Theory of Spin Hall Conductivity in n-Doped GaAs

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We develop a theory of extrinsic spin currents in semiconductors, resulting from spin-orbit coupling at charged scatterers, which leads to skew-scattering and side-jump contributions to the spin-Hall conductivity. Applying the theory to bulk n-GaAs, without any free parameters, we find spin currents that are in reasonable agreement with experiments by Kato *et al.* [Science **306**, 1910 (2004)].

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Generating and manipulating nonequilibrium spin magnetization by electric fields is one of the most desirable goals of semiconductor spintronics, because electric fields have potentialities for accessing individual spins at nanometer scale. Spin-orbit (SO) coupling is a mechanism for achieving this goal. It has the prominent consequence of the spin-Hall effect (SHE), where an electric current can induce a transverse spin current and nonequilibrium spin accumulation near sample boundaries. Recent observations of the SHE are important achievements [1,2]. Theoretically, two different mechanisms of SHE were proposed. The *extrinsic* mechanism [3–5] is based on spin-dependent scattering of electrons by impurities and is mostly due to Mott skew scattering [6]. An *intrinsic* mechanism also has been proposed, based on the recently advanced concept of ''dissipationless spin currents'' in a perfect crystal [7,8].

The theory of spin transport in media with SO coupling is intricate and includes all problems inherent in the theory of anomalous Hall effect (AHE), which has a long history; for reviews, see [9,10]. Also, the precise definition of spin currents is under dispute due to spin nonconservation in media with SO coupling. For small spin-relaxation rates, spin currents with nonzero divergence can lead to spin accumulations, which are experimentally observable quantities [11–13]. However, spin currents do not necessarily vanish in thermodynamic equilibrium, so their relation to spin transport and accumulation is not obvious [14].

These problems inherent in the spin-transport theory make identification of physical mechanisms underlying the SHE observed in Refs. [1,2] rather challenging. On one hand, Wunderlich *et al.* [2] observed a strong SHE in two-dimensional (2D) layers of p-GaAs and ascribed it to the intrinsic effect because of the large magnitude of the effect and large splitting of the energy spectrum typical of heavy holes. On the other hand, Kato *et al.* [1] attribute their measurement of SHE in 3D n-GaAs layers (2 μm thick) to the extrinsic mechanism. We believe that this is indeed the case as we explain in this work. Although an intrinsic spin-Hall effect, driven by the k^3 Dresselhaus SO coupling [15], could give rise to spin accumulation in this system, as proposed in Ref. [16], its estimated size, when impurity scattering is taken into account, is an order of magnitude smaller than the observations. Further, because of the large sample thickness, a specifically 2D mechanism of spin accumulation relying on the properties of near-edge states [17,18] cannot play a role in the geometry of Ref. [1].

In the following, we develop a theory of extrinsic spin currents in a 3D electron system. It results from intrinsic SO coupling in the bulk crystal that produces a SO contribution to the impurity potential. (This effect can occur even in an inversion symmetric crystal.) We show that scattering by charged impurities and SO interaction in n-GaAs are strong enough to support spin currents that are in reasonably good agreement with findings by Kato *et al.* [1] without using any adjustable parameters.

We consider an electron Hamiltonian of form

$$H = \frac{\hbar^2 k^2}{2m^*} + V(\mathbf{r}) + \lambda \boldsymbol{\sigma} \cdot (\mathbf{k} \times \nabla V), \tag{1}$$

where the potential energy $V(\mathbf{r})$ varies slowly on the scale of the host lattice constant. In vacuum, the last term of Eq. (1) results from relativistic corrections in the Pauli equation and is known as the Thomas term, with $\lambda = -\hbar^2/4m_0^2c^2 \approx -3.7 \times 10^{-6}$ Å^2, vacuum electron mass m_0, and velocity of light c. In direct gap cubic semiconductors such as GaAs, a SO interaction of the same form develops in the framework of the $\mathbf{k} \cdot \mathbf{p}$ model due to the

