

<https://doi.org/10.1038/s41699-025-00562-4>

High-throughput calculations of spin Hall conductivity in non-magnetic 2D materials

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Spin Hall effect (SHE) in two-dimensional (2D) materials is promising to effectively manipulate spin angular momentum and identify topological properties. In this work, we implemented an automated Wannierization with spin-orbit coupling on 426 non-magnetic monolayers including 210 metal and 216 insulators. Intrinsic spin Hall conductivity (SHC) has been calculated to find candidates exhibiting novel properties. We discover that $\text{Y}_2\text{C}_2\text{I}_2$ has an unconventional SHE with canted spin due to low crystal symmetry, Ta_4Se_2 is a metallic monolayer with exceptionally high SHC, and the semi-metal Y_2Br_2 possesses efficient charge-to-spin conversion induced by anti-crossing in bands. Moreover, quantum spin Hall insulators are investigated for quantized SHC. The present work provides a high-quality Wannier Hamiltonian database of 2D materials, and paves the way for the integration of 2D materials into high-performance and low-power-consumption spintronic devices.

Spin Hall effect (SHE) is a phenomenon emerging from spin-orbit coupling (SOC) where an external electric field can induce a transverse spin current¹. As an effective method to control spin transport, the SHE has been extensively investigated in bulk heavy metals². However, simple heavy metals are constrained by symmetry and only present conventional SHE where spin current, electric field, and spin orientation are orthogonal to each other. Constrained by this strict rule, an external magnetic field is required to switch perpendicular magnetization³, hindering device minimization. Besides, defined as the ratio of spin Hall conductivity (SHC) to charge conductivity, the spin Hall ratio (SHR) is reduced by high charge conductivity in heavy metals, leading to a low charge-to-spin conversion efficiency². Van der Waals materials emerge as promising candidates to overcome these challenges. SHCs have been studied in two-dimensional (2D) transition-metal dichalcogenide (TMDC) materials^{4–11}. Compared with high-symmetry bulk heavy metals, 2D materials with low-symmetry structures can induce unconventional SHEs^{12,13}. In addition, the reduced charge conductivity in 2D materials enables efficient charge-to-spin conversion^{14,15}. More importantly, the reduced dimension induces quantum spin Hall effect in topological 2D materials¹⁶, offering robust and quantized SHCs in bandgaps of quantum spin Hall insulators (QSHIs)^{17–19}. However, current researches on SHE of 2D materials are mostly limited to TMDC materials, while a large-scale study on 2D materials for spintronics is still absent.

Wannier functions^{20,21} are extensively used to study the SHC²². Recent works have developed frameworks for the high-throughput Wannierization. Symmetrized Wannier Hamiltonians have been

generated for the strained III-V semiconductor materials by applying post-processing symmetrization²³. In addition, the single-criterion decision-making algorithm with no need for an initial guess has been validated for band interpolations on a dataset of 200 materials²⁴. Moreover, Wannier Hamiltonians including SOC for 1406 3D and 365 2D materials were generated using hydrogenic orbital projections with hints from pseudopotentials²⁵. Recently, the projectability-disentangled Wannier functions have been proposed where the initial guess is given by a projectability measure for each Bloch state onto atomic orbitals^{26,27}.

In this work, we perform a high-throughput study of spin Hall conductivity in non-magnetic 2D materials using density functional theory (DFT) and Wannier functions. Leveraging the scalar-relativistic MC2D database^{28,29}, all the rare-earth-free monolayers with up to 6 atoms per unit cell are considered, yielding 426 exfoliable materials. These materials are investigated with *ab initio* calculations including SOC. Intrinsic SHCs are calculated in all the materials to reveal exotic SHE. Unconventional SHE is found in a low-symmetry metal, materials with high SHC values are listed, and a high SHR is found in a semi-metal. Finally, SHCs of topological insulators are studied to understand the importance of spin conservation for quantized SHC.

