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| **Author(s)** | Yang, H.-Y.; Huang, Chunli; Ochoa, H.; Cazalilla, M. A. |
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Extrinsic spin Hall effect from anisotropic Rashba spin-orbit coupling in graphene

H.-Y. Yang,1 Chunli Huang,2,1 H. Ochoa,3 and M. A. Cazalilla4,5
1Department of Physics, National Tsing Hua University, Hsinchu 30013, Taiwan, Republic of China
2Division of Physics and Applied Physics, School of Physical and Mathematical Sciences, Nanyang Technological University, Singapore 637371
3Fundación IMDEA Nanociencia, Cantoblanco 28049, Madrid, Spain
4Department of Physics and National Center for Theoretical Sciences (NCTS), National Tsing Hua University, Hsinchu City, Taiwan 300, Republic of China
5Donostia International Physics Center (DIPC), Manuel de Lardizabal, 4. E-20018 San Sebastian, Spain

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We study the effect of anisotropy of the Rashba coupling on the extrinsic spin Hall effect due to spin-orbit active adatoms on graphene. In addition to the intrinsic spin-orbit coupling, a generalized anisotropic Rashba coupling arising from the breakdown of both mirror and hexagonal symmetries of pristine graphene is considered. We find that Rashba anisotropy can strongly modify the dependence of the spin Hall angle on carrier concentration. Our model provides a simple and general description of the skew scattering mechanism due to the spin-orbit coupling that is induced by proximity to large adatom clusters.

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I. INTRODUCTION

The spin Hall effect (SHE) [1–5] has been investigated intensively over the past few decades due to its potential applications in spintronics [5,6]. Generally speaking, the microscopic mechanisms of the SHE can be classified into either intrinsic [7,8] or extrinsic [9,10]. In both cases, the existence of spin-orbit coupling (SOC) in the material or heterostructure is required. The intrinsic mechanism is a consequence of the band structure of the material, whereas the extrinsic mechanism stems from scattering of the charge carriers by impurities that locally induce SOC.

Since SOC is a relativistic effect that is typically strongest in materials containing heavy atoms, the SOC in graphene [11,12] is expected to be weak [13–15]. Therefore, graphene has been suggested as an ideal material for passive spintronics, for which a long spin diffusion length is required, and SOC in the material is a major limiting factor [16].

However, motivated by the search of materials exhibiting the quantum spin Hall effect [17], it has been theoretically predicted [18,19], and experimentally observed [20], that SOC can be greatly enhanced in graphene by means of adatom deposition. In the limit of a dilute number of impurities (i.e., adatoms), in which the excellent charge-carrier mobility properties of graphene are not strongly modified, heavy adatom clusters have been predicted to induce a sizable SHE [10]. Experimentally, a large spin Hall angle (∼0.2) has been reported by Balakrishnan and co-workers [20] in devices made from chemical-vapor-deposited (CVD) graphene. The phenomenon was explained [20] by the combination of resonant scattering and skew scattering by residual Cu clusters resulting from the CVD process. It was experimentally estimated that the latter can induce SOC of the order of ∼10 meV.

Nevertheless, the nature of the SOC induced by the adatoms depends on their arrangement relative to a hexagonal unit cell of graphene. The latter can lower the symmetry from the hexagonal symmetry of the carbon monolayer. It also depends on the symmetry of the orbitals that hybridize with the π bands of graphene, since this hybridization is ultimately responsible for both the proximity-induced SOC and the resonant scattering. This also applies to heavy-metal substrates or large clusters, where certain crystalline ordering is possible. In this regard, it is worth mentioning, for instance, the differences between gold intercalation [21], which leads to a conventional Rashba splitting of graphene bands, and lead [22], which results in proximity-induced Rashba SOC where the two terms of the coupling have different weight. The latter is a consequence of the reduced orthorhombic symmetry of the composite (graphene + substrate) system. Such coupling is therefore an anisotropic generalization of the Rashba SOC, which arises due to the breakdown of both mirror and sixfold rotation symmetry. Similar features have been reported recently in graphene intercalated with platinum [23].

In this work, we will investigate the skew scattering mechanism arising from the SOC induced by extrinsic scatterers. Unlike previous studies [10,20], we will focus on understanding the effects of Rashba anisotropy on the charge and especially spin transport properties, and in particular the spin Hall angle. Toward that end, we will first solve the scattering problem of an anisotropic SOC-active scatterer [24,25]. From the single-impurity scattering data, we will derive the relaxation times that parametrize the collision integral of the linearized Boltzmann transport equation (BTE) [26], which allows us to compute the spin Hall angle. Finally, we will also compare the interplay and interference between different scattering potentials.

The large SOC induced by absorbates (e.g., the ∼10 meV SOC reported in Ref. [20]) is the result of hybridization between the π orbital of the carbon atoms in the graphene layer and the orbitals of the adatoms [18,27,28]. This mechanism is fundamentally different from the one responsible for the existence of SOC in two-dimensional electron gas (2DEG). In the latter case, the orbital hybridization between electrons in a quantum well and the dopants is suppressed by the large distance between them [29]. As a result, the SOC induced by absorbates in graphene is not necessarily related to the gradient of the impurity scattering potential as it is in the case of 2DEG. This feature sets graphene apart, which added to its
other peculiarities, such as the absence of intrinsic SOC and its linear density of states, which allows for the formation of scattering resonances and makes this system unique for the study of the extrinsic SHE.

The rest of the article is organized as follows. We present the details of our theoretical model in Sec. II. First, we discuss the way the symmetries of monolayer graphene decorated with adatoms conform the form of the SOC in the $k \cdot p$ Hamiltonian of the system. Then, the single scatterer problem is solved. Using the scattering data (i.e., T-matrix) of the single scatterer problem, the linearized Boltzmann transport equation is also solved and the transport properties of the system are obtained. In Sec. III, we discuss the most salient features of our results, namely the change in the spin Hall angle and the conductivity of the system as a function of the anisotropy parameter. A summary of the main results of this work is provided in Sec. IV. Finally, the Appendices contain the most technical details of the work.

II. MODEL

A. Scattering potentials

In this study, we will consider a dilute ensemble of scatterers that create a (disorder) potential that is smooth in the atomic scale of graphene. As a consequence, we will neglect scattering between the two valleys at the opposite corners of the hexagonal Brillouin zone (i.e., $K_{\pm}$). Therefore, most of the discussion below applies to a single valley (i.e., $K_{+}$), unless otherwise stated.

To understand the charge and spin transport properties of the system, we will rely upon the semiclassical Boltzmann transport equation (BTE). The latter applies to doped graphene (i.e., when the Fermi energy measured from the Dirac point $E_F \neq 0$) in the limit where the distance between scatterers is much larger than the Fermi wavelength. Therefore, the results obtained from the BTE should be regarded as providing some sort of interpolation between the hole-doped Fermi liquid ($E_F \ll 0$) and the electron-doped Fermi liquid (i.e., $E_F \gg 0$) regime.

In the dilute impurity limit, the collision term of the BTE is determined by the scattering data for a single scatterer [30]. Thus, we first analyze the scattering problem of a single scatterer, for which the $k \cdot p$ Hamiltonian describing the electron dynamics in the long-wavelength limit can be generally written as follows:

$$\mathcal{H} = \hbar v_F \left( \pm \sigma_x p_x + \sigma_y p_y \right) + \sum_{a=0,1,R} V_a(r),$$  (1)

where the sign $\pm$ applies to the valley at crystal momentum $K_{\pm}$, and $\sigma_a$ ($a = x,y,z$) are the Pauli matrices associated with the sublattice degrees of freedom of the wave function. The Pauli matrices acting on electron spin are denoted by $\sigma_a$. The first two terms in Eq. (1) correspond to the $k \cdot p$ Hamiltonian of the pristine graphene, which describes the electronic bands near the $K_{\pm}$ points.

Among the possible time-reversal invariant impurity potentials, we will focus on the scalar potential ($\alpha = 0$), the intrinsic SOC ($\alpha = I$), and the Rashba potential ($\alpha = R$). These three are invariant under the point group $C_{6v}$, which is generated by the sixfold rotation axis perpendicular graphene intersecting the center of the hexagonal unit cell and six reflection planes containing such an axis. The latter group describes the rotation and mirror symmetries of monolayer graphene, excluding the mirror reflection about the graphene plane, which takes $z \to -z$. However, the scalar potential ($r = |r|$),

$$V_0(r) = v_0(r) f_{4,4},$$  (2)

and the intrinsic or Kane-Mele SOC term [17],

$$V_I(r) = \pm \Delta_I(r) \sigma_x \sigma_y,$$  (3)

are invariant under the larger point group $D_{6h}$, which includes the mirror reflection for which $z \to -z$. On the other hand, Rashba SOC is associated with a lack of mirror reflection symmetry. Typically, this symmetry is broken and lowered to $C_{6v}$ by the presence of a substrate, adatoms, and/or ripples.