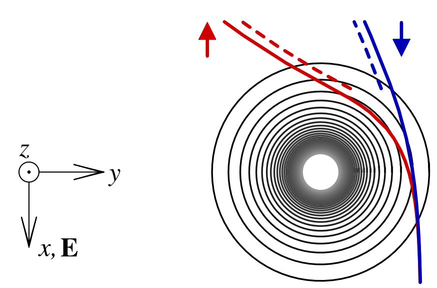

FIG. 1 (color online). Spin-dependent scattering at an attractive impurity. We show the classical trajectories (solid lines), for a screened Coulomb potential and for strongly exaggerated spin-orbit coupling with $\lambda > 0$ and with quantization axis $\hat{\mathbf{z}}$. The skew-scattering current results from different scattering angles for spin-$\uparrow$ and spin-$\downarrow$ and leads to a positive spin-Hall conductivity, $\sigma_{SS}^{SH} = -j_{SS,y}^z/E_x > 0$. Further, we show the horizontal displacement due to the side-jump effect (dashed lines), contributing to the spin current with opposite sign.

coupling of a s-type conductance band to p-type valence bands. For conduction band electrons in the 8×8 Kane model, one finds $\lambda = (P^2/3)[1/E_0^2 - 1/(E_0 + \Delta_0)^2]$ in third-order perturbation theory, with gap E_0, SO splitting Δ_0 between the $J = \frac{3}{2}$ and $J = \frac{1}{2}$ hole bands, and a properly normalized interband matrix element P of the momentum [19]. All these parameters are large (i.e., of atomic scale) since they result from the strong crystal potential. For GaAs, one finds $\lambda = 5.3$ Å^2. Thus, the SO coupling in n-GaAs is by 6 orders of magnitude stronger than in vacuum and has the *opposite sign*. This enhancement of SO coupling is critical for developing large extrinsic spin currents. Further, we ignore the k^3 (Dresselhaus) SO correction, which is small and is absent in the 8×8 Kane model. Thus, in our 3D system, SO coupling comes through V, which contains the potential of the driving electric field $\mathbf{E}$ and of the impurity centers.

We now analyze the effect of the SO coupling in Eq. (1) on the scattering at impurities, which leads to the extrinsic SHE. This comprises two contributions, one resulting from the skew-scattering at the impurities [6] and the other from the shift of the scattered wave packet [20] (side-jump contribution). They are shown schematically in Fig. 1.

The skew-scattering contribution is calculated in the lowest order both in SO interaction and in $1/k_F\ell$, with Fermi momentum k_F and mean free path ℓ. To this end, we describe the system by a spin-dependent Boltzmann equation and a distribution function written as a 2×2 spin matrix $\hat{f} = [f_0(\mathbf{k}) + \phi(\mathbf{k})]\mathbb{1} + \mathbf{f}(\mathbf{k}) \cdot \boldsymbol{\sigma}$, with equilibrium distribution function f_0. Below, we suppress the identity matrix $\mathbb{1}$. The collision integral on the right-hand side of the Boltzmann equation has the form

$$-\left(\frac{\partial \hat{f}(\mathbf{k})}{\partial t}\right)_{\mathrm{coll}} = n_i \sum_{\mathbf{k}';k'=k} \frac{\hbar k}{m^*} \frac{d\overleftrightarrow{\sigma}}{d\Omega}[\hat{f}(\mathbf{k}) - \hat{f}(\mathbf{k}')], \tag{2}$$

where n_i is the impurity density. The scattering cross section $d\overleftrightarrow{\sigma}/d\Omega$ is spin dependent and mixes spin components of the incoming flux. In the general case, this spin dependence is rather complex [21]. However, it simplifies essentially when we expand Eq. (2) in SO coupling and neglect the cross terms containing spin-dependent contributions of both $d\overleftrightarrow{\sigma}/d\Omega$ and $\hat{f}$. For a central symmetric impurity potential, we can then write

$$\frac{d\overleftrightarrow{\sigma}}{d\Omega}[\hat{f}(\mathbf{k}) - \hat{f}(\mathbf{k}')] = I(\vartheta)[\hat{f}(\mathbf{k}) - \hat{f}(\mathbf{k}')] - I(\vartheta)S(\vartheta)\boldsymbol{\sigma}\cdot\mathbf{n}[\phi(\mathbf{k}) + \phi(\mathbf{k}')], \tag{3}$$

where $\vartheta = \vartheta_{\mathbf{k}\mathbf{k}'}$ is the angle between $\mathbf{k}'$ and $\mathbf{k}$, and $\mathbf{n} = \mathbf{k}' \times \mathbf{k}/|\mathbf{k}' \times \mathbf{k}|$ is the unit vector normal to the scattering plane. The coefficient $I(\vartheta)$ is the spin-independent part of the scattering cross section, while $S(\vartheta)$ is the so-called Sherman function [6,21,22], which measures the polarization of outgoing particles scattered into direction $\mathbf{k}$ from an unpolarized incoming beam of momentum $\mathbf{k}'$.

