Orbital ac Spin-Hall Effect in the Hopping Regime

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The Rashba and Dresselhaus spin-orbit interactions are both shown to yield the low temperature spin-Hall effect for strongly localized electrons coupled to phonons. A frequency-dependent electric field $\mathbf{E}(\omega)$ generates a spin-polarization current, normal to $\mathbf{E}$, due to interference of hopping paths. At zero temperature the corresponding spin-Hall conductivity is real and is proportional to ω^2. At nonzero temperatures the coupling to the phonons yields an imaginary term proportional to ω. The interference also yields persistent spin currents at thermal equilibrium, at $\mathbf{E} = 0$. The contributions from the Dresselhaus and Rashba interactions to the interference oppose each other.

Introduction.—The possibility to control electron spins by an electric field, due to spin-orbit (SO) interactions, has obvious potential applications. Present-day research has focused on the spin-Hall effect: an in-plane electric field applied on a two-dimensional electron gas creates an in-plane spin-current, flowing in the perpendicular direction. In a two-dimensional electron gas confined to the $x - y$ plane, the spin-current is the flux of electrons with spins polarized along z. The spin-Hall conductivity, defined as the ratio between the spin current and the electric field, takes a universal value [1] in a pure, homogeneous infinite electron gas, but disappears [2] in the presence of arbitrarily small static disorder. Consequently, the spin-Hall effect recently observed in a two-dimensional hole system [3] is apparently due to the sample's edges.

Spin-orbit interactions in two-dimensional electronic systems originate from bulk inversion asymmetry (due to the Dresselhaus [4] term), or from the structural inversion asymmetry of the potential confining the electrons to the plane (the Rashba [5] term). While many of the theoretical studies concentrate on either term [6], it has been noted [7–9] that there is, in fact, a competition between the two terms, implying the intriguing experimental possibility to control the direction of the spin-polarization flow by modifying, e.g., the asymmetry of the confining potential.

Whereas the spin-Hall effect of itinerant electrons in the diffusive regime seems to be well understood, far less has been done concerning its realization in strongly localized electronic systems coupled to a phonon bath [10]. To lowest order, the SO interaction appears as a 2×2 phase-factor matrix multiplying the hopping amplitudes [11,12],

$$\hat{J}_{i\ell} = J_{i\ell} e^{-i\mathbf{d}_{i\ell} \cdot \boldsymbol{\sigma}}. \tag{1}$$

Here, $J_{i\ell} = J_{\ell i}$ is the overlap of two wave functions localized at sites i and ℓ (which can be chosen to be real), $\boldsymbol{\sigma}$ is

the vector of Pauli matrices, and the vector $\mathbf{d}_{i\ell} = -\mathbf{d}_{\ell i}$ is calculated below for the combined Rashba and Dresselhaus interactions. Therefore, the spin-Hall effect in insulators is due to *interference* of hopping paths, reminiscing the origin of the ordinary Hall effect in insulators [13]. This interference also leads, as we show below, to the appearance of "persistent spin currents," flowing at thermal equilibrium in the absence of any external fields, and surviving the coupling to the phonon bath—again in analogy with the ordinary Aharonov-Bohm persistent charge current in the hopping regime [14]. However, this spin-persistent current does not cause any local spin accumulation [15]. To achieve the latter accumulation, and to establish a spin-Hall polarization, a frequency-dependent electric field, $\mathbf{E}(\omega)$, is required. Below we calculate the spin-Hall-conductivity matrix of a three-site triad (the smallest cluster of localized electronic sites in which the SO interaction is manifested), and show that it consists of off-diagonal matrix elements alone, such that the spin-current density in the plane, $\mathbf{j}^{\mathrm{P}}(\omega)$, always flows normally to the field,

$$\mathbf{j}^{\,\mathrm{P}}(\omega) = \sigma_{\mathrm{Hall}}^{\mathrm{P}}(\omega) \begin{bmatrix} 0 & 1 \\ -1 & 0 \end{bmatrix} \mathbf{E}(\omega). \tag{2}$$

At zero temperature and low ω, we find that the spin-Hall conductivity, $\sigma_{\mathrm{Hall}}^{\mathrm{P}}$, is real and quadratic in ω, and is robust against the self-averaging effect of our highly disordered system. As the temperature is increased, the coupling to the thermal bath comes into play, yielding an imaginary term in $\sigma_{\mathrm{Hall}}^{\mathrm{P}}$ which results from dissipation and is linear in ω.

