Intrinsic spin Hall effect in topological insulators: a first-principles study

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The intrinsic spin Hall conductivity of typical topological insulators Sb$_2$Se$_3$, Sb$_2$Te$_3$, Bi$_2$Se$_3$, and Bi$_2$Te$_3$ in the bulk form, is calculated from first-principles by using density functional theory and the linear response theory in a maximally localized Wannier basis. The results show that there is a finite spin Hall conductivity of 100-200 ($\hbar/2e$)(S/cm) in the vicinity of the Fermi energy. Although the resulting values are an order of magnitude smaller than that of heavy metals, they show a comparable spin Hall angle due to their relatively lower longitudinal conductivity. The spin Hall angle for different compounds are then compared to that of recent experiments on topological-insulator/ferromagnet heterostructures. The comparison suggests that the role of the bulk in generating a spin current and consequently a spin torque in magnetization switching applications is comparable to that of the surface including the spin-momentum locked surface states and the Rashba-Edelstein effect at the interface.

I. INTRODUCTION

A topological state of matter is distinguished by its insulating bulk, but conducting surface states that are robust against disorder. The surface states in a topological insulator carry opposite spins while propagating in opposite directions [1]. Due to their strong spin-orbit coupling, topological insulators are capable of switching an adjacent thin-film ferromagnet in a bilayer structure without the need to apply any external magnetic fields [2]. Both the bulk and the surface states are reported to be involved in generating an electric-field-induced spin torque on the ferromagnet [3]. The spin polarization on the surface is due to the spin-momentum locking [1][4] of the surface states as well as the Rashba-Edelstein effect [5][6] at the interface with the ferromagnet, while the bulk of the topological insulator contributes via the intrinsic spin Hall effect [7][8]. Several experimental studies [3][4][9] have attempted to distinguish the contribution of the surface and the bulk states through magnetization switching in topological-insulator/ferromagnet heterostructures. However, theoretical estimates of the intrinsic bulk contribution are limited. In this work, we quantify the role of the bulk in spin generation by calculating the intrinsic spin Hall conductivity of four topological insulators namely Sb$_2$Se$_3$, Sb$_2$Te$_3$, Bi$_2$Se$_3$, and Bi$_2$Te$_3$, by using first-principles calculations. These materials, along with their alloys, are of the first experimentally realized [1][10][12] three dimensional topological insulators and have been studied more extensively especially in terms of the spin Hall effect.

The spin Hall effect is the accumulation of spin on the surface of a material in response to an applied electric field. Dyakonov and Perel [13] introduced the idea of generating spin polarization with a charge current via the Mott scattering which is a spin-dependent scattering off a Coulomb potential in the presence of spin-orbit coupling. They introduced a phenomenological spin electric coefficient term, which models the generation of a transverse spin current via an external electric field. The scattering potential by impurities and phonons can also result in an asymmetric scattering cross section leading to the accumulation of spin, an effect called the intrinsic spin Hall effect [14][15]. However, it has been shown [16] that even in the absence of extrinsic effects, spin accumulation occurs due to the finite spin-orbit coupling of the underlying crystal. Therefore, the contribution of non-zero orbital angular momentum in the Bloch wavefunctions along with an external electric field gives rise to a spin accumulation, a phenomenon known as the intrinsic spin Hall effect [17][18].

Topological insulators are a distinct state of quantum matter where the band structure is topologically different than that of ordinary/trivial insulators due to inverted bands at the Fermi level. Although insulating in the bulk, they are conducting on their surfaces where they meet a trivial insulator that is the vacuum [12][19]. Recently, topological insulators have been used to generate spin current to electronically switch the magnetization of a proximal thin-film magnet [2]. Compared to heavy metals, such as platinum [20] and tantalum [21], which are also used as spin-current generators, topological insulators are expected to consume less energy while yielding the same spin Hall angle [8], which is beneficial in realizing low power spintronics.

In this work we focus on the intrinsic ability of the bulk of topological insulators in generating spin currents through the spin Hall effect. We calculate the spin Hall conductivity of the four compounds (Sb/Bi)$_2$(Se/Te)$_3$ from first principles, that is by solving the Schrödinger equation in the framework of density functional theory and using the solution to calculate the linear response coefficients. Previous theoretical studies on the intrinsic spin Hall conductivity in topological insulators are limited to those of HgTe [22] by using first principles and...
Bi$_{1-x}$Sb$_x$ [23] and Bi$_2$Se$_3$ [24, 25] by using a tight binding and an effective Hamiltonian. It is worth noting that the calculation of spin Hall conductivity requires integration over the entire Brillouin zone and that all the bands below the Fermi energy contribute toward the spin Hall effect. Hence, the effective Hamiltonian, which is valid only in the vicinity of the Fermi energy, may not provide an adequate description of the spin Hall conductivity. First-principles calculations are therefore necessary to accurately quantify the intrinsic strength of spin generation in typical topological insulators, as well as understanding the bulk contribution in magnetization switching applications. We furthermore provide estimates of the spin Hall angle in different compounds using the large body of experimental studies on the topological-insulator/ferromagnet heterostructures.

