Band tilt-induced nonlinear Nernst effect in topological insulators: An efficient generation of high-performance spin polarization

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Topological insulators(TIs) hold the promise as a platform for spintronics applications due to the fascinating spin-momentum locking (SML) of the surface states. One particular interest lies on using TIs as spin polarized sources for spintronics structures. Here, we propose the band tilt-induced nonlinear Nernst effect (NLNE) in TIs as a clean and efficient route to generate high-performance spin polarization (SP). We show in the presence of SML and Hexagonal warping effect, a finite band tilt can induce an imbalance of two spin carriers and effectively give rise to spin-polarized NLNE current in TIs. In our scheme, both the spin current and charge current regime can be achieved under the thermal drive. The obtainable SP can be efficiently tuned either in a smooth or rapid way, exhibiting highly flexible tunability. In addition, near-unity SP can be generated within a wide range of tunable parameters, which is also found to be robust against temperature. Therefore, our work provides a mechanism to realize controllable room-temperature high-degree SP based on TIs, being of essential importance for future spintronics applications.

Introduction.—Topological spintronics which revolves around the topological states and carriers’ spin, in addition to carriers’ charge, has attracted growing attention over the past decade owing to its promising applications in spin-based future technologies [1–10]. A primary requirement for the practical applications of spintronics is to generate spin polarized carriers utilizing proper materials. Though much progress has been made [11–24], the realization of high-performance spin-polarized currents, i.e., with high degree of SP, long lifetime and robustness against temperature, remains to date a critical challenge in spintronics.

3D TIs are inherent SP generators [25, 26], which allows for generating SP without the need of magnetism such as ferromagnetic layers. Several experiments have demonstrated the observation of the (electrical) current-induced SP in TIs [15, 27–34], which are believed to be resulted from the intrinsic SML of the topological surface states (TSSs). Besides SML, however, it is reported that bulk spin Hall effect as well as the trivial 2D electron gas with strong spin-orbit coupling near the surfaces in TIs are also possible contributions [34–40]. This makes the origin of the experimentally-measured SP in TIs unclear. While the search for high-quality TIs that can reach bulk-insulating states will exclude most of the disruptive SP contributors, it can not guarantee the SP generated in TIs currently are applicable for spintronics. As has been pointed out recently [41], the net SP generated in these experiments are mostly contributions from the in-plane spin projection of the intrinsic states, rendering the nonequilibrium ensemble SP obtainable in TIs being relatively minuscule. Consequently, to utilize TIs as spin polarized sources, alternative mechanisms for the SP generation are still highly demanded.

Despite the intense interest and early success in the filed of nonlinear responses [42], transport effects in the respect of SP (or spin-polarized transports), have not yet been discussed in the nonlinear regime so far. Here, we propose the band-tilt induced NLNE in 3D TIs as a new mechanism to generate highly tunable SP. Contrast to the NLNEs reported previously [43–49], the one investigated here has a quantum origin from the finite band tilt effect in TIs, which effectively generates the transverse nonlinear spin and charge currents coplanar with the thermal gradient. Interestingly, accompanied with the SML and hexagonal warping effect in TIs, the band tilt-induced distortion of the Fermi surface results in an imbalance of spin-up and spin-down carriers, as schematically shown in Fig. 1. As an immediate result, nonzero net SP dependent on multi parameters can be generated under a thermal drive. Being a cooperative effect of SML, hexagonal warping, and band tilt, the thermally induced SP carried by NLNE current will not be concealed by any other possible states, in striking contrast to the previous SP generations in TIs. Moreover, since the generated SP is associated with the NLNE, which is a purely nonequilibrium Fermi surface quantity [42, 49, 50], it represents an ensemble SP density exactly caused by momentum asymmetry. Thus the obtainable SP is a pure nonequilibrium ensemble quantity and expected to be fully applicable for spintronics. We further show the proposed NLNE in TIs, remarkably, provides near-unity SP with high tunability as well as robustness against temperature. Therefore, our scheme to generate SP will pave the way for TI-based spintronics applications. In what follows, we present the semiclassical formalism for the spin-polarized NLNE in 3D TIs and demonstrate the generation of high-performance SP in detail.