As is the case for itinerant electrons [7–9], the Rashba and the Dresselhaus terms work one against the other: the interference phase induced by the SO interaction is found to be proportional to the difference between the two respective contributions. This opens the possibility to reverse

the sense of the equilibrium persistent spin current, as well as the direction of the spin polarization induced by an external electric field, in the strongly localized regime.

Spin-orbit interaction in the hopping regime.—The combined Dresselhaus and Rashba SO interactions in a two-dimensional electron gas may be written in the form

$$\mathcal{H}_{\text{SO}} = \mathbf{U}_p \cdot \boldsymbol{\sigma}, \tag{3}$$

where $\mathbf{U}_p = [\alpha_D p_x + \alpha_R p_y, -(\alpha_R p_x + \alpha_D p_y)]$ is a two-dimensional vector, consisting of the in-plane momentum components, and the SO coefficients α_D (for the Dresselhaus term) and α_R (for the Rashba one). To lowest order in the coefficients $\alpha_{D,R}$, the hopping matrix elements between two states localized around sites i and ℓ are given by Eq. (1), with

$$\mathbf{d}_{i\ell} = [p_D R_{i\ell}^x + p_R R_{i\ell}^y, -(p_R R_{i\ell}^x + p_D R_{i\ell}^y)], \tag{4}$$

where $p_{D,R} = m\alpha_{D,R}$ (m is the electronic mass, and $\hbar$ is taken as 1) and $\mathbf{R}_{i\ell} = \mathbf{R}_i - \mathbf{R}_\ell$ is the difference of the radius vectors of sites i and ℓ [16].

Transport properties in the hopping regime are customarily obtained as an expansion in the hopping amplitudes, since the strong localization regime is characterized [17] by $|J_{i\ell}| \ll |\epsilon_{i\ell}|$, where $\epsilon_{i\ell} \equiv \epsilon_i - \epsilon_\ell$, ϵ_i being the single-particle energies of the localized states (assumed to be randomly distributed). The single-bond conductance ($\propto \text{Tr}\{\hat{J}_{i\ell}\hat{J}_{\ell i}\}$, where the trace is in spin space) is then independent of the SO coupling. Spin-orbit interactions affect the hopping at third order in the $\hat{J}$'s (and beyond). At third order, one needs to consider the interference between the direct hopping path $i \rightarrow \ell$ and the indirect one, $i \rightarrow m \rightarrow \ell$. To second order in the SO coefficients, the latter path involves

$$\hat{J}_{\ell m}\hat{J}_{mi} \approx J_{\ell m}J_{mi}\left(1 - \frac{1}{2}|\mathbf{d}_{i\ell}|^2 - i[\mathbf{d}_{\ell i} + \mathbf{d}_{\ell m} \times \mathbf{d}_{mi}] \cdot \boldsymbol{\sigma}\right). \tag{5}$$

At third order in the $\hat{J}$'s, the interference contribution to the *conductance* comes from traces like $\text{Tr}\{\hat{J}_{i\ell}\hat{J}_{\ell m}\hat{J}_{mi}\}$. However, simple algebra shows that, to second order in $\alpha_{D,R}$, this trace is independent of the SO terms. In contrast, the interference terms *do* produce a current of spin polarization, which results from the SO interactions and is quadratic in the $\mathbf{d}$'s. As discussed below, this spin-polarization current requires the calculation of traces like

$$\boldsymbol{J}_{i\ell m} \equiv -i \text{Tr}\{\boldsymbol{\sigma}\hat{J}_{i\ell}\hat{J}_{\ell m}\hat{J}_{mi}\}. \tag{6}$$