The Rashba SOC is invariant under the $C_{6v}$ group of pristine graphene and takes the following form:

$$V_R(r) = \Delta_R(r)(\pm \sigma_z \sigma_y - \sigma_y \sigma_z).$$  (4)

However, in general, the planar symmetry $C_{6v}$ can be broken, for example due to the different symmetries of a graphene lattice and the substrate, or the arrangement (relative to the hexagonal unit cell of graphene) of the adatoms in a large cluster in proximity to the carbon layer. As a result, the symmetry can be lowered from hexagonal (i.e., $C_{6v}$) to rectangular (i.e., $C_{2v}$), for instance. This can be achieved by deposition or intercalation of a metal with either cubic or orthorhombic symmetry [22]. In Ref. [22], it was shown from symmetry arguments and first-principles calculations that the two terms of the Rashba SOC can acquire different weights, which leads to an anisotropic form of the Rashba SOC potential:

$$V_R(r) = \pm \Delta_I(r) \sigma_x \sigma_y - \Delta_2(r) \sigma_y \sigma_z.$$  (5)

In this work, we will study the effect of this anisotropy on the skew scattering mechanism and its contribution to the spin Hall effect.

Before turning our attention to the study of the scattering problem by such an anisotropic Rashba potential, it is useful to analyze the symmetries of (1) in the presence of the anisotropic Rashba SOC potential, Eq. (5). For reasons that will become clear below, it is convenient to write the anisotropic Rashba SOC as the sum of two terms, $V_R(r) = V_{SR}(r) + V_{NR}(r)$, where

$$V_{SR}(r) = \Delta_{SR}(r)(\sigma_x \sigma_y - \sigma_y \sigma_x),$$  (6)

$$V_{NR}(r) = \Delta_{NR}(r)(\sigma_y \sigma_z + \sigma_z \sigma_y),$$  (7)

with

$$\Delta_{NR/\text{SR}}(r) = \frac{\Delta_1(r) \pm \Delta_2(r)}{2},$$  (8)

where $\pm$ applies to $\Delta_{\text{SR}}(\Delta_{\text{SR}})$, respectively. For $\Delta_1 = \Delta_2$ we recover the standard Rashba SOC. In the opposite limit, $\Delta_1 = -\Delta_2$, a SOC to be termed “nonstandard” Rashba is obtained. This representation enables us to display more clearly how the anisotropy in the Rashba SOC violates the conservation of the angular momentum projected onto the $z$
axis. Let us recall the definition of the $z$ component of angular momentum operator:

$$J_z = l_z + \frac{\sigma_z}{2} + \frac{s_z}{2}.$$  

(9)

Notice that since $\Delta_y(r)$ are functions of $r = |r|$, as we have assumed, the scalar potential, intrinsic, and standard Rashba SOC commute with $J_z$. However, when the Rashba SOC is anisotropic, $J_z$ is no longer conserved and the culprit for this violation is the nonstandard Rashba SOC introduced above. Nevertheless, in the special case in which $\Delta_1 = -\Delta_2$ (i.e., $\Delta_{SR} = 0$), the quantity

$$M_z = l_z + \sigma_z - \frac{s_z}{2}$$

(10)

is conserved instead of $J_z$. This will become useful in our investigation of this special limit for a more general type of scatterers than those considered in the following (see Appendix C 1).

Note that the lack of conservation of $J_z$ by the anisotropic Rashba SOC makes it impossible to employ a partial wave expansion to solve the scattering problem as was done in Ref. [10]. Nonetheless, since we are interested in the scattering by clusters of adatoms of characteristic size $R > a$ ($a = 2.46$ $\text{Å}$ being the interatomic distance in graphene), and for typical experimental parameters in doped graphene, $R \ll k_F^{-1}$ (where $k_F$ is the Fermi wave vector), we will approximate the cluster potentials by Dirac $\delta$ functions, i.e.,

$$v_0(r) = \lambda_0 \delta^{(2)}(r),$$

(11)

$$\Delta_1(r) = \lambda_1 \delta^{(2)}(r),$$

(12)

$$\Delta_2(r) = \lambda_2 \delta^{(2)}(r).$$

(13)

Let us also define $v_0 = \lambda_0 / R^2$, $\Delta_1 = \lambda_1 / R^2$, and $\Delta_{1,2} = \lambda_{1,2} / R^2$ as the strength of the potentials in units of energy. In passing, we also note that a similar model (with only intrinsic SOC) was successfully employed to account for the giant SHE observed in CVD graphene and attributed to the SOC induced by residual Cu atom clusters [20]. Thus, the potentials in Eq. (1) take the form

$$V_0(r) = \lambda_0 \Delta_0 \delta^{(2)}(r), \quad \alpha = \{0, I, SR, NR\},$$

(15)

where $\lambda_\alpha$ is the strength of the potential and $\Delta_\alpha$ are $4 \times 4$ matrices acting upon the sublattice-spin degrees of freedom. Explicitly, the $\Lambda$ matrices are

$$\Lambda_0 = I_{4 \times 4}, \quad \Lambda_1 = \sigma_z s_z,$$

(16)

$$\Lambda_{SR} = \sigma_y s_y - \sigma_x s_x = i(\sigma^- s^+ - \sigma^+ s^-),$$

(17)

$$\Lambda_{NR} = \sigma_y s_y + \sigma_x s_x = i(\sigma^- s^+ - \sigma^+ s^-).$$

(18)

When written in terms of $\sigma^\pm$ and $s^\pm$, the conservation of $J_z$ by $V_{SR}$ and the failure to do so by $V_{NR}$ becomes apparent.

Interestingly, the $\Lambda$ matrices form a closed group under (matrix) multiplication. This means that the product of two of these matrices can be written as a linear combination of

$$\Lambda_i \Lambda_j = \sum c_{ijl} \Lambda_l,$$  

(19)

The coefficients $c_{ijl}$ can be read off from Table I. As a mathematical curiosity, it is worth noting that the group is Abelian, as can be expected for a group of order 4. Out of the two possible order-4 groups, this corresponds to the Klein group.

### B. Single scatterer problem

In this subsection, the Lippmann-Schwinger (LS) equation for the single impurity problem with the choice of potentials discussed in the previous section will be solved. The LS wave equation reads

$$\langle r | \psi_p \rangle = \langle r | \phi_{k,\sigma} \rangle + \int d^2r' \mathcal{G}_R(r - r') \langle r' | \mathcal{V}(r') | \psi_p \rangle,$$  

(20)

where

$$\mathcal{V}(r) = \sum_{\alpha = 0, I, SR, NR} V_\alpha(r),$$

(21)

$$\langle r | \phi_{k,\sigma} \rangle = \frac{1}{\sqrt{2}} \left( \frac{1}{e^{i\eta_{\tau}}} \right) e^{i k \cdot \eta_{\tau}},$$

(22)

and $V_\alpha(r)$ given by Eq. (15). In the LS equation, $\langle r | \phi_{k,\sigma} \rangle$ is the incident wave function from a conduction band with momentum $p$ and a spin state described by the spinor $\eta_{\sigma}$, where $\eta_{\tau} = \tau (1, 0)^T$. The spin quantization axis is taken to be the $z$ axis, which is perpendicular to the graphene plane. The angle of incidence is $\theta_k = \tan^{-1}(k_y / k_x)$; the normalization area of the system is taken to be unity. The scattered wave function is $\langle r | \psi_p \rangle$. Note that $\langle r | \psi_p \rangle$ does not carry a spin index because it is not an eigenstate of $s_z$ in general. The (retarded) Green’s function $\mathcal{G}_R(r - r')$ is a $4 \times 4$ matrix acting both in the sublattice pseudospin and electron-spin space (see Appendix A for details).