Surface states in 3D TIs.—The standard Hamiltonian for the surface state of TI is given by

$$H_0 = \left[ v_F \hbar k \times \hat{z} + \lambda k \times \hat{y} (k_z^2 - 3k_y^2) \right] \sigma,$$

where $v_F$, $\hbar$, $\lambda$, and $\sigma$ denote the Fermi velocity, Planck constant, spin-orbit coupling, and Pauli matrices, respectively. The Hamiltonian $H_0$ describes the free electron behavior on the surface of a TI, including the spin-orbit coupling and the warping effect.

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where \( v_f \) is the Dirac velocity and \( \lambda \) denotes the hexagonal warping strength promised by the \( C_{3v} \) rotational invariance the rhombohedral crystal structure (e.g. Bi\(_2\)Te\(_3\)) [51]. It has been shown by Zhang et al. [52, 53], that the TSSs can be efficiently manipulated through adjusting the surface potential, such as introducing the in-plane (surface) magnetization via doping the TIs or the proximity effect of ferromagnetic insulators. In such cases, both \( C_{3v} \) and time-reversal symmetry (TRS) of the system would be broken, and the surface state Hamiltonian will be correspondingly modified. To the leading term, it can be effectively described by the band tilt \( H_t = \nu_{\lambda}(k) \sigma_0 \) \((t = x, y)\). Therefore, an effective TSS Hamiltonian can be written as

\[
H = H_0 + H_t. \tag{2}
\]

In effect, besides the linear band tilt term, the symmetry breaking can also lead to additional terms (higher-order in \( k \)) in the surface Hamiltonian, which might also be important for the physical properties of TSSs. As a conceptual and theoretical work, here we mainly focus on the tilt-induced transport effects for the TSSs.

The surface band dispersion based on Eq. (2) is \( E_{\pm,k} = v_f k_t \pm \sqrt{(v_f \hbar)^2 k_t^2 + \lambda^2 k^2 \cos^2 \phi} \), where \( \phi \) is the azimuth angle of momentum \( k \) with respect to \( k_x \)-axis, “\( \pm \)” denotes the conduction (upper) and valence (lower) band, respectively, and the corresponding eigenstates are defined as \( \pm \), \( \pm \). Note that, the warping term coupled to \( \sigma_z \) in Eq. (2) explicitly introduces an out-of-plane SP along \( s_z \)-direction [54]. However, here we are more interested in the in-plane SP \( \langle \sigma_{||} \rangle \), obtained as \( \langle \pm, k | \sigma_{||} | \pm, k \rangle = \langle \sigma_x, \sigma_y \rangle = \pm \frac{v_f k}{\tau} \), with \( E_0 = |E_{\pm,k}| \). This explicitly reveals the SP is perpendicularly locked to the momentum, independent of the warping and/or band tilting effect, as shown in Fig. 1.

Before proceeding, we want to mention that a particle-hole asymmetry (PHA) term \(( \propto k^2 \) can also exist for the TSSs, which has been found important in many intriguing effects in TIs [50, 55–58]. In effect, as shown by the contour plots in black dashed lines in Fig. 1, it suppresses the warping effect, which in turn inevitably weakens the warping-effect-dependent nonlinear responses. Fortunately, it can be found that in Bi\(_2\)Te\(_3\) the warping and tilting effect is just slightly suppressed by PHA, especially at relatively lower Fermi energies, which thus will not affect the main results of this work [59].

To better elaborate the generation mechanism of the spin polarized NLNE, we here in the main text consider the particle-hole symmetric TSSs for the following discussions.

**Semiclassical Boltzmann transport formalization.**—The nonequilibrium distribution of carriers in response to external fields can be phenomenally depicted by the Boltzmann transport theory. Within the relaxation time approximation, the Boltzmann transport equation in steady state is

\[
\tau (\hat{v} \partial_{\nu} f + \hat{k} \partial_{\nu} f) = -(f - f_0), \tag{3}
\]

where \( f_0 \) and \( f \) is the equilibrium and perturbed Fermi-Dirac distribution function, respectively. For simplicity, the relaxation time \( \tau \) will be treated as a constant.