Results

To match the typical experimental setup, we focus on the SHC where the spin current is along x , the electric field is along y , and the spin orientation is along α direction. The intrinsic SHC in a weak scattering limit can be

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calculated by the Kubo formula^{22,30}:

$$\sigma_{xy}^{\alpha} = \frac{\hbar}{2e} \frac{e^2}{\hbar} \int_{\text{BZ}} \frac{d^2\mathbf{k}}{(2\pi)^2} \Omega_{xy}^{\alpha}(\mathbf{k}), \quad (1)$$

where BZ denotes the Brillouin zone, and the spin Berry curvature (SBC) is given as

$$\Omega_{xy}^{\alpha}(\mathbf{k}) = \sum_n f_{nk} \Omega_{n,xy}^{\alpha}(\mathbf{k}), \quad (2)$$

$$\Omega_{n,xy}^{\alpha}(\mathbf{k}) = \hbar^2 \sum_{m \neq n} \frac{-2 \text{Im} \{ [nk | \hat{j}_{\alpha} | mk] \langle mk | \hat{v}_y | nk \rangle \}}{(\epsilon_{nk} - \epsilon_{mk})^2 + \eta^2}. \quad (3)$$

In the above equations, f_{nk} denotes the Fermi-Dirac distribution $f_{nk}(\mu, T) = \frac{1}{e^{(\epsilon_{nk} - \mu)/(k_B T)} + 1}$, where μ is the Fermi energy. $\hat{j}_{\alpha} = \frac{1}{2} \{ \hat{\sigma}_{\alpha} \hat{v}_x + \hat{v}_x \hat{\sigma}_{\alpha} \}$ is the spin current operator, $\hat{\sigma}_{\alpha}$ is the Pauli operator, $\hat{v}_x$ and $\hat{v}_y$ are velocity operators, η is the broadening approximated as extrinsic scattering³¹.

To compute the SHC on ultra-dense $\mathbf{k}$ grids, we construct an ab initio Wannier Hamiltonian database and assess its quality by comparing the average differences between DFT bands and Wannier bands, given as^{26,27}

$$\eta_{\nu} = \sqrt{\frac{\sum_{nk} \tilde{f}_{nk} (\epsilon_{nk}^{\text{DFT}} - \epsilon_{nk}^{\text{Wan}})^2}{\sum_{nk} \tilde{f}_{nk}}} \quad (4)$$

where $\mathbf{k}$ denotes eigenstates along the high-symmetry $\mathbf{k}$ -path of the band structure, $\tilde{f}_{nk} = \sqrt{f_{nk}^{\text{DFT}}(E_F + \nu, T) f_{nk}^{\text{Wan}}(E_F + \nu, T)}$ where $E_F + \nu$ represents energy position, thus η_{ν} denotes the difference given by comparing $\epsilon_{nk}^{\text{DFT}}$ and $\epsilon_{nk}^{\text{Wan}}$ below $E_F + \nu$. Since the Wannier Hamiltonian describes the low-energy subspace, it is expected that the Wannier-interpolated bands will deviate more from the DFT results in the higher energy region. Therefore, the higher ν is, the larger η_{ν} is expected to be. The symbols η_1 and η_2 denote the band distance for the bands below $E_F + 1\text{eV}$ and $E_F + 2\text{eV}$, respectively. For η_1 , 98% of materials show a band distance below 10 meV, and 92% of materials show a small band distance below 2 meV. For η_2 , 93% of materials show a distance below 10 meV, and 70% of materials show a distance below 2 meV, highlighting the overall quality of Wannier Hamiltonians in our database. η distributions are provided in Supplementary Fig. 1b. Bands of all the metals and insulators are present in Supplementary Sections 4 and 5.

Unconventional spin Hall effect

Neumann's principle states that symmetries of the physical property must include all symmetries of the crystal³², indicating that a high-symmetry crystal would impose more restrictions on the allowed property. As a result, high-symmetry monolayers can only present one unique non-zero SHE component σ_{xy}^z ³³, which is called conventional SHE where the spin orientation is perpendicular to the xy -plane as shown in Fig. 1a. In contrast, low-symmetry materials can lead to unconventional SHE³³. The unconventional SHE is important for the field-free switching of perpendicular magnetization, since it can provide a canted torque to trigger the spin dynamics without external magnetic field³⁴.