Section II provides an overview of the experimental studies and techniques in estimating the spin Hall angle. Theoretical details of calculating the spin Hall conductivity are given in Section III. The symmetry properties of the crystal and the spin Hall conductivity tensor are discussed in Section IV. Results are presented in Section V along with a comparison of different values of the spin Hall angle reported in the literature. Section VI concludes the paper. Computational details and the first-principles setup are presented in the Appendix.

II. REVIEW OF EXPERIMENTS

The ability of a material to generate a spin current via the spin Hall effect is measured by the spin Hall angle (efficiency) $\theta$ which is proportional to the ratio of the spin current density $J_\alpha$ to the charge current density $J_\beta$, i.e., $\theta = (2e/h)J_\alpha^*/J_\beta = (2e/h)\sigma_{\alpha\beta}^*/\sigma_{\beta\beta}$ where $\sigma_{\alpha\beta}^*$ is the spin Hall conductivity and $\sigma_{\beta\beta}$ is the longitudinal charge conductivity. The spin Hall angle is usually measured via three different techniques namely the spin-torque ferromagnetic resonance (ST-FMR), the second harmonic Hall voltage, and the helicity-dependent photoconductance. These methods are briefly introduced in this section. In section V we show the spread of experimentally reported values of $\theta$ in various topological materials using different measurement schemes.

The ST-FMR technique was introduced by Liu et al. [20] in the context of spin Hall effect in heavy-metal/ferromagnet heterostructures such as platinum/permalloy bilayers. Later, Mellnik et al. [2] utilized this technique for topological-insulator/ferromagnet heterostructures. The ST-FMR method is based on the spin-torque driven magnetization resonance when a radio frequency (RF) charge current flows in the proximal spin-torque driven magnetization resonance when a radio frequency (RF) charge current flows in the proximal heavy metal or topological insulator layer. However, the magnetic field that is used in the second harmonic method is not static, but rotates in a plane perpendicular to the sample and parallel to the RF charge current. The first frequency component of the resulting Hall voltage in the sample is proportional to the RF current with the Hall resistance as the proportionality constant. The second harmonic component is shown to be proportional to the spin-transfer torque with a proportionality constant that depends on the anomalous Hall coefficient and the relative orientation of the magnetic field and the magnetization. Experimental works that utilize this measurement technique to quantify spin Hall angle include [9, 32, 33].

The photoconductive method has only recently been utilized to study spin Hall effect in topological insulators [7, 8]. Unlike the ST-FMR and the second harmonic method, the photoconductive method does not rely on the presence of a coupled ferromagnetic thin film to measure the spin Hall angle. In this method, spin accumulation that appears on the lateral edges of the sample due to a charge current is directly probed. That is, by shining a laser light with modulated helicity, the population of the spins can be locally changed. This change in the population of the spins reflects in a transverse spin-dependent voltage, which is shown to be proportional to the spin Hall angle with a proportionality constant that depends on the material geometry and transport properties such as sample resistivity. Therefore, by measuring the helicity-dependent photovoltage across the sample one can extract the value of the spin Hall angle.

III. SPIN HALL CONDUCTIVITY

In the linear response theory, the spin conductivity $\sigma_{\alpha\beta}^*$ is a tensor that connects the applied electric field $E_\gamma$ to the a spin current density $J_\alpha^*$ which is the response of the system, i.e., $J_\alpha = \sigma_{\alpha\beta}^*E_\beta$. The spin Hall conductivity is then a component in which $\alpha$ and $\beta$ are perpendicular to each other. Utilizing the Kubo formula, the spin conductivity can be written in terms of a Berry-like curvature
The spin Berry curvature is given as
\[ \sigma_{\alpha\beta} = -\frac{e^2}{h} \left( \frac{\hbar}{2e} \right) \int \frac{d^3 k}{(2\pi)^3} \sum_n f(\epsilon_n, k) \Omega^\gamma_{\alpha\beta,n}(k), \quad (1) \]
where \( f(\epsilon_n, k) \) is the Fermi-Dirac distribution function.