Besides electrical generation, applying thermal gradient is another feasible approach to generate SP [60–65]. Specifically, the linear thermoelectric effect has been recently reported to generate and control the spin-polarized current in topological materials [66]. Here let’s consider a configuration in which only the thermal gradient is present, i.e., \( E, B = 0, \nabla T \neq 0 \). Thus, Eq. (3) can be reduced to \( \tau (\hat{v} \partial_{\nu} f + \hat{k} \partial_{\nu} f) = 0 \), where \( g_k^{(n)} \) implies the \( n \)-th-order \((n \geq 1)\) correction in \( \nabla T \) to \( f_0 \), i.e., \( f = f_0 + \sum_n g_k^{(n)} \). The corrections \( g_k^{(n)} \) are generally obtained via the iterative substitutions in Boltzmann equation [59, 67], through which we have

\[
g_k^{(1)} = \frac{\tau}{\hbar} \frac{\partial f}{\partial \mu} \nabla T, \quad g_k^{(2)} = \frac{\tau^2}{\hbar^2} \frac{\partial f}{\partial \mu} \nabla T - \frac{\partial^2 f}{\partial \mu^2} \nabla T \nabla T. \tag{4}
\]

Without loss of generality, a thermal gradient applied along \( x \)-direction is considered above. Note that, here the first-order correction \( g_k^{(1)} \) generally leads us to the linear responses, while the correction \( g_k^{(2)} \) is the one plays the key role in the nonlinear transport discussed in the following. Analogous to the charge and energy current [68, 69], the spin current operator can be defined as \( j_{\nu}^{\sigma^\prime} = \frac{\hbar}{2} \{ \nu^\prime, \sigma^\prime \} \), where \( \nu^\prime = \partial H_k / \partial k_n \), and \( s_{\nu^\prime} \) indicates the \( \nu^\prime \)-spin component with Pauli matrix \( \sigma^\prime \). In the absence of electric and magnetic field, the semiclassical equations of motion of Bloch electrons are simplified as \( \dot{k} = 0, \dot{\nu} = \langle \nu^\prime \rangle_n = \frac{\partial E_n}{\partial \mu} \) [68]. Therefore, the charge current and the spin current are respectively given as:

\[
\begin{align*}
&j_{\nu} = \sum_n \int [dk] f_{n,k} \langle -\nu \hat{v}_{\nu} \rangle_n, \\
j_{\nu}^{\sigma^\prime} = \sum_n \int [dk] f_{n,k} \langle \hat{\sigma}_{\nu^\prime} \rangle_n,
\end{align*}
\]

where \( f_{n,k} \) and \( \langle \ldots \rangle_n \) represents the distribution function and the average with regard to \( n \)-th eigenstate \( \varepsilon_{n,k} \), respectively. Note that, \( j_{\nu}^{\sigma^\prime} \) implies the spin current along \( \nu \)-direction with spin pointing in direction \( \nu \). In this work, we focus on the nonlinear responses to temperature gradient stemming from the second-order correction \( g_k^{(2)} \). It is important to note that,
and $E$ or $s$ indicates the magnitudes and the black lines are shown for $v$ that remains basically unchanged even with varying magnetic contrast to the previously reported nonlinear planar effects can be further increased by a finite band tilt [Fig. 2(a)], in$nal warping-induced enhancement [49], it shows the NLSNE sections at of additional symmetry, or considering the band warping/band for Eq. (2), the second-order nonlinear spin Nernst current only requires broken TRS are required to guarantee a finite second-order nonlinear charge current.

