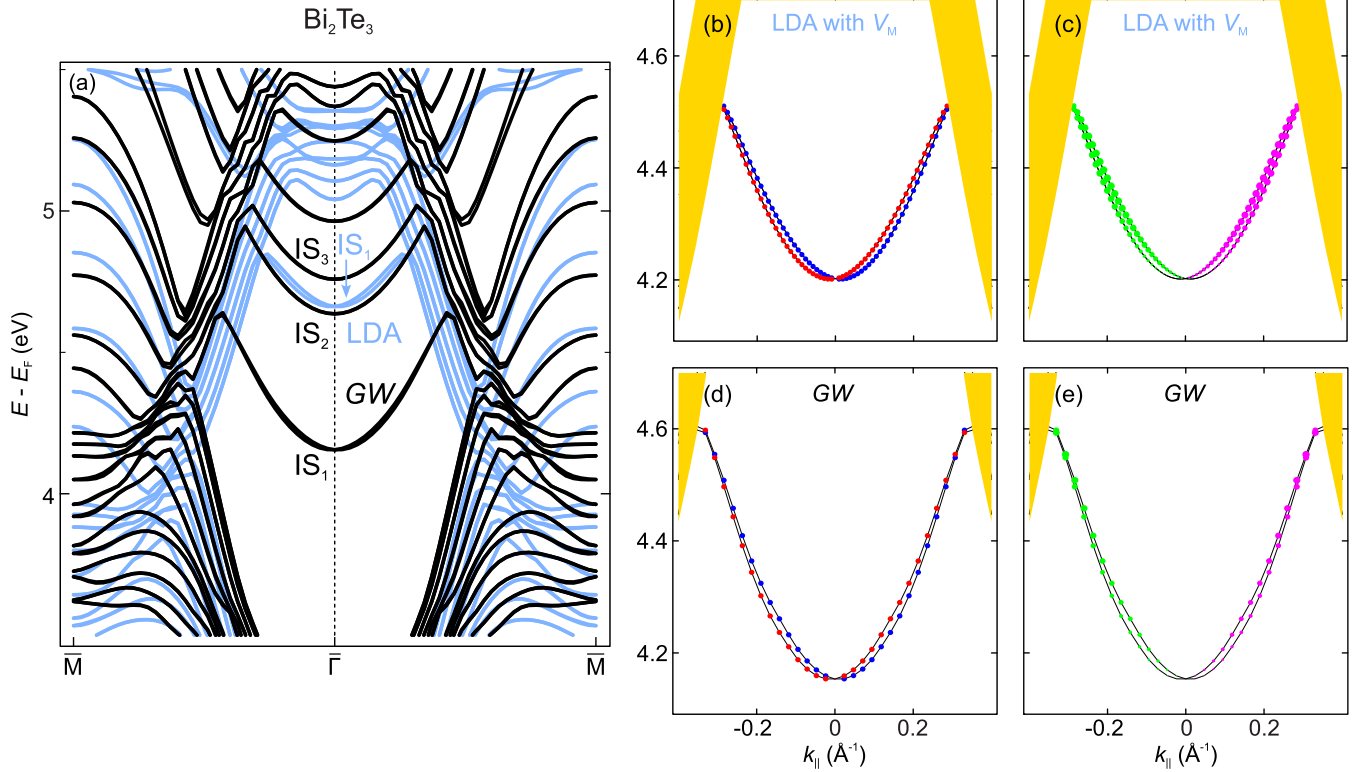


FIG. 5. (a) Band structure of six QL of Bi₂Te₃ calculated with LDA (no V_M , light blue) and GW (black). (b) and (c) show a zoom-in for LDA with V_M (see main text Fig. 3), and compare well to GW shown in (d) and (e).

APPENDIX B: BAND STRUCTURES

Our many-body perturbation theory calculations with the GW approximation have been performed similar to previous studies [36]. Only the number of evaluated bands is increased to 336 due to the large energy (with respect to E_F) of the image potential states. In Fig. 5(a) an unoccupied part of the band structure for Bi₂Te₃ is shown, which compares the DFT(LDA) results (light blue) to GW (black). As expected, most GW-derived bands are shifted to higher energies in comparison to the DFT bands. In contrast, the image state is clearly lowered in energy by about 0.6 eV. Thus, it exists as a localized state about half the $\bar{\Gamma}$ - $\bar{M}$ path. In Figs. 5(b) and 5(c) the results of the spin polarization and the orbital angular momentum are replotted from the main text (see Fig. 3 for further discussion). The model potential with $\lambda = 1/a_B$ leads to a similar downward shift of the image state as observed in GW. Also the spin polarization and orbital angular momentum compare well to the GW result in Figs. 5(d) and 5(e), and the same holds for the Rashba parameter (see main text). We note in passing, that a similarly good agreement of LDA employing V_M and GW is observed for Bi₂Se₃.

Figure 6 shows the band structures (DFT + V_M) of Bi₂Se₃ and Bi₂Te₃ on a larger energy scale showing the $n = 1$ and $n = 2$ image states, as well as the topological surface states close to the Fermi level.

