Generation of Spin Currents and Spin Densities in Systems with Reduced Symmetry

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We show that the spin-current response of a semiconductor crystal to an external electric field is considerably more complex than previously assumed. While in systems of high symmetry only the spin-Hall components are allowed, in systems of lower symmetry other non-spin-Hall components may be present. We argue that, when spin-orbit interactions are present only in the band structure, the distinction between intrinsic and extrinsic contributions to the spin current is not useful. We show that the generation of spin currents and that of spin densities in an electric field are closely related, and that our general theory provides a systematic way to distinguish between them in experiment. We discuss also the meaning of vertex corrections in systems with spin-orbit interactions.

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Ground-breaking work in past years has turned semiconductor spin electronics into a richly rewarding field, both theoretically and experimentally. The application of an electric field to a semiconductor sample gives rise to a nonequilibrium spin current [1–4] as well as a steady-state spin density in the bulk of the sample [5–7]. The latter was first observed several decades ago [5], while the recent imaging [8] and *direct* measurements of spin currents [9,10], together with the achievement of the room-temperature spin-Hall effect [11] have stimulated an enormous amount of research [12]. Other schemes for generating and detecting spin currents have been implemented or proposed [13].

The development of electrical spin manipulation techniques has brought the fundamental physics of spin transport under intense scrutiny [14–21]. Debate has focused on definitions of spin currents [14], on whether spin currents are transport or background currents [15], whether scattering-independent or scattering-dependent contributions are dominant [12], on the role played by spin Coulomb drag [19], and on spin accumulation at the boundary [22]. Whereas most studies to date have paid significant attention to common semiconductors and asymmetric quantum wells (QW) [22–36], recent developments call for an in-depth investigation of neglected aspects.

In this Letter, we first discuss the relationship between spin currents in a crystal and the symmetry of the underlying lattice. Such an analysis has been enlightening in the context of nonequilibrium spin densities excited by an electric field. If the response of the spin density $\boldsymbol{s}$ to an electric field $\boldsymbol{E}$ is given by $s^\sigma = Q^\sigma_j E_j$, nonzero components for the material-specific spin density response tensor Q^σ_j are permitted only in gyrotropic crystals [5]. Yet such an analysis has not been done for spin transport. We therefore determine the components of the spin-current response tensor allowed by symmetry in an electric field, providing systematic proof that spin currents in response to an electric field can be much more complex than the spin-Hall effect [37]. This result is completely general and is not sensitive to the definition of the spin current or to whether the electric field is constant or time dependent. The subsequent calculations confirm these predictions by determining the corrections to the density matrix present in an electric field and demonstrating their intimate relationship with spin precession. We argue that, if spin-orbit interactions are present only in the band structure, only one contribution to the spin current exists, which appears intrinsic in the weak momentum-scattering limit and extrinsic in the strong momentum-scattering limit. We show that spin currents and bulk spin densities in an electric field arise from linearly independent contributions to the density matrix, and that certain setups can measure effects due solely to spin currents. Our work considers realistic scattering potentials and is very relevant to experiment, where the high symmetry often assumed in theoretical approaches is usually not present.