**Band tilt-enhanced spin current.—** Using the SP obtained for Eq. (2), the second-order nonlinear spin Nernst current (NLSNE) is found to be

$$j_y^s = \tau^2 \frac{\hbar}{2} \int |dk| v_f h_k E_0 \left[ \frac{2h v_x \partial f_0}{E} \right] \frac{2h v_x \partial f_0}{\partial k_x} + (E_k - \mu) \left( \frac{E_k}{\hbar^2 T^2} \right) \left( \frac{2h v_x \partial f_0}{\partial k_x} \right) (\nabla_x T)^2,$$

where the upper branch of the surface state (denoted as $E_k$) is considered. Note that, $j_y^s$ is finite even without the breaking of additional symmetry, or considering the band warping/band tilting effect. Such a feature is implied by the non-zero intersections at $\lambda = 0$ in Fig. 2(a). In addition to the hexagonal warping-induced enhancement [49], it shows the NLSNE can be further increased by a finite band tilt [Fig. 2(a)], in contrast to the previously reported nonlinear planar effects that remains basically unchanged even with varying magnetic fields [49, 50]. Such evident enhancement along with the $v_t$ and $E_f$-dependency are presented in Fig. 2(b), where the colors indicates the magnitudes and the black lines are shown for isovalues at 0, 0.3, 1.0, respectively. Line cuts with different chemical potentials $E_f$ is also given in Fig. 2(c). Obviously, even a moderate tilting strength ($v_t/h \sim 0.4$) can easily increase the NLSNE by hundreds of times on its order of magnitude compared to the non-tilted case at $v_t = 0$, especially at higher chemical potentials.

One finds that, under the drive of thermal gradient $\nabla_T$, only the NLSNE current with transverse SP ($j_x^s$ and $j_y^s$) can exist [59]. Here we focus on $j_y^s$, the component transverse to $\nabla_T$. Particularly, both $j_x^s \propto (\nabla_x T)^2$ and $j_y^s \propto (\nabla_y T)^2$ will be simultaneously generated, given a misalignment between the externally applied thermal gradient and the principal axes. In such a case, a real-space spin texture of the generated NLSNE can be determined as $|j_y^s| = \sqrt{(j_x^s \sin \theta)^2 + (j_x^s \cos \theta)^2}$, where $\theta$ stands for the polar angle in the rotational coordinate system that is commonly exploited in the conventional transport measurement [42]. The numerically calculated spin current textures $|j_y^s|$ with $E_f = 0.8$ eV (solid) and $E_f = 0.5$ eV (dashed) are plotted in Fig. 2(d). Interestingly, the SML texture is still preserved for the NLSNE, irrespective of the anisotropy in magnitude. Another interesting angular dependency may come from the crossed thermal gradient and band tilt [59].

**Nernst current and the spin polarization.—** Based on Eq. (4), the second-order NLNE is obtained as

$$j_y = -e \tau^2 \int |dk| v_f \left[ \frac{2h v_x \partial f_0}{E} \right] \frac{2h v_x \partial f_0}{\partial k_x} + (E_k - \mu) \left( \frac{E_k}{\hbar^2 T^2} \right) \left( \frac{2h v_x \partial f_0}{\partial k_x} \right) (\nabla_x T)^2,$$

which differs from the NLSNE in Eq. (5) merely by factor $-\frac{\hbar}{2e} \langle \sigma_x \rangle$ inside the integral. It is worthy noting that $j_y$ vanishes unless $v_y$ contains an extra momentum-independent contribution from the finite band tilt, i.e., $v_y \rightarrow v_t/h \pm v_y$. As such, we find the generated NLNE current only flows along the direction of the band tilt, e.g. $y$-direction ($\nabla_T \sin \theta$) in the current configuration. This is different from NLSNE which are generated in both transverse and longitude directions. In effect, a longitudinal or transverse nonlinear charge current can be generated if the Dirac cone state is perfectly tilted along $x$- or $y$-direction, respectively [59].

Unlike the NLSNE, the NLNE $j_y$ is asymmetric with respect to tilting strength $v_t$, which undergoes a sign change when modulating $v_t$ into $-v_t$ [Fig. 3(a)]. As restricted by TRS, NLNE current vanishes $\{j_y = 0\}$, see Fig. 3] in the absence of band tilt ($v_t = 0$). Yet, nonzero NLNE current immediately emerges when band tilt is finite. Interestingly, it rapidly increases by enhancing the warping effect and/or band tilting effect, and modulating the Fermi energy can also effectively tune the NLNE. These prominent tunabilities of NLNE are consistent to the NLSNE.