To discover intrinsically unconventional SHE, we focus on metals with non-zero σ_{xy}^x and σ_{xy}^y as illustrated in Fig. 1a. A symmetry precision of 0.01 was applied in the ASE package³⁵ to screen materials with low-symmetry space group. After screening, we find 17 metals with low symmetry. Most of them present small SHCs. Notably, $\text{Y}_2\text{C}_2\text{I}_2$ is found to present a high σ_{xy}^x at the Fermi energy as shown in Fig. 1b. With the C2/m space group, $\text{Y}_2\text{C}_2\text{I}_2$ only has one mirror symmetry m_y as shown in Fig. 1c, allowing the spin current with spin orientation in xz -plane. To analyze the σ_{xy}^x and σ_{xy}^z behaviors, Fig. 1d presents the bands projected by SBC along the Γ - X - Γ path. The SOC results in an anti-crossing gap in

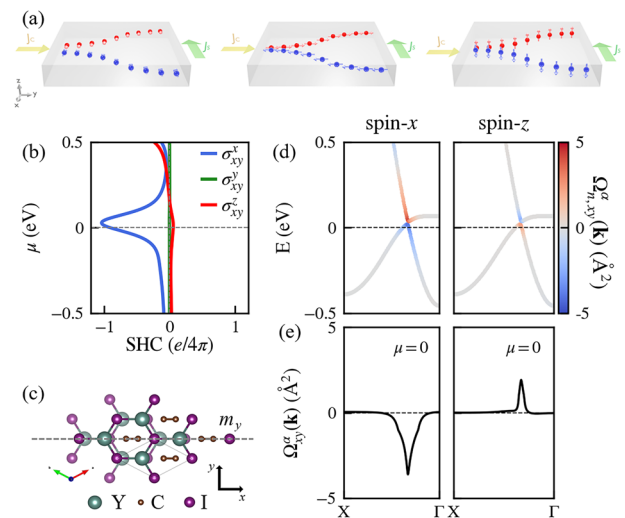


Fig. 1 | Canted spin current in $\text{Y}_2\text{C}_2\text{I}_2$. **a** Diagram of spin Hall effect components σ_{xy}^x , σ_{xy}^y , and σ_{xy}^z , respectively. **b** Fermi energy μ -dependent room-temperature spin Hall conductivity components, **c** atomic structure and mirror symmetry, **d** band-resolved spin Berry curvature defined by Eq. (3), and **e** $\mathbf{k}$ -resolved spin Berry curvature defined by Eq. (2) for $\text{Y}_2\text{C}_2\text{I}_2$ [C2/m] with unconventional spin Hall effect.

Table 1 | Stable metals with high conventional spin Hall conductivity σ_{xy}^z

Material	Space group	σ_{xy}^z ($e/4\pi$)
Ta_4Se_2	P4/nmm	-3.4
Sn_2P	P1	-2.7
Y_2Br_2	P-3m1	2.4

$\text{Y}_2\text{C}_2\text{I}_2$. The positive and negative $\Omega_{n,xy}^x$ are perfectly separated by the Fermi energy, leading to a peak of the integral Ω_{xy}^x . In contrast, σ_{xy}^z is limited, leading to a slightly canted spin orientation in the xz -plane. The magnitude of SBC was dealt with a logarithm function as Eq. (52) in ref. 22 for simplicity. The dynamical stability has been validated by its phonon dispersion in Supplementary Fig. 7.

High spin Hall conductivity

Although only a few materials have unconventional SHE components, the conventional component σ_{xy}^z is allowed for all the materials. The known σ_{xy}^z values in 2D materials are limited, for instance, MoS_2 monolayer only presents $\sigma_{xy}^z \approx 0.2e/4\pi$ with doping³⁶. To find high values for σ_{xy}^z in metals, SHC calculations are performed on all 210 metals including semi-metals, where 15 materials display exceptional $|\sigma_{xy}^z| > 2e/4\pi$, see details in Supplementary Section 2. Unfortunately, most of them are unstable according to phonon dispersions. Table 1 lists 3 materials whose mechanical stabilities have been validated by positive phonon dispersions, details are given in Supplementary Section 3.

Ta_4Se_2 monolayer presents the highest value $\sigma_{xy}^z = -3.4 e/4\pi$. Considering that Ta_4Se_2 is composed of heavy elements, its high SHC is attributed to strong SOC which opens up an anti-crossing band gap around Fermi energy. Figure 2a, b shows the bands of Ta_4Se_2 projected by SBC and energy-dependent σ_{xy}^z . For $\mu = 0$, the positive and negative SBCs compete as shown in Fig. 2c. The SHC of Ta_4Se_2 can be further enhanced by tuning the Fermi energy position. For $\mu = 0.21 \text{ eV}$, the negative SBC is dominant in the whole BZ as shown in Fig. 2d, maximizing the σ_{xy}^z amplitude to be $-12 e/4\pi$. This finding suggests that doping is an effective method to increase SHC in Ta_4Se_2 .

