Electrical Spin Orientation, Spin-Galvanic, and Spin-Hall Effects in Disordered Two-Dimensional Systems

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In disordered systems, the hopping conductivity regime is usually realized at low temperatures where spin-related phenomena differ strongly from the cases of delocalized carriers. We develop the unified microscopic theory of current-induced spin orientation, spin-galvanic, and spin-Hall effects for the two-dimensional hopping regime. We show that the corresponding susceptibilities are proportional to each other and determined by the interplay between the drift and the diffusion spin currents. Estimations are made for realistic semiconductor heterostructures using the percolation theory. We show that the electrical spin polarization in the hopping regime increases exponentially with the increase of the concentration of localization sites and may reach a few percent at the crossover from the hopping to the diffusion conductivity regime.

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Introduction.—Spin physics is a rapidly growing area of research in condensed matter science aimed at the creation, manipulation, and detection of spins in various systems [1]. Important and fundamentally interesting results, also promising for possible future applications, have been obtained in semiconductors and semiconductor nanostructures [2]. The cornerstones in semiconductor spintronics are spin orientation, spin transfer, and spin readout. Remarkable progress has been achieved in the last decade's experiments in all three directions including ultrafast optical spin injection [3,4], low-dissipation spin current manipulation [5], and nearly nondestructive spin measurements [6]. A challenging problem in the spin physics is how to affect the spin by instantaneous nonmagnetic methods, in particular, by electric fields [7,8]. The key to the electrical spin control is the spin-orbit interaction [9], which linearly couples spin and momentum components of carriers. It allows for the current-induced spin polarization (CISP) — a phenomenon where the electric current flow is accompanied by a homogeneous orientation of carrier spins. This problem is mostly studied in semiconductors, see Ref. [10] for review. Recent progress in the field is related to precise electrical control of spin in semiconductor epilayers [11,12]. The problem of CISP in two-dimensional (2D) semiconductor heterostructures is investigated theoretically in detail, including nonlinear regimes of CISP [13,14].

There are two more phenomena closely related to CISP. The first one is a generation of an electric current in systems with a nonequilibrium spin polarization referred to as the spin-galvanic effect (SGE) [15]. SGE has been studied in various 2D semiconductor systems where nonequilibrium spin polarization has been created by means of optical excitation [16]. One more phenomenon is the spin-Hall effect (SHE) consisting in a generation of the spin current in the presence of the electric current [2]. All three effects are phenomenologically introduced as follows:

$$\boldsymbol{s} = \hat{\boldsymbol{\sigma}}_{\rm CISP}\boldsymbol{E}, \qquad \boldsymbol{j} = \hat{\boldsymbol{\sigma}}_{\rm SGE}\boldsymbol{s}, \qquad \boldsymbol{\mathcal{J}} = \hat{\boldsymbol{\sigma}}_{\rm SHE}\boldsymbol{E}. \quad (1)$$

Here, $\boldsymbol{s}$ is the nonequilibrium spin polarization, $\boldsymbol{E}$ and $\boldsymbol{j}$ are the electric field and electric current density, and $\boldsymbol{\mathcal{J}}$ is the component of the spin current describing the flux density of spins oriented along the normal to the 2D plane.

Despite a deep investigation of CISP, SGE, and SHE, all previous activities were devoted to delocalized electrons, which weakly feel the static disorder as a source of rare momentum scattering. However, the role of the disorder is drastically enhanced at low temperatures when carriers are localized in minima of potential energy. In contrast to free electron systems, the localized carriers preserve their spin coherence for hundreds of nanoseconds due to suppression of the Dyakonov-Perel spin relaxation mechanism [17]. The record spin coherence times have been demonstrated for semiconductor quantum dot structures [18–20]. For this reason, the spin properties of localized electrons attract rapidly growing attention. Application of an electric field to such systems induces directed hops of electrons between localization sites, so called *hopping conductivity regime*. Spin relaxation [21–24], spin dynamics [25,26], spin noise [27,28], and ac spin-Hall effect [29] have been recently studied in the hopping regime. However, CISP, SGE, nor dc SHE have been considered. In this Letter, we fill this gap and describe the effects of the spin, electric current, and spin current mutual conversion in the hopping regime.

Model.—The effective electron Hamiltonian describing spin-orbit interaction in 2D heterostructures grown along [001] direction has the form

$$\mathcal{H}_{\rm SO} = \beta_{\mu\nu}\sigma_\mu k_\nu = \beta_{xy}\sigma_x k_y + \beta_{yx}\sigma_y k_x. \quad (2)$$

Here, $x\|[1\bar{1}0]$ and $y\|[110]$ are the coordinates in the 2D plane, $\sigma_{x,y}$ are the Pauli matrices, $\boldsymbol{k} = -{\rm i}\boldsymbol{\nabla}$, and $\beta_{\mu\nu}$ are spin-orbit constants caused by both bulk- and