The Dirac $\delta$ function potential allows us to express the solution to the LS equation in terms of the $\Lambda$ matrices,

$$\langle r | \psi_p \rangle = \langle r | \phi_{k,\sigma} \rangle + \mathcal{G}_R(r) \sum_i \lambda_i \Lambda_i \langle 0 | \psi_p \rangle,$$

(23)

where $\lambda_i$ are the couplings that appear when we solve for $\langle 0 | \psi_p \rangle$.
(see Appendix A for details). In particular, it can be seen that for $\lambda_{NR} \to 0$, $\beta_{NR} \to 0$, and it then follows that the scattered wave function is an eigenstate of $J_z$, because the expression for the scattered wave no longer contains $\Lambda_{NR}$. On the other hand, when $\lambda_{SR} \to 0$, then $\beta_{SR} \to 0$, and the scattered wave function becomes an eigenstate of $M_z$ [cf. Eq. (10)].

Using Eq. (19) and introducing the coefficients

$$y_1 = \sum_{i,j} c_{ij} \lambda_i \beta_j,$$

(24)

the solution to the LS equation (20) takes the following compact form:

$$(r|\psi_p) = (r|\phi_{k,\sigma}) + G_R(r) \sum_{l} y_1 \Lambda_l(0|\phi_{k,\sigma}).$$

(25)

Note that the right-hand side of the above expression contains only known quantities. After expanding the Green function asymptotically at large distances, the scattered wave can be written as the sum of an incident and an outgoing wave:

$$(r|\psi_p) \approx (r|\phi_{k,\sigma}) + \sum_{\sigma = \uparrow, \downarrow} f(\mathbf{p}, \sigma'; \mathbf{k}, \sigma) \frac{e^{i k r}}{\sqrt{2 \pi}} \left(\frac{1}{\gamma} \right) \eta_{\sigma'}. $$

(26)

In the above expression, we have introduced the scattering amplitudes given by $f(\mathbf{k}, \sigma; \mathbf{p}, \uparrow)$ and $f(\mathbf{k}, \sigma; \mathbf{p}, \downarrow)$. From them, the differential scattering cross section can be calculated using

$$\frac{d\sigma}{d\theta} = \sum_{\sigma = \uparrow, \downarrow} \left| f(\mathbf{p}, \sigma'; \mathbf{k}, \sigma) \right|^2
= \sum_{\sigma = \uparrow, \downarrow} |f_{\sigma' \sigma}(\theta)|^2,$$

(27)

(28)

where $\theta = \cos^{-1}(k_F \mathbf{p})$ is the scattering angle. We refer the reader to Appendix A for the detailed form of the scattering amplitude and how it is related with the scattering $\Gamma$-matrix that enters in the collision term of the Boltzmann transport equation.

C. Transport properties

To compute the charge and spin transport properties of a dilute random ensemble of identical clusters of areal density $n_{imp}$, we use the semiclassical BTE [10,26]. The details of its linearization in the approximated notation are reviewed in Appendix B. The exact solution of this equation [10] allows us to obtain the charge and spin Hall conductivities ($\sigma_{tr}$ and $\sigma_{SH}$, respectively):

$$\sigma_{tr} = \frac{e^2}{h} \int d\epsilon \left| \frac{\partial n}{\partial \epsilon} \right| \frac{\tau_{sk} \tau_{sk}^* \tau_{tr}}{\tau_{sk} \tau_{sk}^* + \tau_{tr} \tau_{tr}},$$

(29)

$$\sigma_{SH} = -\frac{e^2}{h} \int d\epsilon \left| \frac{\partial n}{\partial \epsilon} \right| \frac{\tau_{sk} \tau_{sk}^* \tau_{tr}}{\tau_{sk} \tau_{sk}^* + \tau_{tr} \tau_{tr}},$$

(30)

where $n(\epsilon) = (e^{(\epsilon - \mu)/k_B T} + 1)^{-1}$ is the Fermi-Dirac distribution. The different scattering times—$\tau_{tr}$, $\tau_{sk}$, $\tau_{sk}^*$, and $\tau_{tr}^*$—are defined in Appendix B and can be derived from the differential scattering cross section. In particular, we would like to point out that

$$\frac{1}{\tau_{tr}^*} = n_{imp} v_F \sum_{\sigma = \uparrow, \downarrow} \int \left(1 - \eta_{\sigma' \sigma} \cos \theta \right) |f_{\sigma' \sigma}(\theta)|^2 d\theta
\equiv n_{imp} v_F \Sigma_{\sigma^*}.$$

(31)

where $n_{imp}$ and $v_F$ are the areal density of the impurities and the Fermi velocity of pristine graphene, respectively. Here, the notation $\eta_{\uparrow, \downarrow} = \eta_{\downarrow, \uparrow} = -1$ and $\eta_{\uparrow, \uparrow} = \eta_{\downarrow, \downarrow} = +1$. $\Sigma_{\sigma^*}$ is the single-scatterer transport cross section, which exhibits sharp peaks at the resonance energies of the single scatterer.

The figure of merit determining the efficiency in the charge current to spin current conversion, known as spin Hall “angle,” is defined as the ratio

$$\gamma = \frac{\sigma_{SH}}{\sigma_{tr}}.$$

(32)

At zero temperature, the spin Hall angle reduces to the ratio (see Appendix B)

$$\gamma = \frac{-\tau_{sk}^*}{\tau_{tr}}.$$

(33)

III. RESULTS AND DISCUSSION

The following discussion is about the effects of anisotropy in Rashba SOC. The degree of anisotropy in Rashba SOC will be phrased in terms of the anisotropy parameter:

$$\beta = \tan^{-1} \left( \frac{\Delta_2}{\Delta_1} \right) = \tan^{-1} \left( \frac{\lambda_2}{\lambda_1} \right).$$

(34)

Note that for the standard (“isotropic”) Rashba SOC, $\beta = \frac{\pi}{4}$. For $\beta = -\frac{\pi}{4}$, the Rashba-like SOC is of the form $\{\sigma_0 \sigma_z + \sigma_y \sigma_x\}$ (with $\tau = \pm$ for $K_{\pm}$), which has been termed nonstandard Rashba in Sec. II A. In the range $-\frac{\pi}{4} < \beta < \frac{\pi}{4}$, $\beta$ is a measure of the degree to which the $C_{6v}$ symmetry is broken by the adatom arrangement within the clusters. Close to $\beta = \frac{\pi}{4}$, the deviation from Rashba and the perfect $C_{6v}$ symmetric situation is small. On the other hand, having $-\frac{\pi}{4} < \beta < 0$ requires a strong breaking of the $C_{6v}$ symmetry. The latter depends on several factors, such as the chemical nature of the adatoms or their arrangement with respect to the hexagonal lattice of graphene. In the particular case of lead intercalations that motivated this work, the experiments suggest that $\beta$ can take very small values [22].

In Fig. 1, we show the dependence of the spin Hall angle $\gamma$ and the transport cross section $\Sigma_{\sigma^*}$ at zero temperature on the carrier energy $E$ for different values of strength of the scalar potential $V_0 = \lambda_0 / R^2$ [cf. Eq. (11)] for a rather anisotropic Rashba-like SOC corresponding to $\beta = -\pi / 8$. It can be seen that the enhancement of the spin Hall angle still takes place around the values of $E$ for which $\Sigma_{\sigma^*}(E)$ exhibits a peak, that is, a scattering resonance. This is in agreement with what was already pointed out in Ref. [10] for the isotropic Rashba SOC. Physically this is also expected, because at resonance the scattering electron or hole spends most of its time near the scatterer, and therefore it can also experience the effect of the locally induced SOC. The enhancement of $\gamma$ is suppressed at large values of $V_0$. To understand this effect qualitatively, let us recall that $\gamma$ and $\Sigma_{\sigma^*}$ are both determined by the T-matrix.
which obeys the LS equation

$$T(E) = V + V G_R(E) T(E), \quad (35)$$

where $G_R(E)$ is the retarded Green’s function and $V = V_0 + V_{\text{SOC}}$, where $V_0 \propto v_0$ is the scalar potential and $V_{\text{SOC}} = V_I + V_R$ is the SOC part of the potential. In the limit where $V_0 \gg V_{\text{SOC}}$, the solution to Eq. (35) can be (loosely) written as

$$T(E) = \frac{V_0 + V_{\text{SOC}}}{1 - (V_0 + V_{\text{SOC}}) G_R(E)} \approx -\frac{1}{G_R(E)} \left[ 1 + \frac{V_{\text{SOC}}}{V_0} \right], \quad (36)$$

where the last expression applies to the large-$V_0$ limit. Thus, to leading order, the cross section $\Sigma^*_{\text{tr}}$ is determined by the first term on the right-hand side, whereas $\gamma$ is determined by the second term. Hence, $\gamma$ is expected to decrease at large $V_0 \propto v_0$, as shown in Fig. 1.

