Electric generation of spin in crystals with reduced symmetry

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Motivated by recent experimental progress in the detection of spin accumulation in semiconductors, we propose a way of generating a spin polarization in crystals with strong spin-orbit interactions. We show that, in the presence of an electric field, there exists an intrinsic torque term which gives rise to a nonzero spin generation rate. This spin generation rate is experimentally observable. The wide applicability of this effect is emphasized by explicit consideration of a range of examples: bulk wurtzite and strained zincblende (n-GaAs) lattices, as well as quantum well heterojunction systems.

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A considerable amount of attention has been devoted in recent years, both experimentally and theoretically, to the field of semiconductor spintronics. There have been numerous suggestions of advantages offered by the manipulation of the spin degree of freedom, including the increased functionality of spin devices, low power consumption, integration with existing technologies and the fact that in spin transport quantum coherence can be maintained on much larger time scales. Semiconductors can be used in the same spintronic devices as metals, and have the additional advantages brought about by the existence of an adjustable bandgap and by the ability to manipulate the carriers over many orders of magnitude through doping, gating and heterojunction formation. Moreover, a spin accumulation in a semiconductor will generate a much larger voltage because the density of states at the Fermi energy is lower than in a metal, leading to a higher spin splitting [1].

To this end it would be extremely desirable if a practical method existed for the efficient generation of a spin polarization inside a semiconductor, as well as for the transport of spins over *a sizable length scale*. Although optical spin injection [2] has been known for decades, it is impractical for devices, since it is not sufficiently local for nanoelectronics. On the other hand, spin injection from a ferromagnetic metal into a semiconductor requires long spin lifetimes [3] and is impeded by the resistivity mismatch between the two materials [4]. Progress has been achieved with ferromagnetic semiconductors [4, 5, 6, 7, 8, 9], but room temperature semiconductor ferromagnetism [10] has not yet been clearly established. Recent research has focused on the electrical control of spins in semiconductors [11, 12, 13], including theoretical work on the possibility of observing a spin-Hall effect [14], together with various efforts to generate a spin current [15] and the suggestion by Rashba [16] that spin currents can exist in thermodynamic equilibrium, as well as further theoretical investigations by the same author regarding the nature of the spin current [17]. Nevertheless, despite this energetic theoretical enterprise, a practical framework for detecting and measuring spin currents has not been devised to date.

Recent experimental work [18, 19], on the other hand, has demonstrated the detection of a sizable spin *accumulation* in semiconductors to be a feasible task. Motivated by these findings, in this Letter we present a general theory of the intrinsic electrical spin injection that occurs generically in spin-orbit coupled systems that are not inversion symmetric. The existence of such a mechanism has been pointed out by Levitov [20] and Magarill [21]. Our approach is to be contrasted with the early theories (e.g. [20]) which consider directly the response of a spin polarization to an electric field. Approaches that calculate the spin density directly via linear response do not separate the intrinsic generation terms from the extrinsic scattering effects and thus lose physical transparency. On the other hand, in the present paper we recognize spin generation as an intrinsic effect so that it may be determined from first principles calculations. Experiment [18] has shown that it is possible to measure the spin generation rate separately from the spin relaxation time. The idea we discuss has already been applied to the Rashba Hamiltonian [21], which we also examine below. The interplay of the spin generation and relaxation terms, resulting in a finite spin polarization, emerges in the final analysis.

Spin-orbit interactions can be important in semiconductors for several reasons. The first is the fact that the carriers are clustered near the band extrema around high symmetry points where there exist degeneracies, and the form of these degeneracies is determined by the spin-orbit interaction. Due to the fact that in semiconductors the carriers occupy a narrow width of k-space the spin-orbit interaction plays a crucial role. We will show in this Letter that, because of the fact that spin is not conserved, there exists a term which acts as a bulk source of spin generation. It represents the rate of change of the spin density in response to an external electric field. Aside from the references mentioned above, intrinsic spin-orbit effects have not been taken into account previously, ex-

cept in discussions of the role they play in spin relaxation [22]. Nevertheless, these intrinsic spin-orbit effects have been shown to lead to a non-zero Berry curvature which gives a contribution to the anomalous Hall effect [23, 24, 25], while a spin-orbit-induced metal-insulator transition has been detected by Koga *et al.* [26].

Because both the rate of change of the spin density and the electric field are even under time reversal, ferromagnetism is not required for spin generation. Nevertheless, because the electric field changes sign under spatial inversion while the rate of change of the spin density does not, the crystal must be asymmetric under spatial inversion. The mechanism we outline applies to a wide class of systems, and to illustrate this we discuss two-dimensional heterostructures described by the Rashba model, followed by a model of the conduction band of strained bulk zincblende as well as of unstrained bulk wurtzite structures, and numerical results for bulk n-GaAs.