Apart from Ta_4Se_2 , Sn_2P and Y_2Br_2 are also found to exhibit high σ_{xy}^z values -2.7 and $2.4 e/4\pi$, respectively. Note that the high-

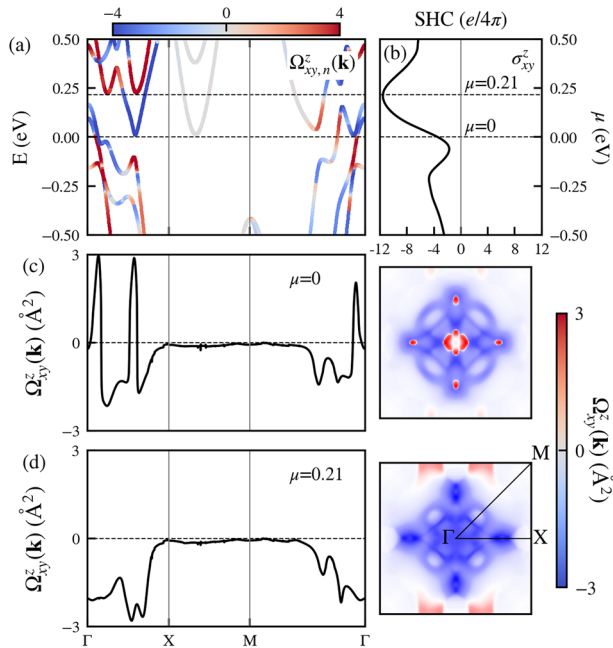


Fig. 2 | High spin Hall conductivity in Ta₄Se₂. **a** Band structure projected by spin Berry curvature, **b** Fermi energy μ -dependent room-temperature spin Hall conductivity σ_{xy}^z , and **c** $\mathbf{k}$ -resolved spin Berry curvatures respectively for $c\mu = 0$ and $d\mu = 0.21$ eV of Ta₄Se₂ monolayer.

symmetry Sn₂P monolayer reported by the MC2D database presents imaginary phonon frequency, thus its structure was relaxed without preserving any symmetry, leading to the P1 space group. It is found that although in a broken-symmetry regime, the relaxed structure is very close to the original structure, namely, the movement of every atom is less than 10^{−3} Å. Still, this low-symmetry Sn₂P monolayer with positive phonon dispersion is employed to study SHC. Besides, the possible magnetism in Y₂Br₂ is excluded by the negligible magnetic anisotropic energy. Additional details are given in Supplementary Section 3.

High spin Hall ratio

The conversion efficiency from charge current to spin current via the spin Hall effect is evaluated by the spin Hall ratio, defined as $\xi = \frac{2e}{h} \left| \frac{\sigma_{xy}^z}{\sigma_{xx}} \right|$ ³⁷. The semi-metal WTe₂ has been reported for an efficient conversion³⁸ since its resistive conductivity σ_{xx} is reduced by limited carrier around Fermi energy. Remarkably, Table 1 includes Y₂Br₂ which is also a semi-metal with high σ_{xy}^z . Figure 3a shows that Y₂Br₂ has a small energy overlap between the conduction band and the valence band. The SOC-induced anti-crossing gaps separate the negative and positive SBCs perfectly, leading to a σ_{xy}^z peak around Fermi energy as given in Fig. 3b. The dynamical stability of Y₂Br₂ is validated by the positive phonon dispersion as shown in Fig. 3c. Our finding suggests that Y₂Br₂ could be a promising candidate for high SHR.