The spin Berry curvature is given as
\[ \Omega^\gamma_{\alpha\beta,n}(k) = \hbar^2 \sum_{m \neq n} -2 \text{Im} \left\{ \langle m | J^\gamma_n | k \rangle \langle k | m | \rangle v_\beta | n k \rangle \right\}, \quad (2) \]
where \( v_\beta \) is the velocity operator and \( J^\gamma_n = \{(v_\alpha, \sigma_\gamma)/2 = (v_\alpha \sigma_\gamma + \sigma_\gamma v_\alpha)/2 \) is the spin velocity operator. The structure of the spin Berry curvature suggests that the spin Hall effect can be viewed as an intermixture of the Bloch functions in the \( k \) space. It is worth noting that the integral of this curvature over the \( k \) space is not quantized \( [17, 22] \), unlike the Berry curvature in integer quantum Hall systems. Based on first-principles calculations, Eq. (1) has been used to study the spin Hall effect in semiconductors \( [35, 37] \) and heavy metals \( [38, 40] \). Previous calculations on topological insulators have been performed for HgTe \( [22] \) based on first principles and also for Bi\(_{1-x}\)Sb\(_x\) \( [23] \) and Bi\(_2\)Se\(_3\) \( [24, 25] \) based on a tight binding and an effective Hamiltonian, respectively. However, first-principles calculations of the spin Hall conductivity for (Bi/Sb)\(_2\)(Se/Te)\(_3\) crystals have not been reported previously.

We evaluate Eq. (1) based on first principles for (Bi/Sb)\(_2\)(Se/Te)\(_3\) crystals. Although this equation can be evaluated directly from the Bloch functions, a more computationally efficient method involves the Wannier functions. The Wannier functions are maximally localized within the same crystal structure which is classified as the trigonal crystal with a single three-fold high symmetry axis. The symmetry of the crystal is described by the symmorphic space group \( R\bar{3}m \) \( (#166) \). Figure 1 illustrates the primitive unit cell of these compounds along with the first Brillouin zone. There are five atoms per unit cell denoted in the figure. The symmetry of the crystal can predict several properties of the system such as the number of degeneracies and the selection rules to identify the components of the linear response tensor that evaluate to zero.

For instance, the group of the wavevector at the \( \Gamma \) point of the Brillouin zone is homomorphic to the point group \( D_{3d} \), and, therefore, the energy levels at the \( \Gamma \) point are labeled by the irreducible representation of the double group for group \( D_{3d} \). Since these irreducible representations are two-dimensional \( [44, 45] \), there are only two-fold degeneracies at the \( \Gamma \) point which represent the Kramers doublets and are protected by the time-reversal symmetry. The group of the wavevector at other points of the Brillouin zone is a subgroup of \( D_{3d} \), and, therefore, the bands do not merge into degenerate ones anywhere in the Brillouin zone. It is possible that accidental degeneracies appear at a number of points in the Brillouin zone where some energy levels get close to each other. These points lead to spikes in the Kubo formula, where the energy difference between different levels appears in the denominator and could make the integration over the Brillouin zone challenging.

Symmetry restricts the spin conductivity tensor through selection rules which determine the zero matrix.


\[
\begin{align*}
\sigma_x &= \begin{pmatrix} 0 & \sigma_{yx} & 0 \\ \sigma_{xy} & 0 & -\sigma_{yx} \\ 0 & \sigma_{xy} & 0 \end{pmatrix}, \\
\sigma_y &= \begin{pmatrix} \sigma_{yx} & 0 & \sigma_{yx} \\ 0 & 0 & -\sigma_{yx} \\ \sigma_{yx} & 0 & 0 \end{pmatrix}, \\
\sigma_z &= \begin{pmatrix} 0 & \sigma_{zy} & 0 \\ -\sigma_{zy} & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}.
\end{align*}
\]

As seen from the above equations, several components are zero, while not all non-zero components are independent. For instance, $\sigma_{yx} = -\sigma_{xy}$. There are only four independent components of the tensor namely $\sigma_{yx}$, $\sigma_{zy}$, $\sigma_{zx}$, and $\sigma_{xy}$. Since the majority of the magnetization switching experiments involve the spin current in the [111] direction (z direction here, i.e., $\sigma_{\alpha\beta}^{\gamma}$ components), we only need to calculate the $\sigma_{xy} = -\sigma_{yx}$ component which is referred to as the spin Hall conductivity from here on. We note that since we are dealing with non-magnetic space groups which contain the time-reversal operator as a group element, the Onsager reciprocity relations are satisfied [45]. As a consequence, the inverse spin Hall conductivity has an equal magnitude to that of the direct one.

V. RESULTS & DISCUSSIONS

First-principles calculations within the density functional theory are performed to obtain the band structure and the spin Hall conductivity of the four topological insulators Sb$_2$Se$_3$, Sb$_2$Te$_3$, Bi$_2$Se$_3$, and Bi$_2$Te$_3$. The details of the first-principles setup and the calculation of spin Hall conductivity are presented in the Appendix. Figure 2 illustrates the band structure of these four compounds on the left column as well as their corresponding spin Hall conductivity, $\sigma_{xy}$ as a function of the energy level on the right column. Both the band structure and the spin Hall conductivity are obtained in the Wannier basis. The bands shown in the energy window of the figure are composed of the s and p orbitals. The fully occupied d orbitals make highly narrow bands far below the Fermi level and, therefore, their contributions to the spin Hall effect is negligible. However, they are included in the pseudopotentials used for the first-principles calculations of the Kohn-Sham orbitals.