The cornerstone of all our results is the observation that

$$\boldsymbol{J}_{i\ell m} = 4J_{i\ell}J_{\ell m}J_{mi}(p_D^2 - p_R^2)\boldsymbol{\mathcal{A}}, \tag{7}$$

where the vector $\boldsymbol{\mathcal{A}} = \mathbf{R}_{mi} \times \mathbf{R}_{i\ell}/2$ is along the z direction, and $|\boldsymbol{\mathcal{A}}|$ is equal to the area spanned by the three sites, i, ℓ, and m, in the $x - y$ plane. It is also seen that the two

SO interactions considered here have opposite effects on the interference phase.

Spin polarization and spin-polarization current density in the hopping regime.—In our calculations we employ the usual electron-phonon Hamiltonian pertaining to electrons in the hopping regime, $\mathcal{H} = \mathcal{H}_0 + \mathcal{V}$, in which

$$\mathcal{H}_0 = \sum_{n\sigma} \epsilon_n n_{n\sigma} + \sum_{\mathbf{q}} \omega_q b_{\mathbf{q}}^\dagger b_{\mathbf{q}}, \; n_{n\sigma} = c_{n\sigma}^\dagger c_{n\sigma}, \tag{8}$$

where $c_{n\sigma}^\dagger$ ($c_{n\sigma}$) creates (destroys) an electron of spin index σ on the state localized at site n, ω_q is the phonon frequency, and $b_{\mathbf{q}}^\dagger$, $b_{\mathbf{q}}$ are the phonon operators. The Hamiltonian $\mathcal{V}$ includes the hopping, the coupling to the phonons [14], and the external electric field,

$$\mathcal{V} = \sum_{nn'} \sum_{\sigma\sigma'} \hat{J}_{nn'}^{\sigma\sigma'} Q_{nn'} e^{i\Phi_{nn'}(t)} c_{n\sigma}^\dagger c_{n'\sigma'}, \tag{9}$$

where the spin-dependent hopping is defined in Eq. (1),

$$Q_{nn'} = \exp\left[\sum_{\mathbf{q}} \frac{v_{\mathbf{q}}^{nn'}}{\omega_q}(b_{-\mathbf{q}}^\dagger - b_{\mathbf{q}})\right] \tag{10}$$

yields the effect of an on-site electron-phonon coupling ($v_{\mathbf{q}}^{nn'} = v_{\mathbf{q}}^n - v_{\mathbf{q}}^{n'}$, with $v_{\mathbf{q}}^n$ representing the electron-phonon coupling on site n) on the hopping, and $\Phi_{nn'}$ is due to an in-plane external electric field, with $i\omega\Phi_{nn'}(\omega) = e\mathbf{E} \cdot \mathbf{R}_{nn'}$ [18].

The z component of the spin-polarization operator at site n is given by $P_n = \text{Tr}\{\sigma^z n_{n\sigma}\}$. The thermal average of its temporal derivative, $\langle dP_n/dt \rangle \equiv \langle i[\mathcal{H}, P_n]\rangle$ (assuming that in the Hamiltonian $\mathcal{H}$ each site is in contact with a grand-canonical electron reservoir [13,14]), up to first order in the electric field, is

$$\langle dP_n/dt \rangle = -\sum_{n'} \sum_{\sigma\sigma'} \sigma_{\sigma\sigma}^z \langle I_{nn'}^{\sigma\sigma'} - I_{n'n}^{\sigma'\sigma} - i\Phi_{nn'}(t)(I_{nn'}^{\sigma\sigma'} + I_{n'n}^{\sigma'\sigma})\rangle, \tag{11}$$

with

$$I_{nn'}^{\sigma\sigma'} = iQ_{nn'} c_{n\sigma}^\dagger \hat{J}_{nn'}^{\sigma\sigma'} c_{n'\sigma'}. \tag{12}$$