Calculation of charge conductivity is required to compute the SHR in Y₂Br₂. For an intrinsic material, the electron-phonon coupling (EPC) is a fundamental interaction that affects the scattering rate, which can be accurately calculated using ab initio approach³⁹. The phonon-limited charge conductivity σ_{xx} is given by⁴⁰

$$\sigma_{xx} = \frac{-e}{S^{uc}} \sum_n \int \frac{d^2\mathbf{k}}{\Omega^{BZ}} v_{nkx} \partial_{E_k} f_{nk}, \quad (5)$$

where S^{uc} is the unit cell area, Ω^{BZ} is the BZ area, and $v_{nkx} = \hbar^{-1} \partial \epsilon_{nk} / \partial k_x$ is the band velocity. The linear variation of the electronic occupation function f_{nk}

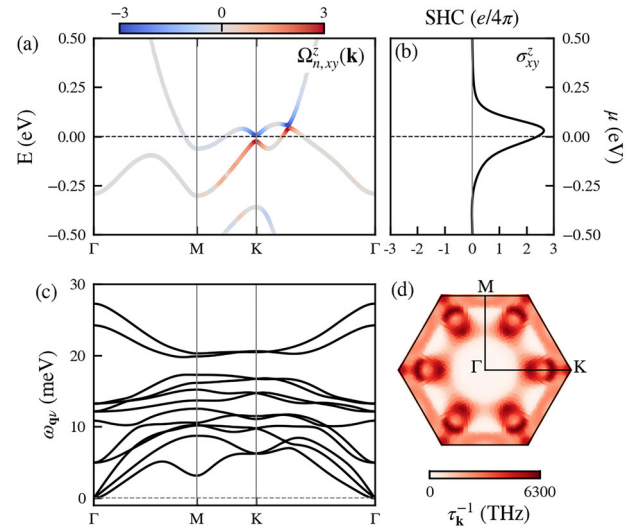


Fig. 3 | High spin Hall ratio in Y₂Br₂. **a** Bands projected by spin Berry curvature, **b** Fermi energy μ -dependent room-temperature spin Hall conductivity σ_{xy}^z , **c** phonon dispersion, and **d** $\mathbf{k}$ -resolved electron-phonon scattering rate of Y₂Br₂.

in response to $E_{\mathbf{k}}$, $\partial_{E_k} f_{nk}$, can be obtained by solving the Boltzmann transport equation⁴¹ which induces the scattering rate given by

$$\tau_{nk}^{-1} = \frac{2\pi}{h} \sum_{m\nu} \int \frac{d^2\mathbf{q}}{\Omega^{BZ}} |g_{mn\nu}(\mathbf{k}, \mathbf{q})|^2 \left[(n_{q\nu} + 1 - f_{mk+\mathbf{q}}^0) \delta(\epsilon_{nk} - \epsilon_{mk+\mathbf{q}} - \hbar\omega_{q\nu}) + (n_{q\nu} + f_{mk+\mathbf{q}}^0) \delta(\epsilon_{nk} - \epsilon_{mk+\mathbf{q}} + \hbar\omega_{q\nu}) \right], \quad (6)$$

where $g_{mn\nu}(\mathbf{k}, \mathbf{q})$ is the electron-phonon matrix element with phonon frequency $\omega_{q\nu}$. The $\mathbf{k}$ -resolved effective scattering rate, defined as $\tau_{\mathbf{k}}^{-1} = \sum_n \frac{-\partial f_{nk}^0}{\partial \epsilon_{nk}} \tau_{nk}^{-1}$, is given in Fig. 3d. Strong scattering occurs around the K points where Fermi energy crosses the anti-crossing gap. The small bandgap facilitates scattering between electron states, enhancing EPC and scattering rates. Besides, the pattern of Fig. 3d is determined by structural symmetry which also forces $\sigma_{xx} = \sigma_{yy}$ in Y₂Br₂.

The SHR of Y₂Br₂ can be obtained from the calculated SHC and charge conductivity. Y₂Br₂ presents an SHC of $\sigma_{xy}^z = 2.4 e/4\pi$, and a charge conductivity of $\sigma_{xx} = 6.1 e^2/h$. Defined as $\xi = \frac{2e}{h} \left| \frac{\sigma_{xy}^z}{\sigma_{xx}} \right|$, the room-temperature SHR in Y₂Br₂ is calculated to be $\xi = 0.4$, much higher than SHRs ~ 0.01 in typical heavy metals². Our results demonstrate that attributed to anti-crossings in semi-metal bands, Y₂Br₂ is a promising candidate for efficient charge-to-spin conversion.