The spin-current operator is defined as $\hat{\mathcal{J}}^\sigma_i = \frac{1}{2}(\hat{s}^\sigma \hat{v}_i + \hat{v}_i \hat{s}^\sigma)$, where $\hat{s}^\sigma$ represents the spin component σ, and the velocity operator $\hat{v}_i = 1/\hbar(\partial \hat{H}/\partial k_i)$, with $\hat{H}$ the Hamiltonian. An alternative, more realistic definition of the spin current has been proposed [14], according to which $\hat{\mathcal{J}}^\sigma_i = d/dt(\hat{r}_i \hat{s}^\sigma)$. Yet we remark that from a symmetry point of view these two definitions are equivalent so that the following symmetry analysis applies to both definitions. The spin current $\hat{\mathcal{J}}$ is a second rank tensor that can be decomposed into a pseudoscalar part, an antisymmetric (spin-Hall) part, and a symmetric part. The pseudoscalar part is $\mathrm{tr}(\hat{\mathcal{J}}) = \frac{1}{3}\hat{\boldsymbol{s}} \cdot \hat{\boldsymbol{v}}$ and represents a spin flowing in the direction in which it is oriented. The symmetric and antisymmetric parts are given, respectively, by $\frac{1}{2}(\hat{s}^\sigma \hat{v}_i \pm \hat{v}_\sigma \hat{s}^i)$. The pseudoscalar and symmetric parts will be referred to as *non-spin-Hall* currents. Under the full orthogonal group only the antisymmetric (spin-Hall) components of $\hat{\mathcal{J}}$ are allowed, indicating that these components are always permitted by symmetry.

In general, the spin-current response of a crystal to an electric field $\boldsymbol{E}$ is characterized by a material tensor $\boldsymbol{T}$ defined by $\mathcal{J}_i^\sigma = T_{ij}^\sigma E_j$. For the 32 crystallographic point groups the symmetry analysis [38] for the tensor $\boldsymbol{T}$ is established by means of standard compatibility relations [39]. One is particularly interested in those groups in which non-spin-Hall components may be present. Our calculations show that the pseudoscalar part of the spin current is only allowed by 13 point groups, while the symmetric part is allowed in all systems except those with point groups O, T_d (zinc blende), or O_h (diamond). Lower symmetries, allowing non-spin-Hall currents, are characteristic of systems of reduced dimensionality.

It is well known that the generation of a spin density by an electric field is the inverse of the circular photogalvanic effect [5,13], while the spin-Hall effect also has an inverse [10]. Since spin densities induced by electric fields are restricted to gyrotropic crystals [5] the same restriction applies to the circular photogalvanic effect. Similarly, the symmetry analysis developed here is also applicable to the inverse spin-Hall effect which needs to be complemented by an inverse non-spin-Hall effect in systems with reduced symmetry.

In order to verify the proposition that non-spin-Hall currents may exist in a variety of crystals, we discuss spin currents induced by an electric field in spin-1/2 electron systems. For electrons the effective Hamiltonian is written as $H = H_{\text{kin}} + H_{\text{so}}$, where $H_{\text{kin}} = \hbar^2 k^2/2m^*$ and $H_{\text{so}} = \frac{1}{2}\boldsymbol{\sigma}\cdot\boldsymbol{\Omega}$, where $\boldsymbol{\Omega}$ is a momentum-dependent effective Zeeman field. In the weak momentum-scattering regime we have $\varepsilon_F \tau_p/\hbar \gg \Omega\tau_p/\hbar \gg 1$, with τ_p the momentum relaxation time and ε_F the Fermi energy, whereas in the strong momentum-scattering regime $\varepsilon_F\tau_p/\hbar \gg 1 \gg \Omega\tau_p/\hbar$.

The system is described by a density operator $\hat{\rho}$, which is expanded in a basis of definite wave vector as

$$\hat{\rho} = \sum_{n,n'}\sum_{\boldsymbol{k},\boldsymbol{k}'} \rho_{nn'\boldsymbol{k}\boldsymbol{k}'}(t)|\psi_{n\boldsymbol{k}}(t)\rangle\langle\psi_{n'\boldsymbol{k}'}(t)|. \tag{1}$$

A constant uniform electric field $\boldsymbol{E}$ is included in the crystal momentum through the vector potential $\boldsymbol{A}$ such that $\boldsymbol{k} = \boldsymbol{q} + e\boldsymbol{A}/\hbar$, and the wave functions are chosen to have the form $|\psi_{n\boldsymbol{k}}(t)\rangle = e^{i\boldsymbol{q}\cdot\boldsymbol{r}}|u_{n\boldsymbol{k}}(t)\rangle$, where $|u_{n\boldsymbol{k}}(t)\rangle$ are lattice-periodic functions that are *not* assumed to be eigenfunctions of the crystal Hamiltonian. In this basis the matrix elements $\rho_{nn'\boldsymbol{k}\boldsymbol{k}'}(t)$ form the density matrix and the impurity potential has matrix elements $\mathcal{U}_{\boldsymbol{k}\boldsymbol{k}'}\mathbb{1} + \mathcal{V}_{\boldsymbol{k}\boldsymbol{k}'}$, where $\mathcal{V}_{\boldsymbol{k}\boldsymbol{k}'}$ is the spin-dependent part.