To lowest order in the electric field, the left-hand side of the Boltzmann equation equals $e\mathbf{E} \cdot \frac{\partial f_0}{\partial \mathbf{p}} = (e\hbar/m^*) \times (\mathbf{E} \cdot \mathbf{k}) \frac{\partial f_0}{\partial \varepsilon}$, where the isotropy of $f_0(\mathbf{k})$ was used and $e < 0$ is the electron charge. The Boltzmann equation may then be solved with the following ansatz. First, the usual spin-independent term is set to $\phi(\mathbf{k}) = \mathbf{k} \cdot \mathbf{E} C_k$ [typically, $C_k = -(e\hbar\tau/m^*)\frac{\partial f_0}{\partial \varepsilon}$ with transport lifetime τ]. Second, we use $\mathbf{f}(\mathbf{k}) = (\mathbf{E} \times \mathbf{k})D_k$ for the spin polarization due to SO interaction. This structure is motivated by the physics of Mott scattering, where the spin polarization is perpendicular to the scattering plane defined by incoming electrons that drift in the direction of $-\mathbf{E}$ and are scattered into $\mathbf{k}$. Here, C_k and D_k are spherically symmetric functions of $\mathbf{k}$.

With this ansatz, we can evaluate the collision integrals on the right-hand side of the Boltzmann equation. Integrating over the direction of $\mathbf{k}'$, $d\Omega(\mathbf{k}') = d\varphi d\vartheta \times \sin\vartheta$, where φ is the azimuth of $\mathbf{k}'$ in the plane perpendicular to $\mathbf{k}$, and suppressing an overall factor of $n_i\hbar k/m^*$ in Eq. (2), we obtain [23]

$$\int d\Omega(\mathbf{k}')I(\vartheta)(\mathbf{k} - \mathbf{k}') \cdot (\mathbf{E}C_k + \boldsymbol{\sigma} \times \mathbf{E}D_k) = [\mathbf{k} \cdot \mathbf{E}C_k + \mathbf{k} \cdot (\boldsymbol{\sigma} \times \mathbf{E})D_k] \int d\Omega I(\vartheta)(1 - \cos\vartheta), \tag{4}$$

where the integral on the right is proportional to the inverse transport time τ^{-1}, and

$$-\int d\Omega(\mathbf{k}')I(\vartheta)S(\vartheta)(\boldsymbol{\sigma} \cdot \mathbf{n})(\mathbf{k} \cdot \mathbf{E} + \mathbf{k}' \cdot \mathbf{E})C_k = -\frac{1}{2}\mathbf{k} \cdot (\boldsymbol{\sigma} \times \mathbf{E})C_k \int d\Omega I(\vartheta)S(\vartheta)\sin\vartheta. \tag{5}$$

Since the left-hand side of the Boltzmann equation only depends on the component of $\mathbf{k}$ along the electrical field, this must also be the case on the right-hand side. Thus, the second term in Eq. (4) must cancel with Eq. (5), determining $D_k = \frac{1}{2}\gamma_k C_k$. We defined the *transport skewness*

$$\gamma_k = \frac{\int d\Omega I(\vartheta)S(\vartheta)\sin\vartheta}{\int d\Omega I(\vartheta)(1 - \cos\vartheta)}, \tag{6}$$

which describes the effect of skew scattering on the distribution function and depends on the structure of the scattering center and on the energy of the scattered particle. Therefore, our ansatz is self-consistent, and the solution of the Boltzmann equation is

$$\hat{f}(\mathbf{k}) = f_0(k) + \mathbf{k} \cdot \left[\mathbf{E} + \frac{\gamma_k}{2}(\boldsymbol{\sigma} \times \mathbf{E})\right]C_k; \tag{7}$$

i.e., the components of $\mathbf{f}(\mathbf{k})$ are $f_\mu(\mathbf{k}) = \frac{\gamma_k}{2}(\mathbf{E} \times \mathbf{k})_\mu C_k$.