APPENDIX C: ORBITAL ANGULAR MOMENTUM

In the context of SOI, the angular momentum operator is multiplied with the gradient of the atomic potential, which is a spatially very localized quantity, i.e., SOI mainly acts in the vicinity of an atom. Therefore, we have calculated the OAM by treating the angular momentum operator as a local object placed at the atom positions. To this end, we evaluate the matrix elements of the angular momentum operator parallel to the surface (in x direction for $\mathbf{k}$ in y direction) as

$$\langle \text{OAM} \rangle^{\mathbf{k}} = \sum_{\alpha, \alpha', \mu} c_{\alpha\mu}^{n\mathbf{k}*} \langle \hat{L}_x \rangle_{\alpha\mu, \alpha'\mu} c_{\alpha'\mu}^{n\mathbf{k}} \quad (\text{C1})$$

$$\langle \hat{L}_x \rangle_{\alpha\mu, \alpha'\mu} = \int_{\mathbb{R}^3} d^3r \varphi_{\alpha\mu}^*(\mathbf{r} - \boldsymbol{\tau}_\mu) \times \{(\mathbf{r} - \boldsymbol{\tau}_\mu) \times \hat{\mathbf{p}}\}_x \varphi_{\alpha'\mu}(\mathbf{r} - \boldsymbol{\tau}_\mu) \quad (\text{C2})$$

with the local angular momentum operator $\hat{\mathbf{L}}$ placed at atom $\boldsymbol{\tau}_\mu$. Here $\boldsymbol{\tau}_\mu$ denotes the position of the atoms, α indicates the character of our Gaussian basis function ($spds^*$), and $c^{n\mathbf{k}}$ is the coefficient of the Bloch basis function.

Note that μ is restricted to the atoms at the corresponding surface. The lattice sums $\sum_{\mathbf{R}_{j'}} e^{i\mathbf{k}(\boldsymbol{\tau}_{\mu'} + \mathbf{R}_{j'})}$, etc. are neglected including all nonlocal effects.

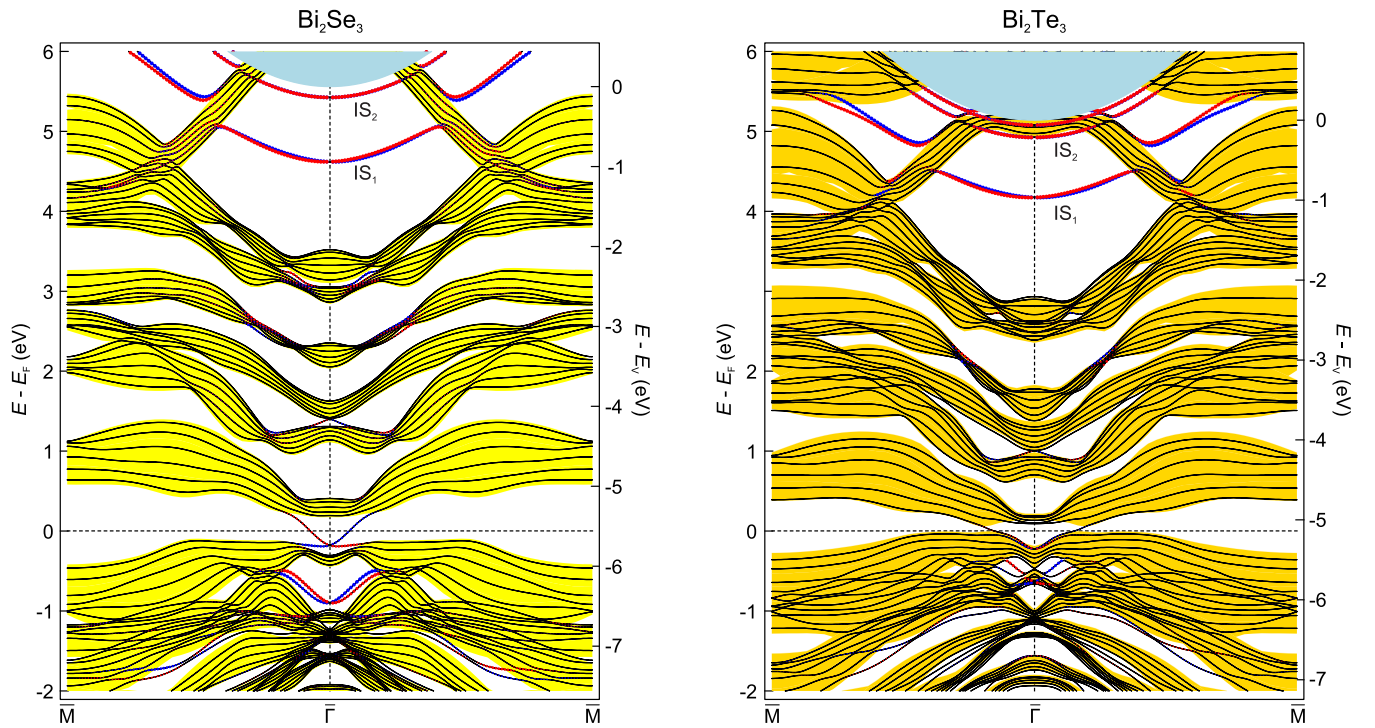


FIG. 6. Band structure of six quintuple layers of Bi_2Se_3 (left) and Bi_2Te_3 (right) along $\bar{\Gamma}\bar{M}\bar{\Gamma}$ from a DFT calculation including the model potential V_M . IS_1 and IS_2 denote the image states with $n = 1$ and $n = 2$, respectively. The light-blue area represents the region of the vacuum continuum states and the yellow area indicates regions of backfolded bulk bands. The spin-up (spin-down) character of the bands is indicated by red (blue) dots. In (a) and (b) the respective energy axis on the left side refers to the Fermi energy while the axis on the right side refers to the vacuum level.

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