We calculate $\langle dP_n/dt \rangle$ to third order in the hopping. Detailed calculations [19] then show that $\langle I_{nn'}^{\sigma\sigma'} + I_{n'n}^{\sigma'\sigma}\rangle$ vanishes, so that the entire contribution to the polarization rate comes from $\langle I_{nn'}^{\sigma\sigma'} - I_{n'n}^{\sigma'\sigma}\rangle$, which will be found up to first-order in the electric field. The polarization rate at each site is thus given by a sum over the bond spin currents, in a complete analogy with the rate of the site (charge) occupation in the hopping regime [13,14]. This parallelness continues further, since (as is shown below) $\Sigma_n \langle dP_n/dt \rangle = 0$, and therefore SO interactions in conjunction with an electric field *do not* lead to spin-polarization accumulation. This enables us to define the spin-polarization current density, $\mathbf{j}^P$, as the rate of change of the corresponding

dipole moment [13],

$$\mathbf{j}^P = \frac{1}{S}\frac{d}{dt}\sum_n \langle P_n \rangle \mathbf{R}_n = \frac{1}{S}\sum_n \left\langle \frac{dP_n}{dt} \right\rangle \mathbf{R}_n. \qquad (13)$$

Here S denotes the area of the system. Below we concentrate on $\mathbf{j}^P$ of a single triad, and therefore this area is replaced by $|\mathcal{A}|$. Note that this definition is independent of the choice of the coordinate origin, since $\Sigma_n \langle dP_n/dt \rangle = 0$.

Persistent spin currents.—The thermal average of the spin current in a single bond, $\langle I_{nn'}^{\sigma\sigma'} - I_{n'n}^{\sigma'\sigma} \rangle$, is nonzero *even when the electric field is absent.* This is a consequence of the trace Eq. (6), which contains $\boldsymbol{\sigma}$: since the SO interaction conserves time-reversal symmetry, it cannot produce persistent charge currents, but it *does* lead to persistent spin current. Indeed, ignoring the electron-phonon coupling, and using third-order perturbation theory in the hopping matrix elements, the contribution of the bond $i - \ell$ in our triad (at $\mathbf{E} = 0$) becomes

$$\sum_{\sigma\sigma'}\sigma_{\sigma\sigma}^z \langle I_{\ell i}^{\sigma'\sigma} - I_{i\ell}^{\sigma\sigma'} \rangle|_{\text{eq}} = 2J_{i\ell m}^z \sum_{\text{per}}\frac{f_i}{\epsilon_{i\ell}\epsilon_{im}}, \qquad (14)$$

where f_i is the Fermi function of the site i occupation, and Σ_{per} stands for the sum over the three permutations $i, \ell, m \to \ell, m, i \to m, i, \ell$.

Hence, there is a persistent spin current flowing around the triad at equilibrium, even at zero temperature, with its sense being determined by the relative locations of the site energies compared to the Fermi level. This finding is in a complete analogy with the persistent charge current flowing in response to an Aharonov-Bohm phase [14]. The analogy continues when the coupling to the phonon bath is switched on: that coupling induces a Debye-Waller factor multiplying the result (14), *and also* a "counter" spin-persistent current, which flows in the opposite direction to the current (14), and vanishes at zero temperature (for details, see Ref. [14]). This equilibrium persistent current does not lead to spin accumulation: the current flowing from i to ℓ is minus the one flowing from i to m. [When ℓ is interchanged with m, the sign of $\mathcal{A}$ is reversed; see Eq. (7).] Consequently $\langle dP_i/dt \rangle|_{\text{eq}}$ vanishes at each site (an analogous calculation shows that at equilibrium, $\langle P_i \rangle|_{\text{eq}}$ vanishes as well). Equilibrium spin currents which do not lead to spin accumulation exist also in conductors lacking inversion symmetry [15]. The question whether they are amenable to an experimental detection (like the charge persistent currents) in small coherent mesoscopic structures is left open.