As seen from Fig. 2, the spin Hall conductivity is non-zero, albeit small compared to that of heavy metals, and is constant inside the gap and also to some extent beyond the gap. This suggests that the bulk in topological insulators could generate a finite spin current even when the Fermi level is located inside the gap and at zero temperature. Due to their finite spin Hall conductivity and limited longitudinal charge conductivity, the spin Hall angle of bulk topological insulators is comparable to that of heavy metals. Therefore, bulk topological insulators are an excellent candidate for energy-efficient charge-to-spin conversion in spin-based devices. This nonzero spin Hall conductivity, however, does not seem to be related to the non-trivial topological order in these materials as previous calculations [46] on trivial gapped semiconductors also reveal a finite spin Hall conductivity inside the gap.

The spin Hall conductivity in all of the four compounds studied here show mostly a similar dependence on the Fermi energy. That is, the spin Hall conductivity has a constant magnitude inside the energy gap and has peaks at certain energy levels where bands get too close to each other and result in accidental degeneracies. The values of spin Hall conductivity of Sb$_2$Se$_3$, Sb$_2$Te$_3$, Bi$_2$Se$_3$, and
FIG. 2. The band structure (left) and the spin Hall conductivity (right) of the four topological insulators Sb$_2$Se$_3$, Sb$_2$Te$_3$, Bi$_2$Se$_3$, and Bi$_2$Te$_3$. The energy axis is relative to the Fermi energy denoted by the gray horizontal line.

FIG. 3. Comparison of spin Hall angle of various bulk topological insulators measured experimentally and computed in this work using first principles. The horizontal axis specifies the average atomic number of the atoms in the unit cell.

Bi$_2$Te$_3$ at the Fermi level are 93.8, 113, 147, and 218 ($\hbar/2e)(S/cm)$, respectively. These values are of the same order of magnitude as the ones reported in the literature for slightly different materials by different methods [22, 23]. As one goes from low spin-orbit strength of Sb$_2$Se$_3$ to the relatively higher spin-orbit strength of Bi$_2$Te$_3$, the magnitude of the spin Hall conductivity at the Fermi level increases monotonically.

We summarise the recent experimental works on spin Hall effect in topological insulators in Table I. For each experiment mentioned in this table, several properties are listed such as the material, the measurement method, the thickness of the topological insulator, the magnitude of the spin Hall conductivity (if reported), $|\sigma_{xy}|$, the spin Hall angle, $\theta$, and the longitudinal charge conductivity $\sigma_{yy}$. The first-principles results in this work are listed in the last two columns where the magnitude of the spin Hall conductivity $|\sigma_{xy}|$ is obtained at the Fermi energy and the value of the spin Hall angle is estimated by using the longitudinal conductivity $\sigma_{yy}$ corresponding to each experiment, i.e., $\theta = (2e/\hbar)|\sigma_{xy}|/\sigma_{yy}$. For the non-stoichiometric compounds we report only estimates of the first-principles spin Hall conductivity by taking a weighted average over that of the stoichiometric ones. We note that since there are no experimental data available for Sb$_2$Se$_3$ and Sb$_2$Te$_3$ compounds, only the first-principles results are reported. It should be noted that, the photoconductive experiments measure a different component of the spin conductivity that is $\sigma_{zxy}$. Therefore, the corresponding first-principles values of $\sigma_{zxy}$ are reported instead.
In the absence of magnon scattering the second harmonic orbit torque. It was confirmed later by Wu et al. [9] that an asymmetric magnon scattering and not by the spin-linearity of the transverse Hall voltage is dominated by in certain configurations, the second harmonic methods of other methods. As mentioned by Yasuda et al. [33], has several data points. On the contrary, two of the data with the ST-FMR data points especially for Bi

**TABLE I. Comparison of spin Hall conductivity \( \sigma^z_{xy} \) and the spin Hall angle (efficiency), \( \theta = \frac{(2e/h)\sigma^z_{xy}}{\sigma_{yy}} \), from first-principles calculations and experimental observations.**