The time evolution of the density operator is given by the quantum Liouville equation, which allows us to derive rigorously the time evolution of the part of the density matrix $\rho \equiv \rho_{nn'\boldsymbol{k}\boldsymbol{k}}$ diagonal in wave vector. We subdivide $\rho = \rho_0 + \rho_E$, where ρ_0 is given by the Fermi-Dirac distribution, and the correction ρ_E is due to the electric field $\boldsymbol{E}$. To first order in $\boldsymbol{E}$, ρ_E satisfies

$$\frac{\partial \rho_E}{\partial t} + \frac{i}{\hbar}[H, \rho_E] + \hat{J}(\rho_E) = \frac{e\boldsymbol{E}}{\hbar}\cdot\frac{\partial\rho_0}{\partial\boldsymbol{k}}, \tag{2}$$

where $\hat{J}(\rho_E)$ is the collision integral to be given below. The eigenvalues of H are $\epsilon_\pm = \varepsilon_0 \pm \Omega/2$, with $\varepsilon_0 = \hbar^2k^2/2m^*$. The spin-current operator is simply $\hat{\mathcal{J}}_i^\sigma = \hbar k_i s^\sigma/m^* + \frac{1}{4}\hbar\partial\Omega^\sigma/\partial k_i$. In electron systems we usually have $H_{\text{so}} \ll H_{\text{kin}}$, thus $\rho_0 \approx f_0\mathbb{1} + H_{\text{so}}\partial f_0/\partial\varepsilon_0$, where $f_0(\varepsilon_0)$ is the Fermi-Dirac distribution. ρ_E is divided into a scalar part and a spin-dependent part, $\rho_E = f_E\mathbb{1} + S_E$. To first order in $H_{\text{so}}/H_{\text{kin}}$ the scattering term can be expressed as $\hat{J}(\rho_E) = (\hat{J}_0 + \hat{J}_s + \hat{J}_v)(f_E) + \hat{J}_0(S_E)$, where

$$\hat{J}_0(f_E) = \frac{2\pi n_i}{\hbar}\int\frac{d^dk'}{(2\pi)^d}|\mathcal{U}_{\boldsymbol{k}\boldsymbol{k}'}|^2(f_E - f_E')\delta(\varepsilon_0 - \varepsilon_0'), \tag{3a}$$

$$\begin{aligned}\hat{J}_s(f_E) = \frac{\pi n_i}{2\hbar}\boldsymbol{\sigma}\cdot\int\frac{d^dk'}{(2\pi)^d}&|\mathcal{U}_{\boldsymbol{k}\boldsymbol{k}'}|^2(f_E - f_E')\{(\hat{\boldsymbol{\Omega}} + \hat{\boldsymbol{\Omega}}')\\ &\times[\delta(\epsilon_+ - \epsilon_+') - \delta(\epsilon_- - \epsilon_-')]\\ &+ (\hat{\boldsymbol{\Omega}} - \hat{\boldsymbol{\Omega}}')[\delta(\epsilon_+ - \epsilon_-') - \delta(\epsilon_- - \epsilon_+')]\},\end{aligned} \tag{3b}$$

$$\hat{J}_v(f_E) = \frac{2\pi n_i}{\hbar}\int\frac{d^dk'}{(2\pi)^d}Y_{\boldsymbol{k}\boldsymbol{k}'}(f_E - f_E')\delta(\varepsilon_0 - \varepsilon_0'). \tag{3c}$$