Spin-Hall effect in the hopping regime.—When an in-plane electric field is applied, the polarization rate $\langle dP_i/dt \rangle$ is nonzero. We first consider it for our triad in the absence of the coupling to the phonon bath, employing linear response theory (with respect to the field),

$$\langle dP_i/dt \rangle(\omega) \equiv \sum_{\sigma\sigma'}\sigma_{\sigma\sigma}^z \langle I_{\ell i}^{\sigma'\sigma}(\omega) - I_{i\ell}^{\sigma\sigma'}(\omega) + I_{mi}^{\sigma'\sigma}(\omega)$$

$$- I_{im}^{\sigma\sigma'}(\omega)\rangle$$

$$= \omega^2 \Phi_{\ell m}(\omega)\frac{2J_{i\ell m}^z}{\epsilon_{i\ell}\epsilon_{\ell m}\epsilon_{mi}}\sum_{\text{per}}\frac{f_i - f_\ell}{\omega^2 - \epsilon_{i\ell}^2}.$$

$$(15)$$

Namely, the average polarization rate at site i is driven by the potential difference across the bond $\ell - m$, and consequently $\Sigma_n \langle dP_n/dt \rangle = 0$. Inserting Eq. (15) into Eq. (13) produces Eq. (2), with the spin-Hall conductivity, σ_{Hall}^P, given by

$$\sigma_{\text{Hall}}^P(\omega) = 16e\omega^2(p_R^2 - p_D^2)|\mathcal{A}|\Gamma_{i\ell m}^e(\omega), \qquad (16)$$

where

$$\Gamma_{i\ell m}^e(\omega) = \frac{J_{i\ell}J_{\ell m}J_{mi}}{\epsilon_{i\ell}\epsilon_{\ell m}\epsilon_{mi}}\sum_{\text{per}}\frac{f_i - f_\ell}{\epsilon_{i\ell}^2 - \omega^2}. \qquad (17)$$

In deriving this result, we have discarded resonance transitions in which the frequency ω compensates the site energy differences. Otherwise, the sum in Eq. (17) would have been augmented by terms of the type $i(f_i - f_\ell)\delta(\epsilon_{i\ell} \pm \omega)/|\omega|$. We will find below that the electron-phonon coupling gives rise to an imaginary part in the spin-Hall conductivity, that is of the same order in ω, and which originates from energy-conserving delta functions of the type $\delta(\epsilon_{i\ell} \pm \omega_q \pm \omega)$. Since the latter are more likely to be satisfied for the randomly distributed site energies, the present discussion is confined to the result (16).

The result (2) demonstrates in a nutshell the origin of the present-day interest in the spin-Hall effect. While an electric field along, say, the x direction, will drive a charge current in the same direction, the spin-current density in this case will be along the y direction, leading to a "separation" of spin and charge.

The spin-Hall conductivity (16) vanishes at small frequencies and tends to zero as ω^{-2} at very high ones [since when $\epsilon_{i\ell}^2 \ll \omega^2$ the sum in Eq. (17) vanishes]. Thus, a dc electric field is incapable of producing the spin-Hall effect. This is in accordance with the behavior found in itinerant electron systems [2]. This conductivity, which depends upon the temperature through the Fermi functions, remains finite at zero temperature when two of the three-site energies are below or above the Fermi energy. Then, at frequencies smaller than the typical site energy differences, and assuming that all the hopping matrix elements $J_{i\ell}$ have the same sign [12], $\Gamma_{i\ell m}^e$ takes a definite sign, independent of the locations of the site energies relative to the Fermi level. Consequently, the *self-averaging* effect of the macroscopic hopping system, which involves many triads, *will not wash out* σ_{Hall}^P.