| Ref. | Material      | Method       | thickness nm | \(|\sigma^z_{xy}| (h/2e)(S/cm)\) | Experiments | first principles (this work) |
|------|---------------|--------------|--------------|-----------------------------------|-------------|-----------------------------|
| 2    | Bi₂Se₃        | ST-FMR       | 8            | 1.1 - 2.0 \times 10^3            | 2.0 - 3.5   | 5.7 \times 10^2 1.47 \times 10^2 0.26  |
| 32   | (Bi₁.₅Sb₀.₅)₂Te₃ | 2nd Harmonic | 3            | 180 - 425 2.227 \times 10^2      | *1.77 \times 10^2 0.79 |
| 27   | Bi₂Se₃        | ST-FMR       | 5 - 10       | 0.02 - 0.34 1 \times 10^3        | 1.47 \times 10^2 0.15 |
| 28   | Bi₂Se₃        | ST-FMR       | 20           | 0.1 2.50 \times 10^3             | 1.47 \times 10^2 0.06 |
| 3    | Bi₂Te₃        | ST-FMR       | 8            | 4.0 \times 10^3                  | 1.0 3.75 \times 10^3 2.18 \times 10^2 0.06 |
| 29   | Bi₂Se₃        | ST-FMR       | 7.4          | 0.16 9.434 \times 10^2           | 1.47 \times 10^2 0.15 |
| 4    | Bi₂Se₃        | ST-FMR       | 20           | 0.3 1.0 \times 10^3              | 1.47 \times 10^2 0.15 |
| 33   | (Bi₁₋ₓSbₓ)₂Se₃ | 2nd Harmonic | 5            | 160 2.44 \times 10^2             | *1.09 \times 10^2 0.45 |
| 8    | Bi₂Se₃        | Photoconductance | 9          | 0.0085 1.18 \times 10^3          | 12.72 \times 10^4 0.023 |
| 9    | Bi₂Te₃        | 2nd Harmonic | 6            | 0.08 1.5 \times 10^3             | 2.18 \times 10^2 0.15 |
|      | Sb₂Se₃        |              |              | 9.38 \times 10^1                 |             |
|      | Sb₂Te₃        |              |              | 1.13 \times 10^2                 |             |

* These values of \( \sigma^z_{xy} \) for alloys are obtained by a weighted average over that of non-alloy compounds.

† This value reflects the \( \sigma^z_{xy} \) component of the spin conductivity tensor.

To compare different experimental techniques and to put the reported values of spin Hall angle into perspective, Table I’s data are plotted in Fig. 3. This figure shows the spin Hall angle of different crystals versus the average atomic number of their unit cell. In this figure the experimental data points are denoted by circles and triangles whereas, our estimates of the spin Hall angle based on first-principles calculations are denoted by + and x symbols with the same color as their corresponding experimental value reported in other works. As seen from the figure the majority of the data points lie in the 0.1 < \( \theta \) < 1.0 range which is comparable to the values reported for the heavy metals such as 0.056 < \( \theta \) < 0.16 for platinum [26] and 0.12 < \( \theta \) < 0.15 for tantalum [21]. As mentioned previously, this suggests that for the same spin current, topological insulators require a lower value of charge current compared to heavy metals, which possess a relatively higher conductivity by more than an order of magnitude.

The first-principles results show a reasonable match with the ST-FMR data points especially for Bi₂Se₃ which has several data points. On the contrary, two of the data points [32, 33] related to the second harmonic Hall voltage method are orders of magnitude higher than that of other methods. As mentioned by Yasuda et al. [33], in certain configurations, the second harmonic methods tend to overestimate the spin Hall angle because the non-linearity of the transverse Hall voltage is dominated by an asymmetric magnon scattering and not by the spin-orbit torque. It was confirmed later by Wu et al. [9] that in the absence of magnon scattering the second harmonic method gives similar values of the spin Hall angle to the ones obtained by the ST-FMR method. The photoconductive experiments [8] provide a lower bound because no interface or ferromagnetic effects are present and therefore, the reported spin Hall angle could be attributed to the intrinsic spin Hall effect in the bulk. Since, the first principles results also reflect only the bulk contribution, one expects a better match with the photoconductive data points than with the other methods. However, in the case of Bi₂Se₃, the first-principles estimate of the spin Hall angle 0.023 is higher, by a factor of 3, than the value reported by Liu et al. [8] that is 0.0085. This discrepancy could be related to the premises in Ref. [8] where it is assumed that the spin accumulation at the lateral edges of the sample, due to a longitudinal charge current in the \( y \)-direction, are \( z \)-polarized because of the \( \sigma^z_{xy} \) component. However, based on the symmetry study in Sec. IV there is another component \( \sigma^z_{xy} \) that also contributes by canting the spin polarization at the edges of the sample which could have affected the measured photovoltage in Ref. [8].

VI. CONCLUSIONS

First-principles calculations of the spin Hall conductivity of typical topological insulators Sb₂Se₃, Sb₂Te₃, Bi₂Se₃, and Bi₂Te₃ show finite values at the Fermi energy. These values are lower by an order of magnitude than those of heavy metals. However, due to their relatively low current conduction capability, the spin Hall
angle in topological insulators is comparable to that reported in heavy metals. We compare theoretical results against experimental observations of spin Hall angle via direct helicity-dependent photovoltage measurements as well as in bilayers of topological insulators and ferromagnets using ST-FMR and second harmonic techniques.