Quantized spin Hall conductivity

As the last part of this work, gapped materials are investigated. It is instructive to compute the SHC of materials to discover topological insulators. In a 2D topological insulator, a pair of helical edge states with opposite spins emerge. Under the protection of time-reversal symmetry, electrons with opposite spins would move in opposite directions, leading to a spin current $\mathbf{J}^z = (\hbar/2e)(\mathbf{J}^\uparrow - \mathbf{J}^\downarrow)$ characterized by a quantized SHC with amplitude $\sigma_{xy}^z = e/2\pi$ composed of two conserved spin channels^{42,43}. However, the SHC plateau is not always well quantized; namely, some $\mathbb{Z}_2 = 1$ QSHIs manifest significant deviations from their expected quantized value⁴⁴.

To clarify the mechanism of quantized SHC, we calculated the σ_{xy}^z of 216 insulators with $T = 0$ K and $\eta = 5$ meV. We find 8 materials with SHC plateau in the bandgap. Among these materials, Bi₂, Hf₂Br₂, CuLi₂As, and Ti₂N₂I₂ have been validated as QSHIs with $\mathbb{Z}_2 = 1$ ^{16,45}. The other 4 materials are Li₂Tl₂, Hg₂S₂, Hg₄O₂, and In₂Bi₂ which possess soft phonon modes. Thus we only list the results of the former 4 materials in Table 2. All 4

Table 2 | List of quantum spin Hall insulators

Monolayer	E_{gap} (meV)	SG	σ_{xy}^z ($e/2\pi$)	S_z
Bi ₂	471	P-3m1	0.5	no
Hf ₂ Br ₂	69	P-3m1	−0.9	no
CuLi ₂ As	41	P-6m2	1.0	yes
Ti ₂ N ₂ I ₂	19	Pmmn	−1.0	yes

SG is the abbreviation of space group, σ_{xy}^z denotes the plateau value in band gap, and S_z illustrates whether the out-of-plane spin is conserved or not.

materials present a non-zero SHC plateau inside the bandgap. However, the SHC amplitudes vary and the β -Bi₂ monolayer shows $\sigma_{xy}^z = 0.5 e/2\pi$ while CuLi₂As presents a quantized SHC of $\sigma_{xy}^z = 1.0 e/2\pi$.

The potentially quantized SHC can be interpreted by spin conservation. SHC tensor is constrained by the crystal symmetry³³. If the SHC component σ_{xy}^z remains invariant under all the crystal symmetry operations, the associated spin current and edge state with spin S_z would be allowed. As a result, if any component other than σ_{xy}^z is allowed, the S_z conservation would be violated, leading to a broken quantized SHC. Symmetry analysis illustrates that in the β -Bi₂ monolayer, SHC components of σ_{xy}^x and σ_{xy}^y are allowed while σ_{xy}^z is prohibited. Indeed, Fig. 4a shows non-zero σ_{xy}^x and σ_{xy}^y SHC plateaus inside bandgap. Besides, the edge-state spin of Bi₂ is neither conserved nor robust as shown in Fig. 4c. In contrast, Fig. 4b shows that only σ_{xy}^z is allowed in CuLi₂As, which presents a pair of helical edge states with well-conserved S_z as shown in Fig. 4d. As a result, we find a quantized SHC $\sigma_{xy}^z = 1.0 e/2\pi$ in CuLi₂As. Besides, with the same space group as Bi₂, Hf₂Br₂ also presents a deviation from quantized SHC due to non-zero σ_{xy}^y and non-zero S_y on edge states. Ti₂N₂I₂ presents similar behaviors as CuLi₂As. It can be concluded that, induced by the crystal symmetry, the non-conserved S_z is destructive for quantized SHC. Calculations of SHC and spin-projected edge states are necessary to predict the value of SHC plateau in QSHI.

Discussion

The experimental feasibility of the promising materials is encouraging. As reported in Supplementary Section 3, all promising materials present positive phonon dispersions, suggesting their mechanical stability. In addition, since the materials have been extracted from the MC2D database, all of them should be exfoliable. For reference, the binding energy of graphene is 20 meV/Å² while Y₂C₂I₂, Y₂Br₂, Bi₂, Hf₂Br₂, and Ti₂N₂I₂ monolayers exhibit binding energies below 20 meV/Å²^{28,29}, indicating their potential for easy exfoliation. This has been demonstrated by the experimental synthesis of flat and buckled Bi₂ monolayers^{46,47}. Moreover, the Ta₄Se₂ monolayer, which has a fairly low binding energy of 33 meV/Å²²⁹, has also been confirmed to be thermodynamically stable⁴⁸. In the context of transport measurements in 2D materials, spin currents have been extensively researched in graphene and MoS₂ monolayers^{49–51}. Notably, quantum spin Hall effects have been observed in WTe₂ monolayers^{52,53}. These findings support the experimental feasibility of investigating SHE in the monolayers reported in this work.