For a given set of $v_0, \Delta_I$, and $\Delta_{\text{SR}}$, Fig. 2 shows the behavior of $\gamma$ and $\Sigma^*_{\text{tr}}$ as a function of the incident electron energy $E$ at different values of anisotropy parameter $\beta$. For the values of $\beta$ close to those corresponding to the nonstandard Rashba SOC (i.e., for $\beta \approx -\frac{\pi}{2}$), the energy dependence is strongly modified. On the other hand, for the case of a $\delta$-function potential, the anisotropy has a less pronounced effect on the cross section $\Sigma^*_{\text{tr}}$.

The observations made above remain largely unchanged when the effect of finite temperature is taken into account; see Fig. 3. As shown there, thermal fluctuations and the associated smearing of the Fermi distribution smooth out the sharper features of the (Fermi) energy dependence $E_F$ of $\gamma$ found at $T = 0$ and suppress the magnitude of $\gamma$. This can be seen in the left panel in Fig. 3 for the case of pure (i.e., $\Delta_I = 0$) anisotropic Rashba SOC and on the right for $\Delta_I \neq 0$. The plots in the left panel also illustrate that for $\beta = 0$ (i.e., $\Delta_2 = 0$), the spin current as well as the spin Hall angle vanish (cf. the second plot from the bottom on the left). This is because the quantization axis for the spin current is aligned along the $z$ axis, whereas for $\beta = 0$, $s_z$ commutes with the Hamiltonian. As pointed out above for $T = 0$, the energy dependence (relative to the isotropic case) is most strongly affected as $\beta$ approaches 085418-5.
FIG. 3. (a)–(d) Temperature dependence of $\gamma$ vs $E_F$ plot at $\Delta_I = 0$ meV and four different $\beta$. (e)–(h) Temperature dependence of $\gamma$ vs $E_F$ plot at $\Delta_I = 15$ meV and four different $\beta$. $v_0 = 0.1$ eV in all eight plots. Other parameters are the same as in Figs. 1 and 2. (Especially, $\frac{\Delta_I + \Delta_2}{2} = \frac{\Delta_{IR}}{2} = 15$ meV). Note that, as mentioned in the Introduction, our semiclassical approach is not accurate near the $E_F \approx 0$ regime.

We have analyzed a simple model in order to understand the effects of the anisotropy of proximity-induced Rashba spin-orbit coupling (SOC) on the spin Hall effect. The anisotropy arises as a consequence of the arrangement of adatoms in the clusters decorating a single layer of graphene, and it takes the form

$$V_R = \Delta_1(r)\sigma_z s_y - \Delta_2(r)\sigma_y s_z. \quad (37)$$

On symmetry grounds, such SOC is effectively generated when the arrangement lowers the symmetry of the system from the hexagonal symmetry (i.e., the $C_{6v}$ group) of graphene.

From our analysis, we conclude that the anisotropy in the Rashba SOC does not modify the observation that the spin Hall angle in graphene is enhanced by the scattering resonances [10] that appear near the Dirac point. In addition, the dependence on the carrier concentration (or equivalently the Fermi energy) of the spin Hall angle is also not strongly modified for weak anisotropy. However, when the parameter $\beta = \tan^{-1}(\Delta_2/\Delta_1) < 0$, and especially when $\beta$ approaches $-\pi/4$, we have found that the Fermi energy dependence deviates strongly from that observed in the isotropic case (corresponding to $\Delta_1 = \Delta_2$ or $\beta = \pi/4$). This conclusion is robust against finite-temperature effects, which smooths out somewhat the Fermi energy dependence and suppresses the value of $\gamma$. It is also not modified by relaxing our assumption of zero-range (i.e., Dirac-$\delta$) potentials.

In our study, we have assumed a single type of scatterer. In a realistic experiment (such as the one reported in Ref. [20]), several kinds of scatterers may be present, some of which do not induce SOC. However, we expect that the above qualitative features will remain unchanged. Experimentally, it would be
interesting to study the differences in the Fermi energy (i.e., doping) dependence of the spin Hall angle for clusters of different atomic species, which can lead to different anisotropic Rashba couplings. Indeed, experimental evidence for intercalated Pb islands obtained in Ref. [22] seems to indicate that this metal can induce a rather anisotropic Rashba coupling with $\Delta_2 \gg \Delta_1$. Similar deviations from the standard Rashba splitting of graphene bands have been reported recently in platinum intercalated devices [23]. Our study identifies the signatures of such deviations in the carriers’ skew scattering properties, providing a way to probe different spin textures in transport.

Lastly, we would like to point out that the skew scattering mechanism on spin transport has also been studied in connection with the extrinsic sources of the anomalous Hall effect [31]. However, in such systems, unlike graphene, there is no pseudospin related to the sublattice degree of freedom. Therefore, in graphene we expect an interesting interplay between the electron spin and the pseudospin degrees of freedom.

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APPENDIX A: SINGLE SCATTERER PROBLEM

In this Appendix, we will provide the details of the solution of the Lippmann-Schwinger (LS) equation. Toward that end, we first recall the form of the retarded Green’s function in the continuum limit of single-layer graphene, $G_R(r-r') = G_R(r-r') \otimes \mathbf{1}$, where $\mathbf{1} = \sum_{\sigma=\uparrow,\downarrow} \eta_\sigma \eta_\sigma$ is the identity matrix in spin space. The function $G_R(r-r')$ is given by

$$G_R(r, r') = \left[ r \right] \frac{1}{E + i0^+ - H_0}|r'|. \tag{A1}$$

Hence,

$$G_R(r, r') = \left( E - i\hbar v_F \sigma \cdot \mathbf{V}_r \right) \times \int \frac{d^2k}{(2\pi \hbar v_F)} \frac{e^{ik(r-r')}}{(E \hbar v_F + i0^+)^2 - |k|^2}. \tag{A2}$$

The integral in the above expression is the Green’s function for the two-dimensional Helmholtz equation, which reads

$$G^H_R(r-r') = -\frac{i}{4\hbar^2 v_F} H_0^{(1)} \left( \frac{E|r-r'|}{\hbar v_F} \right), \tag{A3}$$

where $H_0^{(1)}$ is the Hankel function of the first kind. Inserting this result in the expression for $G_R(r, r')$ and using $\frac{\partial}{\partial z} H_0^{(1)}(z) = \frac{\pi i}{z} H_0^{(1)}(z) - H_1^{(1)}(z)$ yields

$$G_R(r, r') = \frac{-i|E|}{4\hbar^2 v_F} \left[ \text{sgn}(E) H_0^{(1)} \left( \frac{E|r-r'|}{\hbar v_F} \right) + i\sigma_0 H_1^{(1)} \left( \frac{E|r-r'|}{\hbar v_F} \right) \right], \tag{A4}$$

where

$$\sigma_0 = \begin{pmatrix} 0 & e^{-i\theta} \\ e^{i\theta} & 0 \end{pmatrix}. \tag{A5}$$

Due to translational invariance, the Green’s function is only a function of the difference in position. Here $\theta$, with $\cos \theta = (\mathbf{r} - \mathbf{r'}) \cdot \mathbf{\hat{x}}$, is the angle between the vector $\mathbf{r} - \mathbf{r'}$ and the $x$ axis. Note that we have chosen the retarded Green’s function for both electrons (i.e., $E > 0$) and holes (i.e., $E < 0$). Setting $|E| = \hbar v_F k$, we arrive at

$$G_R(r, r') = \begin{cases} \frac{-i k}{4\hbar v_F} \left[ H_0^{(1)}(k|r-r'|) + i\sigma_0 H_1^{(1)}(k|r-r'|) \right] & \text{for } E > 0, \\ \frac{i k}{4\hbar v_F} \left[ -H_0^{(2)}(k|r-r'|) + i\sigma_0 H_1^{(2)}(k|r-r'|) \right] & \text{for } E < 0. \end{cases} \tag{A6}$$

where $\mathbf{p}$ is the momentum of the scattered wave, and $\cos \theta_p = \cos \theta = \frac{x}{\sqrt{x^2+y^2}} \mathbf{\hat{x}}$ is the angle subtended between the scattered momentum with the $x$ axis.