In the picture we consider, the spin-orbit interaction has been taken into account in the bandstructure. The construction of a wavepacket representing a charge and spin carrier, which has real and k-space coordinates $(\mathbf{r}_c, \mathbf{k})$, has been thoroughly treated in [27] and will not be considered at length here. The carriers are described by a Boltzmann phase space distribution $f(\mathbf{r}_c, \mathbf{k}, t)$, the time evolution of which has been discussed in [14, 25]. The study of spin generation necessarily relies on the equation of continuity, which in our case takes the form [14]:

$$\frac{\partial S}{\partial t} + \nabla \cdot \mathbf{J}^s = \mathcal{T} + \int d^3k \frac{df}{dt} \langle \hat{s} \rangle. \tag{1}$$

The terms on the LHS represent the spin density and current, while the last term in the equation takes into account collisions, where $\frac{df}{dt}$ may be modeled by a relaxation time approximation or expressed in terms of collision integrals. We specialize henceforth in homogeneous systems, in which the divergence of the spin current in (1) will be zero. The first term on the RHS, which accounts for spin generation in the absence of collisions, is the focus of this paper. This term, which we shall call the torque density, exists due to the fact that spin is in general not conserved. As discussed in [14], this torque density is defined as:

$$\mathcal{T}(\mathbf{r}, t) \equiv \int d^3r_c \int d^3k f(\mathbf{r}_c, \mathbf{k}, t) \langle \hat{\tau} \delta(\hat{\mathbf{r}} - \mathbf{r}) \rangle, \tag{2}$$

in which $\hat{\tau}$ is understood as $\frac{i}{\hbar}[\hat{H}, \hat{s}]$, $\hat{H}$ is the Hamiltonian, and $\hat{\mathbf{r}}$ is the position operator. In homogeneous systems the torque density simplifies to

$$\mathcal{T} = \int d^3k f \langle \hat{\tau} \rangle, \tag{3}$$

which will be referred to as the *spin generation term*. The fact that we are considering homogeneous systems also implies we can regard the wavepackets as being wide in real space, thus sharp in k-space, and evaluate the expectation value $\langle \hat{\tau} \rangle$ using Bloch wavefunctions. It should be pointed out that, although we arrive at our results semiclassically, one does not require a local description to obtain them, and they can be found using, for example, a Kubo formula approach (see below).

We will treat in what follows a system in which only a constant uniform electric field is present. By means of a gauge transformation, this electric field can be included in the Hamiltonian through the vector potential $\mathbf{A}(\mathbf{r}, \mathrm{t})$ only. This results in a nonadiabatic mixing of the bands, so that the Bloch wavefunctions $|u_i\rangle$ have the following form:

$$|u_i\rangle = |\phi_i\rangle - \sum_{j \neq i} \frac{\langle \phi_j | i\hbar \frac{d}{dt} | \phi_i \rangle}{\epsilon_i - \epsilon_j} |\phi_j\rangle, \tag{4}$$

where the $|\phi_i\rangle$ are the unperturbed Bloch eigenstates. The only time dependence comes from the fact that $\mathbf{k}$ drifts under the action of the electric field, as given by the equation [27]:

$$\hbar \dot{\mathbf{k}}_c = -e\mathbf{E}. \tag{5}$$

Therefore it is legitimate to replace $\frac{d}{dt} = -\frac{e\mathbf{E}}{\hbar} \cdot \frac{\partial}{\partial \mathbf{k}}$ in (4). In this way, the wavefunctions $|u_i\rangle$ depend on the electric field through a reactive term, in other words the field induces a change in the wavefunctions at each $\mathbf{k}$. The $|u_i\rangle$ form a complete set.

In the limit of wide wavepackets, it is easy to prove, starting from Eq. (4), that $\langle \hat{\tau} \rangle$ evaluated in the $\{|u_i\rangle\}$ basis is equal to $\frac{d\langle \hat{s} \rangle}{dt}$, where $\langle \hat{s} \rangle$ is evaluated in the unperturbed $\{|\phi_i\rangle\}$ basis. The former approach is equivalent to using the Kubo formula to find the response of $\hat{\tau}$ to an electric field. Following this line of thought, we find that the spin generation term is always first order in the electric field, and is given by $-\frac{e}{\hbar}\mathbf{E} \cdot \frac{\partial \langle \hat{s} \rangle}{\partial \mathbf{k}}$. To first order in the electric field, we may thus replace f by its equilibrium value f_0. We shall refer to such terms, which depend only on the equilibrium value of the distribution function, as *intrinsic*, as opposed to *extrinsic* terms, depending on the nonequilibrium distribution.