The two opposite spin components can be extracted from the NLNE current, respectively as $j_y^{s_1} = \frac{1}{2} (j_y + \frac{e}{\hbar} j_x)$, and $j_y^{s_2} = \frac{1}{2} (j_y - \frac{e}{\hbar} j_x)$, where the coefficient $2e/h$ is incorporated to match the dimension. The above spin-up $j_y^{s_1}$ and spin-down $j_y^{s_2}$ component of NLNE current are individually tunable by tilting strength, essentially differing from previous works [49, 50]. Such tun-
FIG. 3. The NLNE current \(|j_y/(\nabla_x T)^2|\) (a) as a function of Fermi energy \(E_f\) and band tilting strength \(v_t\) and (b) versus Fermi energy \(E_f\) at different tilting strength \(v_t\). \(\lambda = \lambda_0\) is fixed in (a) and the NLNE current is in units of \(nA \cdot \mu m/K^2\). The other parameters used here are same to Fig. 2.

ability is shown in Fig. 4(a), where \(j_y^{x+\uparrow}/\hbar\) at \(E_f = 0.6\ eV\) are plotted as a function of the tilt parameter \(v_t\). Interestingly, \(j_y^{x+\uparrow}\) and \(j_y^{x-\downarrow}\) get enhanced prominently when the band is tilted along \(y\)-direction \((v_f > 0)\) and negative \(y\)-direction \((v_f < 0)\), respectively. For a given band tilt, an apparent difference in the enhancement on the spin-up and spin-down components is observed. These significant anisotropies in band-tilt dependency (i.e., uneven enhancement on opposite spin) explicitly result in the spin polarized NLNE current (Fig. 1). When fixing \(E_f = 0.15\ eV\) and varying \(v_t/v_f\hbar\) within \([-0.5, 0.5]\), \(j_y^{x+\uparrow}\) is observed to be larger than \(j_y^{x-\downarrow}\) \([j_y^{x+\uparrow} > j_y^{x-\downarrow}]\), inset of Fig. 4(a), which leads to a negative NLSNE consistent with Fig. 2(c). Specifically, in the case of \(j_y^{x+\uparrow} = -j_y^{x-\downarrow}\), a pure nonlinear spin current is generated.

Generally, the SP \(\eta(|\eta| \leq 1)\) is defined as the ratio of the subtraction and the addition of \(j_y^{x+\uparrow}\) and \(j_y^{x-\downarrow}\), namely \(\eta = 2e j_y^{x-\downarrow}/|j_y|\) in our case. As shown in Fig. 4(b), the white region with \([2e j_y^{x-\downarrow}/|j_y| > |j_y|]\), where NLSNE is dominant over NLNE, shall be considered as the spin current regime with an ill-defined SP \(\eta\). The previously proposed nonlinear planar effects [49, 50], which engender from spin-to-charge conversion under magnetic field with conversion rate less than 100% (i.e., \(|j_y| < [2e j_y^{x-\downarrow}/|j_y|]\), exactly belong to such regime. Moreover, the longitudinal nonlinear currents, if generated in our scheme, also belongs to this regime, as the charge current is evidently smaller in magnitude than the spin current [59].

For the sake of practical applications, the charge current regime with \(|j_y| > [2e j_y^{x-\downarrow}/|j_y|]\) that corresponds to the colorful regions in Fig. 4(b), is more interesting to us. Note that, the condition for this regime also implies \(j_y^{x+\uparrow} j_y^{x-\downarrow} > 0\), namely the spin-up and spin-down components of NLNE are expected to flow in the same direction. This also determines a series of threshold strength of the band tilt [59] for generating SP. Remarkably, the SP exhibits considerably high tunabilities in terms of the band tilt and Fermi energy. It undergoes a sign change near the spin balance region that satisfies \(j_y^{x+\uparrow} = j_y^{x-\downarrow} \neq 0\) or \(\eta = 0\), when varying band tilting strength \(v_t\) with fixed Fermi energy or modulating Fermi energy with a selected \(v_t\) [Fig. 4(b)]. This interesting feature may promise the realization of sign-switchable SP, which is also significant for spintronics applications. Moreover, SP varies rapidly before the spin balance while gradually tends to be a constant after going through it [Fig. 4(c), (d)], offering a rapid and smooth tuning regime respectively. Such tunability (sensitiveness) difference is especially evident for the case of a fixed tilt but varying chemical potential [Fig. 4(d)]. In addition, it is also shown that NLNE current with near unity \(\sim 100\%\) SP can be generated in a wide range of Fermi energy and band tilting strength [see the dark blue and dark red colors in Fig. 4(b)].