In conclusion, leveraging the MC2D database, we implemented an automated Wannierization based on orbital information from the pseudopotentials for 426 non-magnetic monolayers including 210 metals and 216 insulators. The quality of Wannierizations has been validated by comparing DFT bands with Wannier bands, and the Wannier Hamiltonians are provided to the community. Exotic spin Hall effects are discovered in diversified materials. For metals, it is found that Y₂C₂I₂ displays canted spin Hall effect due to its low-symmetry structure. Besides, metallic candidates with high SHC values are listed, indicating Ta₄Se₂ as the optimal candidate. Efficient charge-to-spin conversion is reported in the semi-metal Y₂Br₂. For insulators, SHC plateaus are revealed in quantum spin Hall insulators, highlighting the importance of conserved S_z for quantized SHC. This comprehensive work enhances the understanding of spin Hall effect in 2D systems, suggesting promising materials for advanced spintronic

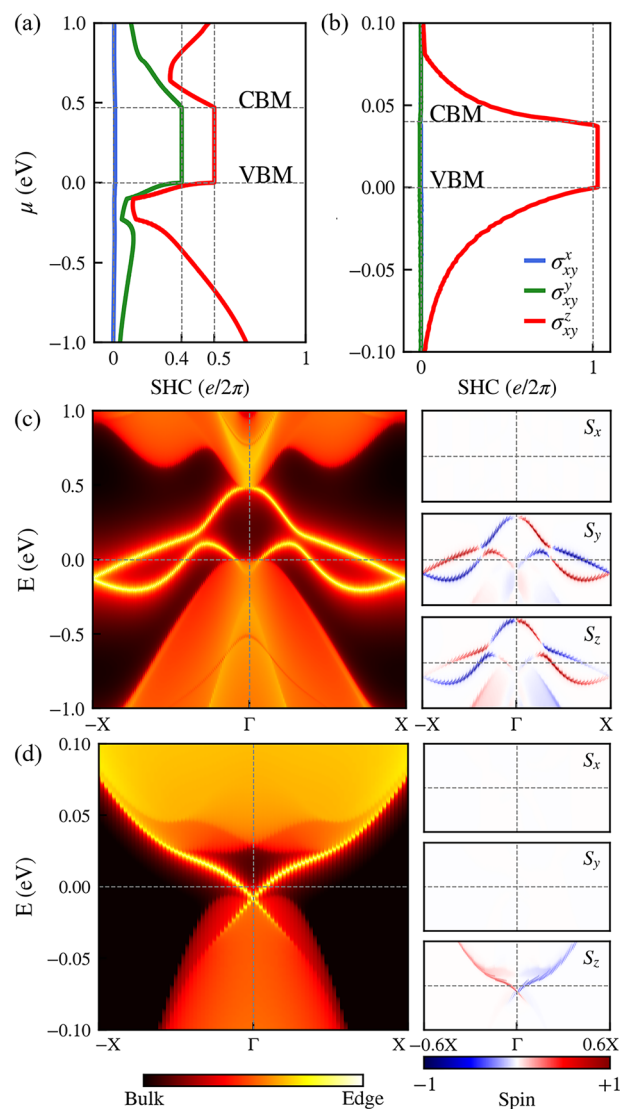


Fig. 4 | Spin Hall conductivities and edge states in quantum spin Hall insulators. Fermi energy μ -dependent spin Hall conductivity components of **a** Bi₂ and **b** CuLi₂As at $T = 0$ K. Edge states and spin projections of **c** Bi₂ and **d** CuLi₂As. The conduction band minimum is abbreviated as CBM, valence band maximum is abbreviated as VBM.

devices. Moreover, the provided Wannier database could be further employed to explore additional spin-orbitronic phenomena.