The spin Hall angle values from first-principles calculations tend to underestimate the measured values. This is because in experiments, mechanisms other than the intrinsic spin Hall effect may be present, which are not included in our theoretical calculations. Yet, theoretical results are within an order of magnitude of measured values. Overall, the first-principles estimates of the spin Hall angle suggest that the intrinsic bulk contribution plays a significant role in spin generation and magnetization switching in bilayers of topological insulators and thin-film magnets. We acknowledge the limitation of first-principles study in terms of accuracy. The general band gap problem of density-functional theory calculations, certainly affects the calculations but the effect on the energy levels below the Fermi energy is minimal. Another issue pertains to the accuracy of the Brillouin zone integration. Since these materials have a large unit cell and, therefore, a large Wannier basis, the integration over the Brillouin zone is quite demanding. We have utilized an adaptive integration scheme to achieve an optimal tradeoff between complexity and accuracy. In our calculations, the numerical error is estimated to be less than 10% for a reasonable mesh size on a large CPU cluster (see Supplementary Information). For a higher accuracy, one might need additional computational power.

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Appendix: Computational Details

First-principles calculations are performed within the framework of density functional theory which is implemented in Quantum ESPRESSO suite \[49, 50\]. Projector augmented-wave [51] pseudopotentials [52] are utilized to reduce the cutoff energies and improve computational efficiency. The detailed setup description and parameters are listed in Table [II]. The crystal parameters of \( \text{Sb}_2\text{Se}_3 \) are obtained from Ref. [53]. The initial crystal parameters and atomic positions of the other three compounds are obtained from Materials Project [54]. The structural relaxation is performed on the crystals to set the total force to zero. The crystal parameters such as the lattice constant \( a \) and the angle between primitive vectors \( \alpha \) along with the relaxed atomic positions in the unit cell are provided in Table [III].

|                | \( \text{Bi}_2\text{Se}_3 \) | \( \text{Bi}_2\text{Te}_3 \) | \( \text{Sb}_2\text{Se}_3 \) | \( \text{Sb}_2\text{Te}_3 \) |
|----------------|-------------------------------|-------------------------------|-------------------------------|-------------------------------|
| **Pseudopotential Type** | Projector Augmented Waves [51] | Generalized gradient approximation [53] | | |
| **Exchange-Correlation functional** | | | | |
| **Kinetic \( E_{\text{cut}} \) (Ry)** | 56 | 56 | 55 | 34 |
| **Charge \( E_{\text{cut}} \) (Ry)** | 457 | 457 | 249 | 242 |
| **\( k \) mesh** | \( 8 \times 8 \times 8 \) | | | |
| **Number of Bands** | 90 | 90 | 70 | 70 |
| **Wannier \( s \) and \( p \) orbitals projections** | | | | |
| **Wannier \( k \) mesh** | \( 50 \times 50 \times 50 \) (adaptive \( 4 \times 4 \times 4 \) mesh) | | | |

The Bloch basis of the first-principles results are converted to the Wannier basis by projecting into \( s \) and \( p \) orbitals, which comprise the bands in the vicinity of the Fermi level. The initial projected Wannier functions are optimized to obtain a maximally localized set via Wannier90 code [45]. Utilizing the post processing module of the Wannier90 code developed by Ref. [59], the maximally localized Wannier functions are then used to calculate the spin Hall conductivity by evaluating the matrix elements that appear in the Kubo formula and integrating the Berry-like curvature over the Brillouin zone. The numerical details of the Wannier methods are listed in Table [II] as well.
SUPPLEMENTARY INFORMATION

In this document, we first review the linear response theory and provide a brief derivation of the Kubo formula for the spin Hall conductivity. Computational details such as the computation time for different stages of the calculations are provided along with a discussion of the accuracy of the calculations and the number of \( k \) points in the mesh required for Wannier interpolation.

Appendix S1: Linear response theory and the Kubo formula

In the linear response theory the response of a system to a perturbation is assumed to be dominated by a linear function of the perturbation which is generally a tensor called linear response tensor. Whenever the perturbation is an electric field and the system responds by generating a spin current, the tensor is called spin conductivity and its function of the perturbation which is generally a tensor called linear response tensor. Whenever the perturbation is \( \mathcal{O}(t) \), to the linear order in \( \mathcal{O}'(t) \), the Kubo formula goes as follows. The change in the expectation value of a given observable \( \mathcal{O}(t) \), can be written in terms of a correlation function, that is

\[
\delta \left\langle \hat{\mathcal{O}}(t) \rightangle = -\frac{i}{\hbar} \int_{-\infty}^{\infty} dt' \theta(t-t') \left\langle \left\{ \hat{\mathcal{O}}(t), \mathcal{O}'(t') \right\} \right\rangle_0,
\]

where the observable \( \hat{\mathcal{O}}(t) \) is in the interaction picture defined as \( e^{i\mathcal{H}_0 t/\hbar} \mathcal{O} e^{-i\mathcal{H}_0 t/\hbar} \). The correlation function includes a \( \left\langle \right\rangle_0 \) which denotes an ensemble average over the occupied states of the unperturbed \( \mathcal{H}_0 \). Taking the Fourier transfer of the above equation results in

\[
\delta \left\langle \hat{\mathcal{O}}_\omega \rightangle = -\frac{i}{\hbar} \int_0^\infty dt e^{i\omega t} \left\langle \left\{ \hat{\mathcal{O}}(t), \mathcal{O}'_{\omega} \right\} \right\rangle_0.
\]