Accounting for the spin degree of freedom, the asymptotic form of the Green’s function reads

$$G_R(r, r') \approx \frac{-i k}{4\hbar v_F} \frac{2}{\pi k r} e^{ikr} \sum_\sigma \left( e^{i\theta_\sigma} \eta_\sigma \right) \times \eta_\sigma (1 e^{-i\theta_\sigma}) e^{-ip r}. \tag{A9}$$

Thus, we are equipped to solve the LS equation of the scattering problem. Assuming an incident electron from the conduction band with momentum $\mathbf{p}$ and $\sigma$ means that (we
work assuming a normalization area equal to unity)
\[
|\langle \mathbf{r} | \phi_{\mathbf{k},\sigma} \rangle |^2 = \frac{1}{\sqrt{2}} \left( \frac{1}{e^{i\theta_0}} \right) e^{i\mathbf{k} \cdot \mathbf{r}} \eta_{s}. \tag{A10}
\]

Thus, the LS equation [Eq. (20)] becomes
\[
|\langle \mathbf{r} | \psi_p \rangle |^2 = |\langle \mathbf{r} | \phi_{\mathbf{k},\sigma} \rangle |^2 + \sum_{\sigma' = \uparrow, \downarrow} \frac{-ik}{4\hbar v_F} \sqrt{\frac{2}{\pi k r}} e^{i(kr - \frac{\pi}{4})} \left( \frac{1}{e^{i\theta_0}} \right) \eta_{s'},
\]
\[
\times \int d^2 r' (1 - e^{-i\theta_0}) \eta_{s'} e^{-i\mathbf{p} \cdot \mathbf{r}'} \langle \mathbf{r}' | T | \phi_{\mathbf{k},\sigma} \rangle
\]
\[
= |\langle \mathbf{r} | \phi_{\mathbf{k},\sigma} \rangle |^2 + \sum_{\sigma' = \uparrow, \downarrow} f_{\mathbf{p},\sigma';\mathbf{k},\sigma} e^{i(kr - \frac{\pi}{4})} \left( \frac{1}{e^{i\theta_0}} \right) \eta_{s'}, \tag{A11}
\]

where we have introduced the T-matrix, which can be defined by the equation \( T(\phi_{\mathbf{k},\sigma}) = V \psi_p \). Note that the scattered wave does not carry a spin index because it is not an eigenstate of \( s_z \). Indeed, for a non-spin-conserving potential such as Rashba, the scattered wave is a combination of the incident wave with momentum \( \mathbf{k} \) and spin \( \sigma \) and a scattered radial spin-up (spin-down) wave with amplitude given by \( f_{\mathbf{k},\sigma';\mathbf{p},\uparrow} \) [\( f_{\mathbf{k},\sigma';\mathbf{p},\downarrow} \)].

From the above result, the scattering amplitude can be related to the T-matrix by the following expression:
\[
f_{\mathbf{p},\sigma';\mathbf{k},\sigma} = -ie^{-i\frac{\pi}{4}} \frac{\sqrt{k}}{2\pi} \langle \mathbf{p},\sigma' | T | \phi_{\mathbf{k},\sigma} \rangle \tag{A12}
\]

Recalling that the impurity potential is given by Eq. (15), the T-matrix can be written as follows:
\[
\langle \phi_{\mathbf{p},\sigma'} | T | \phi_{\mathbf{k},\sigma} \rangle = \int d^2 r \langle \phi_{\mathbf{p},\sigma'} | \mathbf{r} | V(\mathbf{r}) | \psi_p \rangle
\]
\[
= \sum_i \lambda_i \langle \phi_{\mathbf{p},\sigma'} | \mathbf{0} \rangle \Lambda_i | \mathbf{0} \rangle \psi_p \tag{A13}
\]
\[
= \sum_i \lambda_i \beta_j \langle \phi_{\mathbf{p},\sigma'} | \mathbf{0} \rangle \Lambda_j | \mathbf{0} \rangle \psi_p, \tag{A14}
\]

where we have used (see below) \( \langle \mathbf{0} | \psi_p \rangle = \sum_j \beta_j \Lambda_j | \mathbf{0} \rangle \psi_p \).

We have \( \Lambda_i \Lambda_j = \sum_i c_{ijl} \Lambda_l \), where the coefficient \( c_{ijl} \) can be read off from a group multiplication table (cf. Table I). In addition, let us define
\[
\gamma_i = \sum_{i,j} c_{ijl} \lambda_i \beta_j, \tag{A16}
\]

The T-matrix can be obtained as follows:
\[
\langle \phi_{\mathbf{p},\sigma'} | T | \phi_{\mathbf{k},\sigma} \rangle = \sum_i \gamma_i \langle \phi_{\mathbf{p},\sigma'} | \mathbf{0} \rangle \Lambda_i | \mathbf{0} \rangle \psi_p \tag{A17}
\]

In the above equations, it is understood that the indices \( i,j,l \) run over the set \( \{0,1,2\} \). Explicitly, the T-matrix is given by
\[
\langle \phi_{\mathbf{p},\sigma'} | T | \phi_{\mathbf{k},\sigma} \rangle = \gamma_0(k) \cos \frac{\theta}{2} \delta_{\sigma',\sigma} + i \gamma_1(k) \sin \frac{\theta}{2} (s_z)_{\sigma',\sigma} \tag{A18}
\]
\[
+ [\gamma_{SR}(k) + \gamma_{SR}(k)] \sin \frac{\phi}{2} (s_y)_{\sigma',\sigma} \tag{A18}
\]

For elastic scattering (i.e., \( |\mathbf{p}| = |\mathbf{k}| \)), the T-matrix (and hence the scattering amplitude) is a function not only of the scattering angle \( \theta = \theta_k - \theta_p \) but also \( \phi = \theta_k + \theta_p \). Here \( \theta_k = \arccos((\mathbf{k} \cdot \hat{x})/k) \) is the angle subtended by \( \mathbf{k} \) and the \( x \) axis. In this article, we use the convention where the incident momentum is aligned with the \( x \) axis (i.e., \( \theta_k = 0 \), hence the T-matrix and the scattering amplitude are functions of \( \theta \) only.

Upon setting \( \mathbf{r} = \mathbf{0} \) in Eq. (20),
\[
\langle \mathbf{0} | \psi_p \rangle = \langle \mathbf{0} | \phi_{\mathbf{k},\sigma} \rangle + \sum_j G_R(0) \lambda_j \Lambda_j \langle \mathbf{0} | \psi_p \rangle. \tag{A19}
\]

Hence,
\[
\langle \mathbf{0} | \psi_p \rangle = \frac{\sum_j G_R(0) \lambda_j \Lambda_j \langle \mathbf{0} | \psi_p \rangle}{1 - \sum_j G_R(0) \lambda_j \Lambda_j}. \tag{A20}
\]

The coefficients \( \beta_i \) are obtained by inverting the matrix \( 1 - \sum_j G_R(0) \lambda_j \Lambda_j \) and (exactly) projecting onto the basis of \( \Lambda \) matrices, which yields

\[
\beta_0 = \frac{1}{4} \sum_{\eta = \pm} \left( \frac{1}{1 + G_R(0)(\lambda_0 + 2\eta') \lambda_0 - \eta' \lambda_0} \right), \tag{A22}
\]

\[
\beta_1 = \frac{1}{4} \sum_{\eta = \pm} \left( \frac{\eta'}{1 + G_R(0)(\lambda_0 + 2\eta' \lambda_0 - \eta' \lambda_0)} \right), \tag{A23}
\]

\[
\beta_{SR} = \frac{1}{[1 + G_R(0)(\lambda_0 - 2\lambda_0 - \lambda_0)][1 + G_R(0)(\lambda_0 + 2\lambda_0 - \lambda_0)]}, \tag{A24}
\]

\[
\beta_{NR} = \frac{1}{[1 - G_R(0)(\lambda_0 - 2\lambda_0 + \lambda_0)][1 - G_R(0)(\lambda_0 + 2\lambda_0 + \lambda_0)]}. \tag{A25}
\]
In the above expressions, $G_R(0)$ is a scalar and $G_{R}(0) = G_R(0) \otimes I$ is the matrix Green’s function at the origin. The $G_R(0)$ is obtained by imposing a cutoff at high momenta. Setting $r = r' = 0$ in Eq. (A2), we have

$$G_R(0) = \int \frac{dk'}{2\pi \hbar v_F} k \frac{\text{sgn}(E)}{2\pi \hbar v_F} \log |kR| = \frac{ik}{4\hbar v_F}. \quad (A26)$$

The integral is cut off at momenta $k' \sim R^{-1}$, where $R$ is of the order of the actual spatial range of the scatterer potential.