Methods

DFT calculations

DFT calculations were performed using the QUANTUM ESPRESSO package⁵⁴ within the Perdew–Burke–Ernzerhof (PBE) parametrization of generalized gradient approximation (GGA). AIIA⁵⁵ was employed to calculate 426 non-magnetic monolayers from MC2D database^{28,29} with a k -mesh density of 0.15 Å^{−1}, Marzari–Vanderbilt smearing⁵⁶ of 0.01 Ry, and 2D Coulomb truncation scheme⁵⁷. Although the sssp library⁵⁸ was used in MC2D database, it only includes scalar-relativistic pseudopotentials. To investigate SOC effect, our work employed PSEUDODOJO library⁵⁹ which includes both scalar-relativistic and fully relativistic pseudopotentials. The fully relativistic pseudopotentials were applied to MC2D materials without relaxation. After screening out 8 candidates with superior SHC properties, the scalar-relativistic PSEUDODOJO pseudopotentials were used to relax structures and calculate phonon dispersions. For the relaxed 8 structures, the fully

relativistic PSEUDODOJO pseudopotentials were applied to the DFT calculations and Wannierizations.

Wannierizations

In our work, the initial guess for projector is given by the hydrogenic orbital with hints from PSEUDODOJO⁶⁰. The semicore orbitals corresponding to deep bands are excluded from projections. The maximum of the frozen window is set as Fermi energy +2 eV (Fermi energy is the conduction band minimum for insulators) which is numerically verified to be the optimal value as shown in Supplementary Fig. 1a. To provide a high-quality Wannier Hamiltonian database, failed cases (42/426) have been manually corrected by tuning the frozen window, increasing the number of electronic states, or modifying initial projections.

Transport calculations

The spin Hall conductivity was calculated with the WANNIER90 package^{22,61} with a fine $\mathbf{k}$ -mesh density of $0.005 \text{ \AA}^{-1}$ for the Wannier interpolation. A Fermi-Dirac distribution function of $T = 300 \text{ K}$ and a broadening of 2 meV were considered if not specified. The charge conductivity was calculated using the EPW package^{62,63}. A coarse $12 \times 12 \times 1 \text{ k/q}$ -grid was interpolated on a fine $\mathbf{k/q}$ -grid of $360 \times 360 \times 1$. The electron wavefunctions and dynamical matrices were calculated using fully and scalar relativistic pseudopotentials, respectively⁶⁴. An adaptive smearing⁶⁵, a phonon frequency cutoff of 1 cm^{-1} and a 300 K temperature are used. Edge states of topological insulators were calculated by WannierTools⁶⁶.

Data availability

Details including electronic structures and phonon dispersions of promising materials investigated in this work, as well as fully relativistic bands of 426 monolayers, are given in the Supplementary Material. The input and output files, Wannier Hamiltonians, and bands data of promising materials and 426 monolayers are provided on Materials Cloud Archive⁶⁷.

Code availability

All codes used for this work are open-source. The latest versions are provided at <https://www.quantum-espresso.org/>, <http://www.wannier.org>, <https://www.aiida.net/>, and <https://www.wanniertools.org/>.

Received: 26 December 2024; Accepted: 16 April 2025;

Published online: 14 May 2025

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Acknowledgements

S.P. acknowledges the support from the Fonds de la Recherche Scientifique de Belgique (F.R.S.-FNRS). J.Z. and J.-C.C. acknowledge financial support from the Fédération Wallonie-Bruxelles through the ARC Grant “DREAMS” (No. 21/26-116), from the EOS project “CONNECT” (No. 40007563), and from the Belgium F.R.S.-FNRS through the research project (No. T.029.22F). Computational resources have been provided by the PRACE/EuroHPC award granting access to MareNostrum5 at Barcelona Supercomputing Center (BSC), Spain and Discoverer at SofiaTech, Bulgaria (OptoSpin project ID. 2020225411), and by the Consortium des Équipements de Calcul Intensif (CÉCI), funded by the F.R.S.-FNRS under Grant No. 2.5020.11 and by the Walloon Region.

Author contributions

J.Z. designed the study and performed calculations. S.P. and J.-C.C. supervised the project. All authors analyzed the results and contributed to writing the paper.

Competing interests

The authors declare no competing interests.

Additional information

Supplementary information The online version contains supplementary material available at <https://doi.org/10.1038/s41699-025-00562-4>.

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