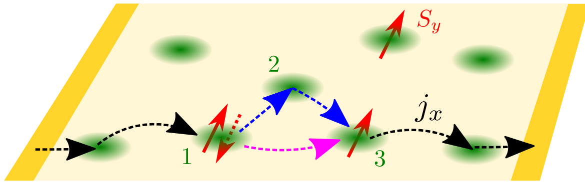

FIG. 1. Illustration of CISP in the 2D hopping regime: electrons rarely hop between localization sites (green areas). In the presence of electric current j_x, the quantum interference between the direct and indirect hopping paths, shown respectively by magenta and blue arrows, leads to spin polarization S_y.

structure-inversion asymmetry [16,30]. We consider a 2D ensemble of electrons localized at random sites in a weak dc electric field, see Fig. 1. The 2D concentration of carriers n is assumed to be much smaller than the concentration of sites n_s. This situation is realized, for example, in ensembles of weakly charged quantum dots or in n-doped quantum wells, compensated by p doping of barriers (D^0 or D^- centers). Note that our theory can be equally applied to the ensembles of holes, but the electron tunneling between the sites is facilitated as compared with holes because the effective mass in the conduction band is, as a rule, smaller than in the valence band.

A microscopic origin of CISP, SGE, and SHE in the hopping regime is the spin-orbit interaction (2). It results in the precession of electron spins during the hops. The electron Hamiltonian in the basis of localized states has the form [29]

$$\mathcal{H}_e = \sum_{i,\sigma} \epsilon_i c^\dagger_{i\sigma} c_{i\sigma} + \sum_{ij} \sum_{\sigma\sigma'} J^{\sigma\sigma'}_{ij} c^\dagger_{i\sigma} c_{j\sigma'}. \tag{3}$$

Here, $c^\dagger_{i\sigma}(c_{i\sigma})$ are the creation (annihilation) operators of an electron at the site i, with the spin projection $\sigma = \pm 1/2$ on the z axis, being the normal to the 2D plane, and we neglect the doubly occupied states, assuming the Hubbard energy to be infinite. The site energies consist of three contributions: $\epsilon_i = E_b + U_i - e\boldsymbol{E}\cdot\boldsymbol{R}_i$, where E_b is the binding energy assumed to be equal for all sites, U_i is the fluctuating electrostatic potential energy at the site, and the last term describes the potential in the external electric field for the site with the 2D coordinate $\boldsymbol{R}_i$. The energies U_i are broadly distributed, and the variable-range hopping regime is realized [31]. The second term in Eq. (3) describes spin-dependent hopping with the amplitudes [17,29,32]

$$\hat{J}_{ij} = J_{ij} e^{-i\boldsymbol{d}_{ij}\cdot\hat{\boldsymbol{\sigma}}}, \qquad \boldsymbol{d}_{ij} = m\hat{\boldsymbol{\beta}}(\boldsymbol{R}_i - \boldsymbol{R}_j)/\hbar^2, \tag{4}$$

where J_{ij} are spin-independent hopping amplitudes between sites i and j, and m is the electron effective mass.

Kinetic equation.—Electron transport in the studied spin-orbit coupled system is described by a kinetic equation for the spin density matrix. Decomposing the on-site density matrix as $\hat{\rho}_i = n_i/2 + \hat{\boldsymbol{\sigma}}\cdot\boldsymbol{S}_i$, we derive a system of coupled equations for the site occupations n_i and the spins $\boldsymbol{S}_i$ [33,34]:

$$\dot{n}_i = \sum_j I_{ij} + \sum_j (\boldsymbol{\Lambda}_{ij}\cdot\boldsymbol{S}_j - \boldsymbol{\Lambda}_{ji}\cdot\boldsymbol{S}_i), \tag{5a}$$

$$\dot{\boldsymbol{S}}_i + \sum_j \boldsymbol{S}_j \times \boldsymbol{\Omega}_{ij} + \frac{\boldsymbol{S}_i}{\tau_s} = \sum_j \boldsymbol{I}^s_{ij} + \sum_j (\boldsymbol{G}_{ij} n_j + \boldsymbol{G}_{ji} n_i). \tag{5b}$$

Here, $I_{ij} = n_j/\tau_{ij} - n_i/\tau_{ji}$ is the particle flow between sites i and j, with τ_{ji} being the hopping time from the site i to the site j. The second sum in Eq. (5a) represents the source of an electric current induced by a nonequilibrium spin polarization being the precursor of SGE.

The left hand side of Eq. (5b) has the form of the Bloch equation, with the effective frequency of spin precession during the hop $\boldsymbol{\Omega}_{ij} = 2\boldsymbol{d}_{ij}/\tau_{ij}$, and the on-site phenomenological spin relaxation time τ_s, caused by the hyperfine interaction. This time is shorter than Dyakonov-Perel spin relaxation time in the hopping regime [45], and for the sake of simplicity, we neglect the possible nonexponential spin relaxation dynamics. The spin current flowing from the site j to the site i, $\boldsymbol{I}^s_{ij}$, is a sum of two contributions

$$\boldsymbol{I}^s_{ij} = \frac{\boldsymbol{S}_j}{\tau_{ij}} - \frac{\boldsymbol{S}_i}{\tau_{ji}} + \boldsymbol{W}_{ij} n_j - \boldsymbol{W}_{ji} n_i. \tag{6}$$

The first two terms describe the spin diffusion, while the latter terms arise due to a difference in spin-conserving tunneling rates for electrons with spins oriented along (↑) and opposite (↓) to the axis α: $W^\alpha_{ij} = (W_{\uparrow\uparrow} - W_{\downarrow\downarrow})/2$. This contribution clearly leads to a spatial separation of electrons with opposite spins in the static electric field, which is a dc SHE.