**APPENDIX B: SOLUTION OF THE BOLTZMANN EQUATION**

In this Appendix, in order to make the article self-contained, we review the solution of the linearized Boltzmann transport equation (BTE) obtained in Ref. [10]. For an external dc electric field, the linearized BTE takes the form [26]

$$(e)E \cdot \nabla \partial_t n^0(\epsilon_k) = -\partial_t n_{\sigma}(k)_{\text{coll}}, \quad (B1)$$

where $E = E|\hat{\mathbf{k}}|$ is the applied electric field, $(e)\epsilon$ is the electron charge, $n^0(\epsilon) = (e(\epsilon - \mu)/\hbar k_T + 1)^{-1}$ is the equilibrium Fermi-Dirac distribution, and

$$v_k = \zeta v_F (\cos \phi(k), \sin \phi(k)) \quad (B2)$$

is the carrier velocity in graphene with the angle $\phi(k) = \arctan k_y/k_x$ (not to be confused with the free Hamiltonian eigenstate $|\phi_{\sigma, \theta}\rangle$); $\zeta = \pm 1$ is the band index (+1 for electrons, -1 for holes) and $n_{\sigma}(k)$ is the distribution function for electrons with spin projection $\sigma = \uparrow/\downarrow$ and Bloch wave vector $k$. The spin quantization axis is chosen to be the axis perpendicular to the graphene plane, which we take to be the $z$ axis. The term $\partial_t n_{\sigma}(k)_{\text{coll}}$ denotes the collision integral [10]

$$-\partial_t n_{\sigma}(k)_{\text{coll}} = \sum_{p, \sigma' = \uparrow, \downarrow} n_{\sigma}(k) - n_{\sigma}(p) W_{\sigma' \sigma}(p, k). \quad (B3)$$

$W_{\sigma' \sigma}(p, k)$ is the scattering rate [30] from state $(k, \sigma)$ to $(p, \sigma')$ due to the presence of impurities:

$$W_{\sigma' \sigma}(p, k) = \frac{2\pi n_{\text{imp}}}{\hbar} |\langle \phi_{\sigma'} | T(e_p) | \phi_{\sigma} \rangle|^2 \delta(\epsilon_k - \epsilon_p). \quad (B4)$$

Here $T(e)$ is the T-matrix that has been explicitly obtained in Appendix A, and $n_{\text{imp}}$ is the density of impurities. The linearized BTE can be solved exactly by the following ansatz for $\delta n_{\sigma}(k) = n_{\sigma}(k) - n^0(\epsilon_k)$:

$$\delta n_{\sigma}(k) = \zeta v_F \{ A_{\sigma} (k) \cos \phi(k) + B_{\sigma} (k) \sin \phi(k) \}. \quad (B5)$$

Introducing this ansatz in (B1) and setting $\phi(k) = 0$ and $\phi(k) = \frac{\pi}{2}$ for the longitudinal and transverse response, respectively, we obtain the following system of algebraic equations for $A_{\sigma}$ and $B_{\sigma}$:

$$\sum_{\sigma' = \uparrow, \downarrow} A_{\sigma} \Gamma^C_{\sigma' \sigma} + B_{\sigma} \Gamma^S_{\sigma' \sigma} - A_{\sigma} \Gamma^T_{\sigma' \sigma} = -X, \quad (B6)$$

$$\sum_{\sigma' = \uparrow, \downarrow} B_{\sigma} \Gamma^C_{\sigma' \sigma} - A_{\sigma} \Gamma^S_{\sigma' \sigma} - A_{\sigma} \Gamma^T_{\sigma' \sigma} = 0, \quad (B7)$$

where $X \equiv -e|E|\frac{2\hbar v_F}{\pi \hbar k_T}$ and the coefficients $\Gamma$ are defined as

$$\Gamma^T_{\sigma' \sigma} = \int \frac{d^2p}{(2\pi)^2} W_{\sigma' \sigma}(p, k), \quad (B8)$$

$$\Gamma^C_{\sigma' \sigma} = \int \frac{d^2p}{(2\pi)^2} \cos[\phi(p) - \phi(k)] W_{\sigma' \sigma}(p, k), \quad (B9)$$

$$\Gamma^S_{\sigma' \sigma} = \int \frac{d^2p}{(2\pi)^2} \sin[\phi(p) - \phi(k)] W_{\sigma' \sigma}(p, k), \quad (B10)$$

where $\phi(p) - \phi(k) = \theta$ is the scattering angle.

Note that time-reversal symmetry imposes several constraints on Eqs. (B6) and (B7). In particular, it requires that $\Gamma^T_{\sigma' \sigma} = \Gamma^T_{\sigma' \sigma}$, $\Gamma^C_{\sigma' \sigma} = \Gamma^C_{\sigma' \sigma}$, $\Gamma^S_{\sigma' \sigma} = -\Gamma^S_{\sigma' \sigma}$, $A_{\sigma} = A_{\bar{\sigma}}$, and $B_{\sigma} = -B_{\bar{\sigma}}$, where we further denote $\bar{\sigma}$ as the opposite spin of $\sigma$ to make the notation more compact. These relations are used to simplify the above system of equations. Using these coefficients $\Gamma$, Eqs. (B6) and (B7) take the following form in terms of four relaxation times:

$$A_{\sigma} = A_{\bar{\sigma}} = \frac{\tau_{sk} \tau_{sk}}{\tau_{sk} + \tau_{tr}} X, \quad (B11)$$

$$B_{\sigma} = -B_{\bar{\sigma}} = -\frac{\tau_{sk} \tau_{sk}}{\tau_{sk} + \tau_{tr}} X. \quad (B12)$$

The four relaxation times—$\tau_{tr}$, $\tau_{sk}$, $\tau_{tr}$, and $\tau_{sk}^*$—are defined as follows:

$$\frac{1}{\tau_{tr}} = \Gamma^T_{\sigma' \sigma} - \Gamma^C_{\sigma' \sigma} + \Gamma^f_{\sigma' \sigma} - \Gamma^C_{\sigma' \sigma}, \quad (B13)$$

$$\frac{1}{\tau_{sk}} = \Gamma^T_{\sigma' \sigma} - \Gamma^C_{\sigma' \sigma} + \Gamma^f_{\sigma' \sigma} + \Gamma^C_{\sigma' \sigma}, \quad (B14)$$

$$\frac{1}{\tau_{sk}} = \Gamma^C_{\sigma' \sigma} + \Gamma^S_{\sigma' \sigma}, \quad (B15)$$

$$\frac{1}{\tau_{sk}^*} = \Gamma^S_{\sigma' \sigma} - \Gamma^S_{\sigma' \sigma}. \quad (B16)$$

The physical meaning of the relaxation times becomes more transparent if we use Eq. (A12) to express the scattering rate $W_{\sigma' \sigma}(p, k)$ in terms of the scattering amplitude:

$$W_{\sigma' \sigma}(p, k) = \frac{4\pi^2 \hbar v_F^2 n_{\text{imp}}}{k} |f_{\sigma' \sigma}(\theta)|^2 \delta(\epsilon_k - \epsilon_p), \quad (B17)$$

where $\theta = \arccos \frac{k \cdot p}{k_F}$ is the scattering angle. Hence,

$$\frac{1}{\tau_{tr}} = n_{\text{imp}} v_F \sum_{\sigma' = \uparrow, \downarrow} \int (1 - \cos \theta)|f_{\sigma' \sigma}(\theta)|^2 d\theta \equiv n_{\text{imp}} v_F \Sigma_{tr}, \quad (B18)$$

$$\frac{1}{\tau_{sk}} = n_{\text{imp}} v_F \sum_{\sigma' = \uparrow, \downarrow} \int (1 - \eta_{\sigma' \sigma} \cos \theta)|f_{\sigma' \sigma}(\theta)|^2 d\theta \equiv n_{\text{imp}} v_F \Sigma_{tr}^*, \quad (B19)$$

$$\frac{1}{\tau_{sk}^*} = n_{\text{imp}} v_F \sum_{\sigma' = \uparrow, \downarrow} \int \sin \theta|f_{\sigma' \sigma}(\theta)|^2 d\theta \equiv n_{\text{imp}} v_F \Sigma_{sk}. \quad (B20)$$
\[ \frac{1}{\tau_{\text{tr}}} = \eta_{\alpha \sigma} v_F \sum_{\sigma' = \pm 1} \int \eta_{\alpha \sigma'} \sin \theta |f_{\alpha \sigma'}(\theta)|^2 d\theta \]

\[ = n_{\text{imp}} v_F \Sigma^*_1, \]  

where \(\Sigma_1, \Sigma^*_1, \Sigma_{3k},\) and \(\Sigma^*_{3k}\) are the single-scatterer scattering cross sections corresponding to the four different scattering rates, and

\[ \eta_{\alpha \sigma'} = \begin{cases} 
1 & \text{if } \sigma' = \sigma, \\
-1 & \text{if } \sigma' = \bar{\sigma}.
\end{cases} \]  