Replacing \( \hat{\mathcal{O}}(t) \) with \( e^{i\mathcal{H}_0 t/\hbar} \mathcal{O} e^{-i\mathcal{H}_0 t/\hbar} \) and inserting a completeness relation \( \sum |m\rangle\langle m| = 1 \), one obtains

\[
\delta \left\langle \hat{\mathcal{O}}_\omega \rightangle = -\frac{i}{\hbar} \int_0^\infty dt e^{i\omega t} \sum_{n,m} f(\epsilon_n) \left[ \langle n|\mathcal{O}|m \rangle \langle m|\mathcal{H}'_{\omega}|n \rangle e^{i(\epsilon_n-\epsilon_m)t/\hbar} - \langle n|\mathcal{H}'_{\omega}|m \rangle \langle m|\mathcal{O}|n \rangle e^{i(\epsilon_m-\epsilon_n)t/\hbar} \right],
\]

where \( f(\epsilon_n) \) is the Fermi-Dirac distribution function. Performing the integral results in

\[
\delta \left\langle \hat{\mathcal{O}}_\omega \rightangle = -i \sum_{n,m \neq n} f(\epsilon_n) \left[ \frac{\langle n|\mathcal{O}|m \rangle \langle m|\mathcal{H}'_{\omega}|n \rangle}{\epsilon_n - \epsilon_m + \hbar \omega} - \frac{\langle n|\mathcal{H}'_{\omega}|m \rangle \langle m|\mathcal{O}|n \rangle}{\epsilon_m - \epsilon_n + \hbar \omega} \right].
\]

Appendix S2: Kubo formula for spin Hall conductivity

In the spin Hall effect, the response of the system is a spin current operator defined as \( \mathcal{O} = \mathcal{J}_\alpha^\gamma = (\hbar/2) \{ v_\alpha, \gamma \} / 2 \). A time varying electric field represented by its frequency components \( E_\beta e^{-i\omega t} \) results in \( \mathcal{H}'_{\omega} = -\mathbf{J} \cdot \mathbf{A} \) where \( \mathbf{J} = -e\mathbf{v}_\beta \) is the charge current operator and \( \mathbf{A} = E_\beta e^{-i\omega t}/i\omega \) is the vector potential. Therefore, by replacing \( \mathcal{H}'_{\omega} \) with \( E_\beta /i\omega \) and expanding the \( (\epsilon_n - \epsilon_m + \hbar \omega)^{-1} \) terms at the \( \omega \to 0 \) limit, one obtains the spin Hall conductivity \( \sigma_{\alpha\beta}^\gamma = \delta \left\langle \mathcal{J}_\alpha^\gamma \right\rangle / E_\beta \) as follows

\[
\sigma_{\alpha\beta}^\gamma = \hbar (eh/2) \sum_{n,m} f(\epsilon_n) \left[ \frac{\langle n|\{ v_\alpha, \gamma \}/2|m \rangle \langle m|v_\beta|n \rangle - \langle n|v_\beta|m \rangle \langle m|\{ v_\alpha, \gamma \}/2|n \rangle}{(\epsilon_n - \epsilon_m)^2} \right],
\]

Relabeling the energy eigenstates with the Bloch functions \( |n,k\rangle \) for a crystalline system and rearranging the sums one obtains

\[
\sigma_{\alpha\beta}^\gamma = -\left( \frac{e^2}{h} \right) \left( \frac{\hbar}{2e} \right) \int \frac{d^3k}{(2\pi)^3} \sum_n f(\epsilon_{n,k}) \Omega_{\alpha\beta,n}^\gamma(k),
\]

where \( \Omega_{\alpha\beta,n}^\gamma(k) \) is called the spin Berry curvature where

\[
\Omega_{\alpha\beta,n}^\gamma(k) = \hbar^2 \sum_{m \neq n} \frac{2Im \{ \langle n|k|v_\alpha, \gamma \rangle /2|m|k \rangle \langle m|v_\beta|n k \rangle \}}{(\epsilon_{n,k} - \epsilon_{m,k})^2}.
\]
We note that both the spin and the charge current operators need to be divided by the volume of the system to give the correct units of the current density. However, since there are two sums over the Brillouin zone, the volumes would be cancelled as the sums are rewritten in terms of $k$-space integrals.