Now we calculate the contribution of skew scattering to the spin current, $\mathbf{j}_{\mathrm{SS}}^{\mu} = n\langle\sigma_\mu \mathbf{v}_0\rangle$, with density n and with $\mathbf{v}_0 = \hbar\mathbf{k}/m^*$ (SO contributions to the velocity are analyzed below). We obtain

$$j^{\mu}_{\mathrm{SS},\kappa} = \mathrm{Tr}\sigma_{\mu} \int \frac{d^3k}{(2\pi)^3} \frac{\hbar k_{\kappa}}{m^*} \hat{f}(\mathbf{k}) = \frac{\gamma}{2e} \varepsilon^{\kappa\mu\nu} (\mathbf{J}_0)_{\nu}, \quad (8)$$

where $\mathbf{J}_0 = 2e \int d^3k (2\pi)^{-3} (\hbar\mathbf{k}/m^*) \mathbf{k} \cdot \mathbf{E} C_k$ is the charge current in the absence of SO coupling, and summation over ν is implied. Assuming low temperatures, we need to evaluate γ_k only near the Fermi energy E_{F} and we defined $\gamma \equiv \gamma_{k_{\mathrm{F}}}$ [24]. If there are different species of impurity potentials, the weighted average of the corresponding transport skewnesses γ [Eq. (6)] should be taken. Note that repulsive impurities generally lead to the opposite sign of γ. This can result in a partial suppression of spin-Hall currents due to impurity compensation.

Next we evaluate the contribution of the side jump [20] to spin currents, where the wave function is laterally displaced during the scattering event. (This displacement does not modify the scattering angle at large distances; i.e., it does not affect the scattering cross section.) Side-jump currents were analyzed in detail for the AHE [9], and we now relate the AHE to the SHE. In the AHE, a net polarization in combination with SO interaction at impurities leads to electrical Hall currents. For the SHE, electrons are unpolarized in equilibrium and we consider induced spin currents $\mathbf{j}^{\mu}$. Because λ is small and spin relaxation is of order λ^2 [25,26], one can understand the $\hat{\varepsilon}_{\mu}$ component of spin current as the difference in particle currents of two spin species with polarizations $\pm\hat{\varepsilon}_{\mu}$. For noninteracting electrons, each of these species carries the anomalous Hall current $\mathbf{J}^{\uparrow,\downarrow}_{\mathrm{AH}}$ of a system with density $n_{\mathrm{AH}} = \frac{1}{2}n$ and with spins fully aligned along the $\pm\hat{\varepsilon}_{\mu}$ direction. We can express the spin-Hall current as

$$\mathbf{j}^{\mu}_{\mathrm{SH}} = e^{-1}(\mathbf{J}^{\uparrow}_{\mathrm{AH}} - \mathbf{J}^{\downarrow}_{\mathrm{AH}}). \quad (9)$$

For the AHE, the side-jump contribution was found to be $\mathbf{J}^{\mathrm{SJ},\uparrow}_{\mathrm{AH}} = -2n_{\mathrm{AH}}\lambda(e^2/\hbar)\hat{\varepsilon}_{\mu} \times \mathbf{E}$ [9]. It results from SO corrections $\delta\dot{\mathbf{r}}$ to the velocity operator during impurity scattering. Nozières and Lewiner [9] clarified that this anomalous velocity $\delta\dot{\mathbf{r}}$ comprises two equal SO contributions. The first is $\delta_1\dot{\mathbf{r}} = (i/\hbar)[H, \mathbf{r}] - \mathbf{v}_0 = (\lambda/\hbar)(\nabla V \times \boldsymbol{\sigma})$ and becomes $\delta_1\dot{\mathbf{r}} = \lambda(\boldsymbol{\sigma} \times \dot{\mathbf{k}})$ after the equation of motion, $\dot{\mathbf{k}} = -\nabla V/\hbar$, is taken into account. The second originates from the correction to the coordinate operator, the Yafet term $\delta\mathbf{r}_{\mathrm{SO}} = \lambda(\boldsymbol{\sigma} \times \mathbf{k})$ [26], and contributes as $\delta_2\dot{\mathbf{r}} = \delta_1\dot{\mathbf{r}}$. Note that $\delta_2\dot{\mathbf{r}}$ leads to a factor of 2 which is often ignored. Heuristically, we can now understand the current $\mathbf{J}^{\mathrm{SJ}}_{\mathrm{AH}}$ as follows. For impurity scattering with momentum transfer $\delta\mathbf{k}$, the lateral displacement it $\delta\mathbf{r} = 2\lambda(\boldsymbol{\sigma} \times \delta\mathbf{k})$. The anomalous Hall current is recovered from $\mathbf{J}^{\mathrm{SJ}}_{\mathrm{AH}} = en\delta\mathbf{r}/\tau$, by using that the momentum dissipated per scattering event is $\hbar\delta\mathbf{k} = -e\mathbf{E}\tau$. Using $\mathbf{J}^{\mathrm{SJ}}_{\mathrm{AH}}$ [9] and Eq. (9), we readily obtain the side-jump contribution to the SHE,