Having solved the linearized BTE, we are ready to compute the longitudinal current \(j_\alpha\) (assumed to be along the \(x\) axis) and spin Hall current \(j_{\text{SH}}\) (assumed to be along the \(y\) axis):

\[ j_\alpha = -e \int \frac{d^2k}{(2\pi)^2} [n_\sigma(k) + n_{\bar{\sigma}}(k)] \xi v_F \cos \phi(k) \]

\[ = -\frac{e^2}{2\pi^2} \hbar \int d\epsilon \epsilon A_\sigma X \]

\[ = -\frac{e}{2\pi^2} \hbar \int d\epsilon |e| \frac{\tau_{3k}^* \tau_{\alpha k}^* \tau_{\alpha k}}{\tau_{3k} \tau_{\alpha k} + \tau_{\alpha k} \tau_{\alpha k}^*} \left[ -e|E| \frac{\partial n^0(\epsilon_k)}{\partial \epsilon} \right] \]

\[ = e^2 \hbar \int d\epsilon |e| \frac{\partial n^0(\epsilon_k)}{\partial \epsilon} \left[ \frac{\tau_{3k}^* \tau_{\alpha k}^* \tau_{\alpha k}}{\tau_{3k} \tau_{\alpha k} + \tau_{\alpha k} \tau_{\alpha k}^*} |E| \right] \equiv \sigma_\alpha |E|, \]  

(B23)

\[ j_{\text{SH}} = -e \int \frac{d^2k}{(2\pi)^2} [n_\sigma(k) - n_{\bar{\sigma}}(k)] \xi v_F \sin \phi(k) \]

\[ = -\frac{e^2}{2\pi^2} \hbar \int d\epsilon \epsilon B_\sigma (k) X \]

\[ = -\frac{e}{2\pi^2} \hbar \int d\epsilon |e| \frac{\tau_{3k}^* \tau_{\alpha k}^* \tau_{\alpha k}}{\tau_{3k} \tau_{\alpha k} + \tau_{\alpha k} \tau_{\alpha k}^*} \left[ -e|E| \frac{\partial n^0(\epsilon_k)}{\partial \epsilon} \right] \]

\[ = e^2 \hbar \int d\epsilon |e| \frac{\partial n^0(\epsilon_k)}{\partial \epsilon} \left[ \frac{\tau_{3k}^* \tau_{\alpha k}^* \tau_{\alpha k}}{\tau_{3k} \tau_{\alpha k} + \tau_{\alpha k} \tau_{\alpha k}^*} |E| \right] \]

\[ = \sigma_{\text{SH}} \eta_{\alpha \sigma} |E|, \]  

(B24)

From the above expressions, we can read off the expressions for the longitudinal transport and spin Hall conductivities, Eqs. (29) and (30), respectively. The spin Hall angle measures the fraction of the charge current transformed into spin current, i.e.,

\[ \gamma = \frac{j_{\text{SH}}}{j_\alpha} = \frac{\sigma_{\text{SH}}}{\sigma_{\alpha}}. \]

In the zero-temperature limit, the spin Hall angle can be further simplified to yield the following expression:

\[ \gamma = \frac{\sigma_{\text{SH}}}{\sigma_{\alpha}} = \frac{\Sigma_{3k}^*}{\Sigma_{3k}}. \]  

(B26)

APPENDIX C: CIRCULAR SCATTERER

In this Appendix, we consider the extreme case for which the couplings in the Rashba potential have opposite sign, that is, \(\Delta_2/\Delta_1 = -1,\) which has been termed “nonstandard” Rashba in the main text. We show that, in this case, under the assumption of a circular (i.e., “pill-box” shaped) scatterer, a solution of the scattering problem in terms of partial wave expansions is still possible.

However, what makes “nonstandard” Rashba different from the “standard” Rashba SOC (i.e., \(\Delta_2/\Delta_1 = 1\)) is that the conserved quantity is no longer the total angular momentum projection along the \(z\) axis, \(J_z = l_z \pm \sigma_z/2 + s_z/2\) (where \(\pm\) applies to \(K_\pm,\) respectively), but \(M_z = l_z \pm \sigma_z/2 - s_z/2,\) with \(l_c \equiv x p_x - y p_y.\)

Below we consider in parallel the two cases, and we study the scattering solutions for the following potentials:

\[ V_{\text{SR}}(r) = [v_0 + \Delta_{\text{SR}}(\tau_\sigma \sigma_s \bar{\sigma} - \tau_\sigma \sigma_s \bar{\sigma})] \Theta(R - r), \]

\[ V_{\text{NR}}(r) = [v_0 + \Delta_{\text{NR}}(\tau_\sigma \sigma_s \bar{\sigma} + \tau_\sigma \sigma_s \bar{\sigma})] \Theta(R - r), \]

where \(R\) is the radius of the circular scatterer potential. Note that we have also considered a scalar potential \(v_0\) in addition to the standard Rashba/nonstandard Rashba-type spin-orbit coupling potential.

1. Nonstandard Rashba case

As mentioned in Sec. II A, the conserved quantity that plays the role of angular momentum when the Rashba coupling is of the nonstandard kind is \(M_z = l_z \pm \sigma_z/2 - s_z/2.\) Therefore, the scattering waves can be expanded on the basis of eigenstates of \(M_z,\)

\[ \psi_m(r, \theta) \sim \left( A_m(r) e^{im\theta} \right) \eta_\uparrow + \left( C_{m-1}(r) e^{i(m-1)\theta} \right) \eta_\downarrow, \]

(C3)

This wave function is an eigenstate of \(M_z,\) where \(M_z \psi_m(r, \theta) = m \hbar \psi_m(r, \theta).\) \(A, B, C,\) and \(D\) are the unknowns to be determined by solving the Schrödinger equation. The real-space angle \(\theta = \arctan z/r\) here should not be confused with the scattering angle defined in the previous Appendix.

For \(r > R,\) the Hamiltonian reduces to the kinetic term \(H_0 = v_F (\pm \sigma_x p_x + \sigma_y p_y).\) In this region, the wave function can be expanded as follows:

\[ \psi_m^\uparrow(r, \theta) = \left( J_m(kr) e^{im\theta} \right) \eta_\uparrow + t_{m+1}^{\uparrow} \left( H_m(kr) e^{i(m+1)\theta} \right) \eta_\uparrow \]

\[ + t_{m-1}^{\uparrow} \left( H_{m-1}(kr) e^{i(m-1)\theta} \right) \eta_\downarrow, \]

(C4)

where we have assumed that the incident electron is from the conduction band with spinor \(\eta_\uparrow,\) \(J_m(kr)\) is the Bessel function of the first kind and \(H_m(kr)\) is the Hankel function of the first kind. \(t_{m+1}^{\uparrow} (t_{m-1}^{\uparrow})\) is related to the non-spin-flip (spin-flip) partial wave scattering amplitude, which must be the case if there is no scattering potential.