The spin components involved in the nonzero spin-polarized NLNE always remain orthogonal to the charge and spin flows [see Eqs. (5, 6)], which enables the generation of perfect transverse SP with high performance. Since both the NLSNE and NLNE are nearly unchanged in the presence of a finite magnetic field [59], the SP generation in our scheme cannot be affected easily by the external magnetic field. Additionally, the impact of varying temperature on our proposed spin-polarized NLNE is found to be negligible (see SM [59]), consistent with the nonlinear planar Nernst effect studied in Ref. [49]. Thus our scheme can also achieve robust room-temperature SP, being of essential importance in the spintronics applications.
Discussion and conclusion— The proposed spin-polarized NLNE in this work can be ascribed to the joint effect of SML, hexagonal warping, and band tilting effect, where the former two ingredients are quite generic in realistic materials. The tilt of Dirac cone can be also realized in various materials and manifests interesting effects on the low-energy behaviors of Dirac fermions [57, 58, 70–76]. For TSSs in TIs, the finite band tilt is symmetry-allowed (or naturally exist) when TRS and $C_{3v}$ are both broken, as pointed out earlier in this work. Additionally, the magnetic topological semimetals that hold tilted Dirac cones, provided IS is further broken, can also support nonzero spin-polarized NLNE in principle.

The spin-polarized NLNE can be quantitatively estimated via the charge current $I = j_y \times l$ with $l$ the length of the material. Using the model parameters fitted for the TSSs of realistic material $\text{Bi}_2\text{Te}_3$ [51, 77–83], estimations of $I \sim 0.03 \mu \text{A}, \eta \sim 0.30$ can be obtained for a film with $l \sim 100 \mu\text{m}$ [50], when Fermi energy $E_f = 0.49 \text{eV}$, in-plane thermal gradient $\nabla^2 T \sim -1.3 K\mu\text{m}^{-1}$ [84] and a moderate band tilt $v_t/v_f \hbar = 0.2$ are considered. Remarkably, the higher SP and/or charge current magnitude can be realized by tuning different parameters. For instance, a much higher SP $\eta \sim 0.965$ carried by the NLNE can be achieved at relatively lower Fermi energy $E_f = 0.21 \text{eV}$, and when the strength of the band tilt is further increased e.g., $v_t/v_f \hbar = 0.45$, a spin-polarized current $I$ with magnitude around $0.12 \mu\text{A}$ can be realized. The above estimations are of the similar order of the magnitude as that made in the recent work [49], which are accessible in relevant experiment.

Though the top and bottom surfaces contribute oppositely to the surface transport effects, it is general that one of the surfaces is dominant as demonstrated in previous studies [54, 85–88], rendering the total responses to be nonzero. Therefore, our model analysis involving only the top surface state is sufficient to capture the essential feature of the proposed spin-polarized nonlinear effect. It should be mentioned that the introduced mechanism to generate SP by our work can be generalized to other transport effects (e.g., the nonlinear Hall effect and thermal Hall effect) in other potential materials e.g., the trigonally warped graphene and bilayer graphene [89–91], etc. Besides, to better reveal the tilt-induced spin-polarized NLNE, a more detailed analysis of the scattering processes that especially incorporate magnetic disorders will be necessarily important. It is also possible in principle to realize the alternating (AC) or second-harmonic-type SP through our mechanism, provided the external driving force (thermal gradient or electric field) is time (frequency)-dependent. These interesting topics beyond the scope of the current work are all left for future studies.

In conclusion, we successfully demonstrate a nonlinear transport effect that can remarkably realize the high-degree (near-unity) SP with flexible parameter tunabilities as well as robustness against temperature, which thereby hints significant advance and possible applications in spintronics.

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