The effect of the coupling to the phonons on the above results may be divided into three. First, there is the overall Debye-Waller exponent, due to loss of coherence. This

factor exists even at zero temperature. (For brevity, it will not be presented below.) Second, virtual electron-phonon processes contribute additional terms to $\Gamma^{e}_{i\ell m}$, which are proportional to the electron-phonon coupling and involve complicated combinations of site Fermi functions and Bose occupation numbers of the phonons. We do not present these terms since in the weak electron-phonon coupling they are smaller than the purely electronic ones given in Eq. (17). Third and most importantly, real (energy-conserving) electron-phonon transitions give rise to an imaginary part in the Hall conductivity, which now becomes

$$\sigma^{P}_{Hall}(\omega) = 16e(p_R^2 - p_D^2)|\mathcal{A}|[\omega^2 \Gamma^{e}_{i\ell m}(\omega) - i\Gamma^{ep}_{i\ell m}(\omega)] \quad (18)$$

with

$$\Gamma^{ep}_{i\ell m}(\omega) = \frac{\sinh(\beta\omega)}{2\beta} \sum_{per} \left(\frac{1}{\epsilon_{\ell m}^2} + \frac{1}{\epsilon_{mi}^2} \right) \frac{G_{i\ell}}{e^2} \frac{J_{\ell m}J_{mi}}{J_{i\ell}}, \quad (19)$$

where $G_{i\ell} \propto \exp[-\beta(|\epsilon_{i\ell}| + |\epsilon_i| + |\epsilon_\ell|)/2]$ is the usual temperature-dependent hopping conductance of the $i - \ell$ bond, and β is the inverse temperature. The result (19) is derived in the small ω limit, assuming the frequency to be smaller than the site energy differences. Hence, Γ^{ep} vanishes at zero temperature. We note that Γ^{ep} has again a definite sign, and hence will survive self-averaging.

To obtain the temperature dependence of the Hall conductivity (18) at low frequencies, we consider the situation in which the leading electrical conductance of our triad takes place along the bond $i - \ell$, and site m provides the interference path necessary for the spin-Hall effect. Thus we imagine ϵ_i and ϵ_ℓ to be below and above the Fermi level, but close to it, while ϵ_m lies far away from the Fermi energy. In that case $\Gamma^{e}_{i\ell m} \sim J_{i\ell}J_{\ell m}J_{mi}/|\epsilon_{i\ell}^3|\epsilon_m^2$, while $\Gamma^{ep}_{i\ell m} \sim \Gamma^{e}_{i\ell m}\gamma_{i\ell} \sinh(\beta\omega)|\epsilon_{i\ell}| \exp(-\beta|\epsilon_{i\ell}|)$, where $\gamma_{i\ell} = |v|^2 \mathcal{N}(|\epsilon_{i\ell}|)$ is the density of phonon states at $|\epsilon_{i\ell}|$ multiplied by the electron-phonon matrix element squared. It then turns out that one may define a characteristic frequency of the system [18],

$$\nu_{i\ell}(T) = \gamma_{i\ell}\beta|\epsilon_{i\ell}|e^{-\beta|\epsilon_{i\ell}|}, \quad (20)$$

which vanishes at zero temperature. For $\beta\omega < 1$ the spin-Hall conductivity is then

$$\sigma^{P}_{Hall}(\omega) \propto \omega[\omega - i\nu_{i\ell}(T)]. \quad (21)$$

At very low temperature, this conductivity is proportional to ω^2; as the temperature is increased, real electron-phonon processes give rise to an imaginary part due to dissipation, which is linear in ω.

In conclusion, we have derived the spin-Hall conductivity of a triad of localized sites, and have shown that, in the small frequency limit, it has a definite sign and is therefore expected to reflect the spin-Hall conductivity of the macroscopic system. Together with the competition between the Rashba and the Dresselhaus SO interactions, this opens the possibility to control the magnitude and the phase (compared to the driving ac field) of the spin-Hall current in the hopping regime.

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