$$j^{\mu}_{\mathrm{SJ},\kappa} = -2n\lambda \frac{e}{\hbar} \varepsilon^{\kappa\mu\nu} E_{\nu}. \quad (10)$$

The sum $\mathbf{j}_{\mathrm{SS}} + \mathbf{j}_{\mathrm{SJ}}$ provides the total spin-Hall current.

Mathematically, one could consider a model where the electron charge is cancelled by a uniform positive background, with only small fluctuations in the potential V. In such a case, we could let τ become arbitrarily large. Although the side-jump contribution to the spin-Hall conductivity is independent of τ, the skew-scattering contribution, given by Eq. (8), would grow with τ. This growth would be cut off when τ becomes comparable to $1/\Delta$, with SO splitting Δ at the Fermi level due to the k^3 Dresselhaus term. For sufficiently small potential fluctuations, $\gamma \ll \Delta/E_{\mathrm{F}}$ and the skew-scattering contribution will be smaller than the intrinsic contribution by a factor of order $\gamma E_{\mathrm{F}}/\Delta$.

We now evaluate the skewness γ [Eq. (6)] for a screened attractive Coulomb potential. For this, we make use of the long-established theory of single electron scattering by an atom [21]. We rescale the parameters to make a connection between the atomic Hamiltonian and the Hamiltonian [Eq. (1)] with $V = -e^{-q_s r} e^2/\epsilon r$, effective mass m^*, permittivity ϵ, and screening length $1/q_s$. We match V by setting the atomic number to $Z = 1/\epsilon$. Further, to match the SO interaction, we define an "effective" speed of light c^* such that $\lambda = \hbar^2/4(m^* c^*)^2$. Finally, when the sign of λ differs from its vacuum value, we replace $S(\vartheta)$ by $-S(\vartheta)$ in Eq. (6) [23]. For GaAs, $\epsilon = 12.4$ and $m^* = 0.0665 m_0$, thus $c^* \approx c/79$ and $\alpha^* Z \approx 1/21$, with fine structure constant $\alpha^* = e^2/\hbar c^*$. When evaluating $I(\vartheta)$, screening of the long-range Coulomb interaction is required, otherwise the denominator in Eq. (6) diverges, e.g., $\tau \to 0$. For exponential screening and in second order Born approximation, $I(\vartheta)$ is given by the Dalitz formula [21]. We assume Thomas-Fermi screening with inverse screening length $q_s = \sqrt{3e^2 n/2\epsilon E_{\mathrm{F}}}$. For $S(\vartheta)$, we use the exact expression for an unscreened potential [6,21]. We find that for q_s in a range near its experimental value, the contributions to Eq. (6) come primarily from angles near $\pi/2$ and the resulting value of γ depends only weakly on screening. (However, in the limit $q_s \to 0$, γ vanishes logarithmically as small angles dominate the denominator.) The small parameter controlling $S(\vartheta)$ is $(Z\alpha^*)^2 = 4|\lambda|/(a^*_{\mathrm{B}})^2$, with effective Bohr radius $a^*_{\mathrm{B}} = \hbar^2\epsilon/m^* e^2$. Estimating $\gamma \sim S$, we get $1/500$ for γ and also $j_{\mathrm{SS}}/j_{\mathrm{SJ}} \sim -\tau E_B/\hbar$ with impurity binding energy $E_B = \hbar^2/m^*(a^*_{\mathrm{B}})^2$.