In the above n_i is the impurity density, d is the dimensionality of the system, primed quantities denote functions of $\boldsymbol{k}'$, $\hat{\boldsymbol{\Omega}}$ is a unit vector along $\boldsymbol{\Omega}$, and $Y_{\boldsymbol{k}\boldsymbol{k}'} = \int d^dk''/(2\pi)^d \times (\mathcal{U}_{\boldsymbol{k}\boldsymbol{k}''}\mathcal{V}_{\boldsymbol{k}''\boldsymbol{k}'} + \mathcal{V}_{\boldsymbol{k}\boldsymbol{k}''}\mathcal{U}_{\boldsymbol{k}''\boldsymbol{k}'})$. Equation (3a) is the usual scalar scattering term, Eq. (3b) is due to band structure spin-orbit coupling, and Eq. (3c) is due to spin-orbit coupling in the impurities. We will assume henceforth that band structure spin-orbit interactions are much stronger than those due to impurities.

For f_E we obtain the standard correction $f_E = (e\boldsymbol{E}\tau_p/\hbar)\partial f_0/\partial\boldsymbol{k}\mathbb{1}$. As a result, the effective source term that enters the equation for S_E is $\Sigma_s - \hat{J}_s(f_E)$, where Σ_s is the spin-dependent part of $(e\boldsymbol{E}/\hbar)\partial\rho_0/\partial\boldsymbol{k}$. In analogy with the Gram-Schmidt orthogonalization of vectors, this effective source term can be divided into two parts, $\Sigma_s - \hat{J}_s(f_E) = \Sigma_\parallel + \Sigma_\perp$, of which $\Sigma_\parallel$ commutes with the spin-orbit Hamiltonian

$$\Sigma_\parallel = \frac{\text{tr}\{[\Sigma_s - \hat{J}_s(f_E)]H_{\text{so}}\}}{\text{tr}(H_{\text{so}}^2)}H_{\text{so}}, \tag{4}$$

while $\Sigma_\perp$ is the remainder. In matrix language $\Sigma_\perp$ is *orthogonal* to the Hamiltonian, thus $\text{tr}(\Sigma_\perp H_{\text{so}}) = 0$. To find $\Sigma_\parallel$ and $\Sigma_\perp$ we define projectors $P_\parallel$ and $P_\perp$ onto and orthogonal to H_{so}, respectively. Acting on the basis matrices $\boldsymbol{\sigma}$, $P_\parallel\boldsymbol{\sigma} = 2\boldsymbol{\Omega}H_{\text{so}}/\Omega^2$, while $P_\perp\sigma_x = [(\Omega_y^2 + \Omega_z^2)\sigma_x - \Omega_x\Omega_y\sigma_y - \Omega_x\Omega_z\sigma_z]/\Omega_k^2$, and the remaining terms are obtained by cyclic permutations.

S_E can likewise be divided into two linearly independent contributions, $S_\parallel$ and $S_\perp$, the former of which commutes with $H_{\rm so}$ while the latter is orthogonal to it. It is helpful to think of $S_\parallel$ as the distribution of *conserved* spins parallel to $\boldsymbol{\Omega}$ and of $S_\perp$ as the distribution of *precessing* spins perpendicular to it. $S_\parallel$ satisfies

$$\frac{\partial S_\parallel}{\partial t} + P_\parallel[\hat{J}_0(S_E)] = \Sigma_\parallel. \tag{5}$$