We note that the torque term must be present even in a clean system if the Hamiltonian contains spin-nonconserving terms. In the presence of scattering mechanisms, the *intrinsic* spin *generation* term is balanced by the *extrinsic* spin *relaxation* term so that a net spin polarization can be reached in the steady state. As mentioned previously, the spin generation rate and relaxation time can be measured separately [18]. In addition, in the systems we consider, we assume scattering is strong enough to keep the distribution function near equilibrium and the scattering time small, but not strong enough to make interband mixing important.

In order to clarify the examples below, some words are in order regarding the spin-orbit interaction and asym-

metry. The terms in the spin-orbit interaction which are odd in k rely upon the inversion asymmetry of the system under study. This asymmetry can be of two kinds, depending on the dimensionality of the system. In three dimensions, the inversion asymmetry is a property of the underlying material, and is referred to as bulk inversion asymmetry (BIA). In two dimensions, an asymmetric confinement potential can provide an additional source of inversion asymmetry, known as structure inversion asymmetry (SIA). Moreover, the application of strain along a particular direction further reduces the symmetry of the structure, with important consequences which will be examined below.

We begin with a study of the Rashba Hamiltonian. This Hamiltonian describes, based on symmetry arguments, the SIA of two dimensional structures and is usually the dominant source of spin-orbit coupling. The effective Hamiltonian has the form:

$$H = \frac{\hbar^2 k^2}{2m} + \alpha(\sigma \times \mathbf{k}) \cdot \hat{\mathbf{z}}, \tag{6}$$

in which α is the spin orbit constant and σ is the vector of Pauli spin matrices. α has the form $a_{46}E_z$, where a_{46} is a material specific parameter while E_z is the magnitude of the electric field in the z direction. Therefore, the magnitude of the Rashba interaction can be tuned by an external gate voltage by an amount which has been shown to be as much as 50%[28]. It is customary to view the term multiplying the spin as a momentum dependent effective magnetic field [14] in which the spin precesses. In the absence of an external magnetic field, the bands in the Rashba model are degenerate at $\mathbf{k} = 0$, and each band contains the same number of spin up and spin down carriers [29]. The Hamiltonian has eigenvalues $\epsilon_\pm = \frac{\hbar^2 k^2}{2m} \pm \alpha k$, which will be labeled by + and - respectively. The spin generation term takes the following form:

$$\langle \hat{\tau} \rangle_\pm = \mp \frac{e\mathbf{k}}{2k^3} (\mathbf{k} \times \mathbf{E}) \cdot \hat{\mathbf{z}}, \tag{7}$$

with $\langle \hat{\tau} \rangle$ defined in Eq.(3). Interestingly, it does not depend on the spin-orbit constant. However, the total torque term, summed over the two bands, depends on the difference in Fermi wavevectors, which is proportional to the spin-orbit constant. We find that

$$\mathcal{T} = \frac{e\alpha m}{4\pi\hbar^2} \mathbf{E} \times \hat{\mathbf{z}}, \tag{8}$$

which vanishes in the limit in which $\alpha \to 0$, is in agreement with [21].

Using symmetry arguments, we find that spin-orbit in the conduction band of bulk wurtzite structures is also described by a Rashba-type Hamiltonian, with a spin-orbit constant defined analogously. This conclusion is supported by [30] based on group theory arguments. The only terms allowed by symmetry are $\beta(\sigma_x k_y - \sigma_y k_x)$, and the Hamiltonian is:

$$H = \frac{\hbar^2 k^2}{2m} + \beta(\sigma_x k_y - \sigma_y k_x) \tag{9}$$

with eigenvalues (labeled as before) $\frac{\hbar^2 k^2}{2m} \pm \beta k$. The spin generation term has a form very similar to (7):

$$\langle \hat{\tau} \rangle_\pm = \mp \frac{e\mathbf{k}_\perp}{2k_\perp^3} (\mathbf{k}_\perp \times \mathbf{E}) \cdot \hat{\mathbf{z}} \tag{10}$$

In the above, $\mathbf{k}_\perp = (k_x, k_y, 0)$. The total torque term is:

$$\mathcal{T} = \frac{em^2\beta}{8\pi\hbar^4} \sqrt{\beta^2 + \frac{2\varepsilon_F \hbar^2}{m}} \mathbf{E} \times \hat{\mathbf{z}} \tag{11}$$

which again vanishes as $\beta \to 0$.