We now estimate γ for Si-doped GaAs and for electron density $n = 3 \times 10^{16}\ \mathrm{cm}^{-3}$ as reported in Ref. [1], i.e., $E_{\mathrm{F}} = 5.3$ meV and $q_s^{-1} \approx 9$ nm. As a test, we first evaluate the electrical conductivity using an impurity density $n_i = n$ and the Drude formula, $\sigma_{xx} = e^2 n\tau/m^*$ with $\tau^{-1} = n_i v_{\mathrm{F}} \int d\Omega I(\vartheta)(1 - \cos\vartheta)$, and arrive at $\sigma_{xx} \approx 1.8 \times 10^3\ \Omega^{-1}\,\mathrm{m}^{-1}$. This is within 10% of the experimentally observed conductivity at low voltages—a surprisingly good agreement, given that the rather small value $k_{\mathrm{F}}\ell \approx 2$ restricts the accuracy of the Boltzmann approach. (Note that τ is not needed explicitly in our evaluation.) Next we evaluate Eq. (6) and find $\gamma \approx 1/900$. This value is rather stable and changes by less than 30% when q_s or E_{F} increase or decrease by a factor of 2.

Next, we estimate the spin-Hall currents. The measurements were performed at electrical fields $E \approx 20$ mV μm^{-1} where the conductivity increased to $\sigma_{xx} \approx 3 \times 10^3\ \Omega^{-1}\,\mathrm{m}^{-1}$ due to electron heating. We assume that γ is not very sensitive to these heating effects and we still use Eq. (8) but with the increased conductivity. For an electrical field $\mathbf{E} = \hat{\mathbf{x}} E_x$, we find both contributions to the spin-Hall conductivity $\sigma^{\mathrm{SH}} \equiv -j_y^z/E_x$, namely, $\sigma_{\mathrm{SS}}^{\mathrm{SH}} = -(\gamma/2e)\sigma_{xx} \approx 1.7\ \Omega^{-1}\,\mathrm{m}^{-1}/|e|$ and $\sigma_{\mathrm{SJ}}^{\mathrm{SH}} = 2n\lambda e/\hbar \approx -0.8\ \Omega^{-1}\,\mathrm{m}^{-1}/|e|$. In total, we arrive at the extrinsic spin-Hall conductivity $\sigma_{\mathrm{theor}}^{\mathrm{SH}} \approx 0.9\ \Omega^{-1}\,\mathrm{m}^{-1}/|e|$. The magnitude is within the error bars of the experimental value of $|\sigma_{\mathrm{expt}}^{\mathrm{SH}}| \approx 0.5 \Omega^{-1}\mathrm{m}^{-1}/|e|$ found from spin accumulation near the free edges of the specimen [1,27].

The sign of $\sigma_{\mathrm{expt}}^{\mathrm{SH}}$ reported in Ref. [1] is actually opposite to the one we calculate. However, there remains uncertainty about the absolute sign in the experiments [28], and further experimental work is needed. Further, since our theoretical result is the difference of two terms with comparable magnitudes, and we have made numerous approximations, including the neglect of dynamic electron-electron interactions and multiple scattering effects, and since the parameter $k_{\mathrm{F}}\ell$ is not large, it is possible that our calculation would give an incorrect sign. Nevertheless, the fact that our result has the correct order of magnitude is highly encouraging, and is a strong argument that the observations of Ref. [1] can be understood as arising from the scattering mechanism we consider.

Further experimental tests would be possible if one can construct samples with larger values of $k_{\mathrm{F}}\ell$, or compensated samples, with negative as well as positive impurities. The latter should increase side-jump and reduce the skew-scattering contributions, giving a negative shift to σ^{SH}.

Bernevig and Zhang calculated intrinsic spin currents due to k^3-Dresselhaus interaction [16]. In contrast to the case of spin-orbit interactions linear in $\mathbf{k}$, where vertex corrections lead to a cancellation of the spin current [12,29,30], for the k^3 interaction this is not true; e.g., for s-wave scatterers, the vertex corrections vanish identically. However, in the dirty limit $\tau\Delta/\hbar \sim 10^{-2}$, and for experimental parameters of Ref. [1], intrinsic spin currents were found to be small, only $\sigma_{\mathrm{D}} \approx 0.02\,\Omega^{-1}\,\mathrm{m}^{-1}/|e|$ [16].

In conclusion, we solved analytically the kinetic equation including skew scattering at impurities. We obtained the contributions to the extrinsic spin-Hall effect and found, without using any free parameters, reasonable agreement with experimental data.

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