This equation can be solved iteratively for any scattering. To solve the equation for $S_\perp$, on the other hand, one needs to expand $S_\perp$ in the strength of the scattering potential $|\mathcal{U}|^2$, as $S_\perp = S_{\perp 0} + S_{\perp 1} + O(|\mathcal{U}|^4)$. (It is easily shown that the first term in the expansion must be zeroth order in $|\mathcal{U}|^2$, while $S_\parallel$ starts at order -1.) The equations for these contributions are

$$\frac{\partial S_{\perp 0}}{\partial t} + \frac{i}{\hbar}[H_{\rm so}, S_{\perp 0}] = \Sigma_\perp - P_\perp[\hat{J}_0(S_\parallel)], \tag{6a}$$

$$\frac{\partial S_{\perp 1}}{\partial t} + \frac{i}{\hbar}[H_{\rm so}, S_{\perp 1}] = P_\perp[\hat{J}_0(S_{\perp 0})]. \tag{6b}$$

It follows that if $\Sigma_\perp - P_\perp[\hat{J}_0(S_\parallel)]$ vanishes in Eq. (6a), as it does for the linear Rashba model, then all the contributions to $S_\perp$ vanish. In general, the equation for $S_{\perp 0}$ is also solved iteratively for any scattering, but a closed-form solution for S_E is not always possible. An enlightening closed-form solution is, however, possible for short-range impurities, where $\hat{J}_0(S_E) = (S_E - \bar{S}_E)/\tau_p$, with the bar denoting averaging over directions in k space and $\tau_p = \hbar^3/(m^*|\mathcal{U}|^2 k^{D-2} V_{uc})$, with D the dimensionality and V_{uc} the unit cell volume. The series of equations of increasing order in $|\mathcal{U}|^2$ give two geometric progressions that sum to

$$S_\parallel = \Sigma_\parallel \tau_p + P_\parallel(1 - \bar{P}_\parallel)^{-1}\bar{\Sigma}_\parallel \tau_p, \tag{7a}$$

$$S_\perp = -\frac{\Sigma_\perp \tau_p + P_\perp \bar{S}_\parallel}{1 + \Omega^2\tau_p^2/\hbar^2} + \frac{\boldsymbol{\Omega} \times ({}_\perp \tau_p + P_\perp \bar{S}_\parallel) \cdot \boldsymbol{\sigma}\tau_p}{2\hbar(1 + \Omega^2\tau_p^2/\hbar^2)}. \tag{7b}$$

In the general case considered below the complex expressions for S_E, $\Sigma_\parallel$ and $\Sigma_\perp$ will not be given explicitly.

Our analysis clarifies the relation between steady-state spin currents and spin densities in electric fields. Since the spin operator is even in $\boldsymbol{k}$ and the spin-current operator is odd in $\boldsymbol{k}$, it emerges, after evaluating $S_\parallel$ and $S_\perp$, that the right-hand side (RHS) of Eq. (7a) is responsible for steady-state spin densities [5–7], while the second term on the RHS of Eq. (7b) gives rise to spin currents [1–4]. [The first term on the RHS of Eq. (7b) vanishes in both the weak and the strong momentum-scattering limits.] We conclude that spin densities arise from $S_\parallel$, the distribution of conserved spin while spin currents arise from $S_\perp$, the distribution of precessing spin. Thus nonequilibrium spin currents are due to spin precession (as first demonstrated in [4]) while nonequilibrium spin densities [5–7] are due to the *absence* of spin precession. The physical picture for the latter mechanism is as follows. Each spin on the Fermi surface precesses about an effective field $\boldsymbol{\Omega}(\boldsymbol{k})$ and the spin component parallel to $\boldsymbol{\Omega}(\boldsymbol{k})$ is preserved. In equilibrium the average of the conserved components is zero, but when an electric field is applied the Fermi surface is shifted and the average of the conserved spin components may be nonzero. This intuitive physical argument [6] explains why the nonequilibrium spin density $\propto \tau_p^{-1}$ and *requires* scattering to balance the drift of the Fermi surface. On the other hand, spin currents contain only terms $\propto \tau_p^{2n}$ with $n = 0, 1, 2, \ldots$.