Finally, we turn our attention to the conduction band of zincblende semiconductors, which has been the focus of experiment in recent months [18]. In order to simulate the experiment, we consider an n-doped $In_xGa_{1-x}As$ heterostructure grown on GaAs, with x=0.07, with a strain of 0.046% directed along (001) and $n = 3 \times 10^{16}\text{cm}^{-3}$, as given in [18] and references therein. The lattice constants in the x and y directions remain unaltered. The symmetry of zincblende does not allow terms linear in k in the conduction band in the bulk. As a result, when strain is applied these linear in k terms will be first order in the strain. This makes them much smaller than the cubic terms already present in the bulk. We therefore neglect the effect of these terms in this calculation, although we take into account the effect of strain on the effective masses. We will take into account only the spin-orbit terms cubic in k which are present in the bulk [29], namely $\lambda\sigma_x k_x(k_y^2 - k_z^2) + c.p.$, where λ is the spin-orbit constant and c.p. stands for cubic permutations. The Hamiltonian for this system is:

$$H = \frac{\hbar^2 k_\perp^2}{2m_\perp} + \frac{\hbar^2 k_z^2}{2m_z} + \lambda\sigma_x k_x(k_y^2 - k_z^2) + c.p., \tag{12}$$

where $\mathbf{k}_\perp$ has been defined above. It has eigenvalues $\varepsilon_\pm = \frac{\hbar^2 k_\perp^2}{2m_\perp} + \frac{\hbar^2 k_z^2}{2m_z} \pm \lambda\Delta$ (labeled as before), with Δ given by $\sqrt{[k_x^2(k_y^2 - k_z^2)^2 + c.p.]}$. In this model the x-component of spin takes the form:

$$\langle \hat{s}^x \rangle_\pm = \frac{\hbar k_x(k_y^2 - k_z^2)}{2\Delta}, \tag{13}$$

with the other components given by the cubic permutations. The x-component of the spin generation term is:

$$\langle \hat{\tau}^x \rangle_\pm = \mp \frac{eE_x(k_y^2 - k_z^2)(k_y^2 k_z^4 + k_z^2 k_y^4 - k_x^4 k_y^2 - k_x^4 k_z^2)}{2\Delta^3}. \tag{14}$$

Again the other components can be found by cubic permutation. Note that, if strain were absent so that the effective mass would be isotropic, the bandstructure and thus the equilibrium distribution would have cubic symmetry, making $\langle\hat{\tau}\rangle$ zero after integration over wavevector. To facilitate comparison with experiment, it is more instructive to examine the *electric torque response tensor*, which we define through the equation $\mathcal{T}_i = \chi^{\tau}_{ij} E_j$. Only $\chi^{\tau}_{xx} = -\chi^{\tau}_{yy}$ have finite values, the other components being zero. In contrast to the Rashba model, the diagonal components of the tensor are finite, whereas the off diagonals vanish. To obtain an explicit expression for the total spin torque, we must integrate over the Fermi surface, which in this case is ellipsoidal. We use the equilibrium distribution and the fact that $f(\varepsilon \pm \lambda\Delta) = f(\varepsilon) \pm \lambda\Delta \frac{\partial f(\varepsilon)}{\partial \varepsilon}$, with $\varepsilon = \frac{\hbar^2 k_{\perp}^2}{2m_{\perp}} + \frac{\hbar^2 k_z^2}{2m_z}$ and $\frac{\partial f(\varepsilon)}{\partial \varepsilon} = \delta(\varepsilon - \varepsilon_F)$. We find $\frac{m_z}{m_{\perp}} = 1.023$ [31], and the total result is:

$$\chi^{\tau}_{xx} = -\frac{e\lambda\mathcal{I}}{16\pi^3}\left(\frac{2m_{\perp}}{\hbar^2}\right)^{5/2}\varepsilon_F^{3/2}, \qquad (15)$$

where $\mathcal{I}$ is a dimensionless integral. Using a Monte Carlo integration method, we find that $\mathcal{I}$=-0.03.

We compare our predictions with the experiment of Kato *et al* [18]. In the experimental setup, the electric field is applied first along $(1\bar{1}0)$, which we shall call x', then along (110), which we shall call y'. The spin polarization generated along y' is measured, and is found to be significant in size in the former setup and very small in the latter. Our results agree with these findings. Namely, with the electric field along x' we find $\chi^{\tau}_{y'x'} = 4.3 \times 10^{-10}\mathrm{c/m}^2$, while with the field along y' we obtain $\chi^{\tau}_{y'y'} = 0$. $\chi^{\tau}_{y'x'}$ agrees in sign with the experimental value, but is one order of magnitude higher. A possible explanation of this discrepancy, which the authors of [18] mention, is the fact that in the experiment the strain in the z direction may not be uniform due to strain relaxation. The thickness of the film is 500nm, which is rather large and it is possible that only a small part of the film in the vicinity of the substrate is strained. In addition we note that in the experiment the spins are electrically generated along y', then rotated by a mT magnetic field into the z direction. The angle of rotation provides information about the spin polarization.

Finally, we would like to mention a novel proposed experimental method. In order to verify the existence of this effect on the scale of a wavepacket, one could use cold atoms to construct an individual wavepacket and measure, for example, the size of $\langle\hat{\tau}\rangle$ for this wavepacket, analogous to the method described in [32].

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