Appendix S3: Computational details

The first principles calculations of the common topological insulators with the formula $A_2B_3$ are quite demanding because of their more complex crystal structure compared to that of heavy metals such as Pt. Also, since there are five atoms in the unit cell of these compounds, compared to one atom in Pt, the volume of the unit cell is about an order of magnitude larger than that of Pt. Therefore, one expects to see about an order of magnitude longer computation times compared to the case of Pt. Here we report the computation time for different stages of the first principles calculations such as the structural relaxation (to achieve zero total force), the self-consistent and non-self-consistent field calculations (to obtain eigenstates on a regular basis), maximally localized Wannier set by iterative optimization, and finally integrating the Berry-like curvature of the spin Hall conductivity over the entire Brillouin zone. The computation times are listed in Table S1 for all the four topological insulators considered in this work. We note that these reported times are for a cluster of 96 CPUs. As seen from the table above the last stage, which involves the integral over the Brillouin zone, is computationally the most demanding one.

|                          | Bi$_2$Se$_3$ | Bi$_2$Te$_3$ | Sb$_2$Se$_3$ | Sb$_2$Te$_3$ |
|--------------------------|--------------|--------------|--------------|--------------|
| Structural Relaxation    | 1095 s       | 90 s         | 870 s        | 640 s        |
| Self consistent field (Monkhorst $8 \times 8 \times 8$ mesh) | 380 s       | 780 s        | 960 s        | 720 s        |
| Non-self consistent field (regular $8 \times 8 \times 8$ mesh) | 1320 s      | 2670 s       | 3600 s       | 2365 s       |
| Maximally localized Wannier set (# of iterations) | 9400 s (32000) | 11000 s (32000) | 4000 s (12500) | 10660 s (32000) |
| Wannier $k$ mesh ($50 \times 50 \times 50 (adaptive \ 4 \times 4 \times 4 \ mesh)$) | 70850 s | 107235 s | 27850 s | 44040 s |

Appendix S4: Choosing the mesh size

It has been shown elsewhere \cite{38,39} that the number of the $k$ points required for a fairly accurate integration over the Brillouin zone is of the order of $10^6$. Fig. S1 shows $\sigma_{xy}$ for Bi$_2$Se$_3$ on a regular mesh with $100 \times 100 \times 100$ and $200 \times 200 \times 200$ points, respectively. As seen from the figure the results are very noisy. The reason is that the kernel of the Brillouin zone integral contains several spikes due to accidental degeneracies in the band structure. Therefore, a very fine mesh is needed to capture all those spikes. To alleviate this problem one could resort to an adaptive way of choosing the $k$ points of the mesh. In the adaptive method, the mesh gets finer in the vicinity of a spike while it is coarser in the smoother areas. This way, the spikes can be taken into account more accurately. Figure S1 shows the result for the adaptive mesh which converges for a mesh of $50 \times 50 \times 50$ (with an additional $5 \times 5 \times 5$ mesh around each spike) and is much smoother than the one obtained from a regular mesh with fixed distancing, for an approximately equal number of points in total. For example, a mesh of $50 \times 50 \times 50$ with adaptive meshing of $5 \times 5 \times 5$ around each spike contains about $10^7$ total number of points which is of the same order of magnitude as that of a $200 \times 200 \times 200$ regular mesh but captures the spike better than the regular mesh. This suggests that with roughly equal number of points, one can achieve a more accurate result by distributing the $k$ points more efficiently using the adaptive meshing method. Furthermore, we reproduce the results previously reported for platinum by comparing meshes with different sizes in Fig. S2. As the mesh gets finer, the values of the spin Hall conductivity converge at each energy level. This figure shows that an adaptive mesh with $50^3$ initial points and $5^3$ additional points at each spike can reproduce a good match (within a 5% error) with the results reported in Refs. \cite{38} and \cite{39}.

Appendix S5: Convergence and error

In order to make sure that the finite value of spin Hall conductivity at the Fermi energy is not a numerical error, we perform convergence tests in which we observe the behavior of $\sigma_{xy}$ as the mesh size increases. Figure S3 plots
FIG. S1. The effect of the size and the shape of the integration mesh comparing the regular (a) and the adaptive (b) meshes for Bi$_2$Se$_3$. The adaptive mesh leads to a smoother result and a faster convergence than the one obtained by the regular mesh due to the more efficient distribution of the k points in the vicinity of the spikes.

FIG. S2. The effect of the mesh size on the spin Hall conductivity of Pt. The legend shows the initial size of the adaptive mesh with a 5 × 5 × 5 additional mesh around the spikes. The finest mesh reproduces the previous results reported in Refs. [38] and [39] within a good accuracy (within a 5% difference).

$\sigma_{xy}^x$ versus the number of k points in the mesh considering the mesh is regular and not adaptive. If the we consider the last value corresponding to the finest mesh as the converged value $\sigma_{xy}^x = -1.63 \times 10^2$, the relative error for the 50 × 50 × 50 is -9.8%.
FIG. S3. The convergence of the spin Hall conductivity of Bi$_2$Se$_3$ at the Fermi energy for different number of k points on the mesh. Assuming that the value for a mesh of $300^4$ points is the converged value, that is $-1.63 \times 10^2$, the relative error for the mesh with size $50^3$ is -9.8%.