It can be seen from Eq. (7b) that there is only *one* contribution to the spin current, which in the weak momentum-scattering (intrinsic) limit is independent of τ_p and in the strong momentum-scattering (extrinsic) limit is $\propto \tau_p^2$. It was also noted that, if the RHS of Eq. (6a) vanishes, then $S_\perp$ vanishes to all orders. We conclude that, if spin-orbit interactions exist only in the band structure, the distinction between intrinsic and extrinsic spin contributions to the spin current is not useful.

In calculations of spin currents $\mathcal{J}$ based on Green's functions formalisms, vertex corrections are important [28–30]. The above analysis suggests that contributions due to two processes are contained in vertex corrections. Scattering renormalizes the driving term for S_E to $\Sigma_s - \hat{J}_s(f_E)$, and mixes the conserved and precessing spin distributions, as in Eq. (6a). This implies that vertex corrections to $\mathcal{J}$ contain the influence of a steady-state spin density on $\mathcal{J}$ [40]. As this spin density occurs only in gyrotropic materials [5], we expect vertex corrections to spin currents to vanish in nongyrotropic materials.

Engel *et al.* [32] showed that, when band structure spin-orbit interactions are negligible, spin currents arise from skew scattering. A comparison of our results with those of Ref. [32] shows that scattering can give rise to spin currents of qualitatively different forms depending on whether spin-orbit interactions are present in the band structure or not. A general analysis of band structure spin-orbit interactions and skew scattering on the same footing remains to be undertaken.

For known cases, our theory agrees with previous work. 2D Hamiltonians with spin-orbit coupling linear in $\boldsymbol{k}$ give zero spin current for any scalar scattering potential [30,41], including short-range [20,28–31], and small-angle scattering [20,21,23]. For Hamiltonians characterized by one Fourier component N [21] the spin current $\propto N$. In 3D the *correction* to the spin current due to $\hat{J}_s(f_E)$ vanishes for the k^3-Dresselhaus model and short-range impurities. Our results also agree with previous calculations of spin generation [6].

As a specific example, we investigate the spin-current response tensor $\boldsymbol{T}$ in systems of low symmetry. A strong justification for this choice is that experiment often studies low-symmetry structures whereas theory is often done for high-symmetry models. We concentrate thus on quantum wells and, considering for definiteness a symmetric 150-Å wide GaAs well grown along [113]. The axes of the coordinate system are $x = [33\bar{2}]$, $y = [1\bar{1}0]$ and $z = [113]$. Spin-orbit interactions are described by k-linear and k^3-Dresselhaus terms [42] and impurity scattering by a screened Coulomb potential, with the Thomas-Fermi wave vector $k_0 = 4k_F$ at carrier density $n = 1.6 \times 10^{11}$ cm^{-2}. We do not consider skew scattering, which is important when band structure spin-orbit interactions are weak [11,32]. In units of $e/(8\pi)$, we obtain $T^x_{xx} = 0.398$, $T^x_{yy} = 0.12$, $T^z_{yx} = 0.172$, and $T^z_{xy} = -0.414$, which shows that for realistic scattering in a system of low symmetry many components of $\boldsymbol{T}$ are nonzero. At the boundary one must study also the spin current according to the alternative definition [14]. Previous work [14] has shown this to be of the same order of magnitude as the conventional spin current, with occasional sign differences. As our symmetry analysis holds for both definitions of the spin current, we expect results to conform to the pattern found above.

Taking the QW along [113] considered above, an electric field applied along x will produce a nonequilibrium spin density, a spin-Hall current and a longitudinal spin current composed of spin-x only. If the QW is joined to a material in which no nonequilibrium spin density is generated, then a Kerr rotation [3] or magnetic circular dichroism [43] experiment will give a nonzero signal exclusively due to the injected spin-x current.

In summary, we have shown that in systems with reduced symmetry spin currents are not restricted to the spin-Hall effect, and that this fact can help one distinguish experimentally between electrically induced spin densities and spin currents. We have demonstrated in addition that spin currents in an electric field are associated with spin precession, whereas spin densities are associated with the absence of spin precession.

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