For \(r < R,\) we have to solve for the Dirac equation

\[ (H_0 + V_{\text{NR}}) \psi_m^\uparrow(r, \theta) = E(k) \psi_m^\uparrow(r, \theta). \]

(C5)

The eigenvalue \(E(k)\) is obtained by diagonalizing the Schrödinger equation in momentum space,

\[ E(k) = v_0 + \xi \Delta_{\text{NR}} + k^2 \sqrt{\Delta_{\text{NR}}^2 + k^2}. \]

(C6)
The defined in Eq. (A12), the asymptotic form (\(\lambda\)) Hankel function, where \(\xi\) all the partial waves, \(q\xi\) scattering amplitudes, \(f\) wave, which contains a spin-up and a spin-down component. The first term in Eq. (C9) is the incident wave propagating \(\theta\) direction, and the second term is the scattered radial wave function \(\psi_{sc}\). The total scattered wave function \(\psi_{sc}\) \(\sim\) in the \(\pm\) direction, and \(\pm\) are two band indices. Only elastic scatterings are considered, therefore the scattered wave function must also be in the conduction band, i.e., \(\lambda = +1\). Substituting Eq. (C3) into Eq. (C5), we have to solve for a system of differential equations (on the basis of \(\{A^\uparrow, B^\uparrow, A^\downarrow, B^\downarrow\}\)):

\[
\begin{pmatrix}
-\epsilon_\xi & -i\left[\frac{d}{dr} + \frac{i}{r}(m + 1)\right] & 0 & -2i\Delta_{NR} \\
0 & -\epsilon_\xi & 0 & 0 \\
2i\Delta_{NR} & 0 & -\epsilon_\xi & -i\left[\frac{d}{dr} + \frac{i}{r}(m - 1)\right]
\end{pmatrix}
\begin{pmatrix}
A_m(r) \\
B_m(r) \\
A_{m+1}(r) \\
B_{m+1}(r)
\end{pmatrix}
= 0.
\]

Here \(\epsilon_\xi \equiv E(k) - V_0\). Let \(A_m(r) = J_m(qr)\), where \(q\) is some wave number to be determined. The system of differential equations can be solved as

\[
\psi_m^\sim(r, \theta) = \sum_{\xi=\pm1} \alpha_\xi \left[ \begin{pmatrix} J_m(qr) & iH_m(qr) \end{pmatrix} e^{im\theta} \right] \eta_\xi + \left[ \begin{pmatrix} iJ_{m+1}(qr) & iH_{m+1}(qr) \end{pmatrix} e^{im\theta} \right] \eta_\xi \]

where \(q \xi = \sqrt{\lambda^2 - 2\xi^2 \Delta_{NR}}\) and \(\alpha_\xi\) are some linear combination weights to be determined from boundary conditions. By equating the wave function at the boundary of the circular potential, \(\psi_m^\sim(R, \theta) = \psi_m^\sim(r, \theta)\), we can derive the partial wave scattering amplitudes, \(t_m^\uparrow\) and \(t_m^\downarrow\).

Finally, we will establish the relationship between partial wave amplitudes \(t_m\) defined in this section and the scattering amplitude \(f(p, \sigma^\uparrow, k, \sigma)\) defined in Appendix A. The total scattered wave function \(\psi(r, \theta)\) is obtained by taking a linear combination over all the partial waves, \(\psi(r, \theta) = \sum_m c_m \psi_m^\sim(r, \theta)\). Setting \(c_m = 1\) and summing Eq. (C4) over \(m\) yields

\[
\psi(r, \theta) = \sum_{m=-\infty}^{\infty} \sum_{\xi=\pm1} \left[ \begin{pmatrix} J_m(kr) & iH_m(kr) \end{pmatrix} e^{im\theta} \right] \eta_\xi + \left[ \begin{pmatrix} iJ_{m+1}(kr) & iH_{m+1}(kr) \end{pmatrix} e^{im\theta} \right] \eta_\xi
\]

\[
= e^{ikr \cos \theta} \begin{pmatrix} 1 \\ 1 \end{pmatrix} \eta_\uparrow + \sum_{m=-\infty}^{\infty} t_m^\uparrow \left[ \begin{pmatrix} H_m(kr) & iH_{m+1}(kr) \end{pmatrix} e^{im\theta} \right] \eta_\uparrow + t_m^\downarrow \left[ \begin{pmatrix} H_{m+1}(kr) & iH_m(kr) \end{pmatrix} e^{im\theta} \right] \eta_\downarrow
\]

\[
\approx e^{ikr \cos \theta} \begin{pmatrix} 1 \\ 1 \end{pmatrix} \eta_\uparrow + \left[ f_{\uparrow\uparrow}(\theta) \eta_\uparrow + f_{\uparrow\downarrow}(\theta) \eta_\downarrow \right] \begin{pmatrix} 1 \\ e^{i\theta} \end{pmatrix} e^{ikr \sqrt{r}}.
\]

Thus, the differential cross section is given by

\[
\frac{d\sigma}{d\theta} = \frac{R_{sc}}{J_m}
= |f_{\uparrow\uparrow}(\theta)|^2 + |f_{\uparrow\downarrow}(\theta)|^2.
\]

The above form is clearly identical to Eq. (27), and the relationship between the partial wave amplitude and the scattering amplitude is established in Eqs. (C10) and (C11).

2. Standard Rashba case

In the SR case, the conservation of angular momentum is restored. Therefore, the wave function in real space must be of the form

\[
\psi(r, \theta) \sim \begin{pmatrix} A_n(r) e^{im\theta} \\ B_{n+1}(r) e^{i(n+1)\theta} \end{pmatrix} \eta_\uparrow + \begin{pmatrix} C_{n+1}(r) e^{i(n+1)\theta} \\ D_{n+2}(r) e^{i(n+2)\theta} \end{pmatrix} \eta_\downarrow,
\]

where \(A, B, C, D\) are the unknowns to be derived from the Schrödinger equation. Following similar methods to solve for the NR case, we expand the wave function for \(r > R\)
as follows:
\[
\psi_n^\gamma(r,\theta) = \left( \frac{J_n(kr)e^{in\theta}}{iJ_{n+1}(kr)e^{i(n+1)\theta}} \right) \eta_\uparrow + t_n^{\uparrow\downarrow} \left( \frac{H_n(kr)e^{in\theta}}{iH_{n+1}(kr)e^{i(n+1)\theta}} \right) \eta_\uparrow + t_n^{\downarrow\uparrow} \left( \frac{H_{n+1}(kr)e^{i(n+1)\theta}}{iH_{n+2}(kr)e^{i(n+2)\theta}} \right) \eta_\downarrow,
\]
where we have assumed that the incident electron is from the conduction band with spinor \( \eta_\uparrow \). For \( r < R \), we solve the corresponding Schrödinger equation:
\[
(H_0 + V_{SR})\psi_n^\gamma(r,\theta) = E(k)\psi_n^\gamma(r,\theta),
\]
where the energy eigenvalue \( E(k) \) is diagonalized to be
\[
E(k) = v_0 + \eta \Delta_{SR} + \lambda \sqrt{\Delta_{SR}^2 + k^2}.
\]
Here \( \lambda \) and \( \xi \) are the two band indices. Note that the energy spectra for SR and NR are the same. Following the same convention in the NR case, we choose \( \lambda = +1 \). To solve for the coefficients \( A_n, B_n, C_n, \) and \( D_n \), we substitute Eq. (C15) into Eq. (C17),
\[
\begin{bmatrix}
-\epsilon_n & 0 & 0 & 0 \\
0 & -\epsilon_n & 0 & 0 \\
0 & 2i\Delta_{SR} & -\epsilon_n & 0 \\
0 & 0 & i\frac{\Delta_{SR}}{r} + \frac{i}{2}(n + 1) & -\epsilon_n
\end{bmatrix}
\begin{bmatrix}
A_n(r) \\
B_{n+1}(r) \\
C_{n+1}(r) \\
D_{n+2}(r)
\end{bmatrix} = 0.
\]
Here \( \epsilon_\xi \equiv E(k) - v_0 \). Setting \( A_n = J_n(q_\xi r) \), the system of differential equations can be solved and the solution reads
\[
\psi_n^\gamma(r,\theta) = \sum_{\xi = \pm 1} \alpha_\xi \left[ \left( \frac{J_n(q_\xi r)e^{in\theta}}{iJ_{n+1}(q_\xi r)e^{i(n+1)\theta}} \right) \eta_\uparrow + \left( \frac{-\epsilon_\xi J_{n+1}(q_\xi r)e^{i(n+1)\theta}}{i\lambda J_n(q_\xi r)e^{i(n+1)\theta}} \right) \eta_\downarrow \right].
\]

The wave number \( q_\xi = \sqrt{\epsilon_\xi^2 - 2\xi \epsilon_\xi \Delta_{SR}} \) and \( \alpha_\xi \) are linear combination weights to be determined from the boundary conditions. Using the boundary condition at the boundary of the circular potential \( \psi_n^\gamma(R,\theta) = \psi_n^\gamma(R,\theta) \), the partial wave amplitudes (i.e., \( t_n^{\uparrow\downarrow} \) and \( t_n^{\downarrow\uparrow} \)) can be determined, hence the scattering amplitudes can be determined as well [i.e., \( f_{\uparrow\downarrow}(\theta) \) and \( f_{\downarrow\uparrow}(\theta) \)]; see Eq. (C9).

Using the solution of the Boltzmann equation described in Appendix B, the spin Hall angle \( \gamma \) for both the SR and the NR case can be computed using the scattering amplitudes. We have plotted the results in Fig. 4 showing the \( v_0 \) dependence of \( \gamma \) for both the SR and NR cases.

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