The last term in Eq. (5b) describes the spin generation. It can be expressed via a difference of spin-flip probabilities during the hops as $G^\alpha_{ij} = (W_{\uparrow\downarrow} - W_{\downarrow\uparrow})/2$. We note that the kinetic coefficient Λ^α_{ij} can also be presented as $2(W_{\uparrow\uparrow} + W_{\downarrow\uparrow} - W_{\downarrow\downarrow} - W_{\uparrow\downarrow})$, thus allowing us to find a fundamental relation between the kinetic coefficients

$$\boldsymbol{\Lambda}_{ij} = 4(\boldsymbol{W}_{ij} - \boldsymbol{G}_{ij}). \tag{7}$$

At the microscopic level, the spin dependence of the tunneling rates appears due to an interference of the direct hopping path with the hopping through an auxiliary site [29,32,46,47]. An arbitrary triad of localization sites is shown in the center of Fig. 1. The matrix element of tunneling between the sites 1 and 3 up to the second order in hopping amplitude is equal to

$$\hat{J}_{31} + \frac{\hat{J}_{32}\hat{J}_{21}}{\Delta E_{12}} = \hat{J}_{31}\left(1 + \frac{J_{32}J_{21}}{J_{31}\Delta E_{12}} e^{i\boldsymbol{d}_{31}\cdot\hat{\boldsymbol{\sigma}}} e^{-i\boldsymbol{d}_{32}\cdot\hat{\boldsymbol{\sigma}}} e^{-i\boldsymbol{d}_{21}\cdot\hat{\boldsymbol{\sigma}}}\right), \tag{8}$$

where ΔE_{12} is the energy difference between states 1 and 2, including the phonon energy. Because of noncommutativity of Pauli matrices, the second term in the brackets is not reduced to a scalar: The electron spin orientation is changed after travel over the closed path. This is due to the Berry curvature [9,48] arising from the inversion symmetry breaking in hopping Hamiltonian [49]. Note that the paths $1 \to 2 \to 3$ and $1 \to 3$ interfere because the same phonons are emitted or absorbed at these paths [34]. As a result, the hopping matrix element is essentially spin dependent. Therefore, the kinetic coefficients $\mathcal{K}_{ij}$ ($\mathcal{K} = \Lambda, G, W$) can be presented as a sum over the auxiliary sites $\mathcal{K}_{ij} = \sum_k \mathcal{K}_{ikj}$, and the relation (7) holds for $\mathcal{K}_{ikj}$ as well.

Microscopic calculation shows that, in accordance with the time reversal symmetry, only doubly resonant hops contribute to dc spin effects [34]. The kinetic coefficients read

$$\boldsymbol{G}_{ikj} = 3Q_{ikj}\boldsymbol{A}_{ikj} \times \hat{\boldsymbol{\beta}}\boldsymbol{R}_{ij}\mathrm{Tr}(\hat{\boldsymbol{\beta}}^2), \tag{9a}$$

$$\boldsymbol{W}_{ikj} = Q_{ikj}\mathrm{Tr}(\hat{\boldsymbol{\beta}}^2)\left(\boldsymbol{A}_{ikj} \times \hat{\boldsymbol{\beta}}(\boldsymbol{R}_{jk} + \boldsymbol{R}_{ik}) - 3\frac{\hbar^2}{m}\boldsymbol{A}_{ikj}\right), \tag{9b}$$

where $\boldsymbol{R}_{ij} = \boldsymbol{R}_i - \boldsymbol{R}_j, \boldsymbol{A}_{ikj} = \boldsymbol{R}_{ki} \times \boldsymbol{R}_{ij}/2$ is the oriented area of the triad, and Q_{ikj} is the constant determined by the hopping times and hopping amplitudes between the sites [34]. We note that the spin separation (W^z_{ikj}) appears in the second order in the spin-orbit interaction, while the spin generation rate and spin galvanic current are cubic in the spin splitting.

Results.—The CISP and SGE can be conveniently related to the spin current $\mathcal{J}$ flowing in the system. Indeed, the electric current leads to the generation of the spin current due to the SHE, Fig. 2(a). Then, the spin current is converted to spin polarization. The effects of mutual spin and spin current conversion were introduced for free electrons in Ref. [42] by Kalevich, Korenev, and Merkulov and can be referred to as the KKM effects [50]. For localized carriers, it is illustrated in Fig. 2(b): in the presence of spin current, spin-up electrons (S_z) and spin-down electrons ($S_{\bar{z}}$) hop in opposite directions and experience spin precession with frequency Ω_{SO} in opposite directions, which leads to spin polarization S_y. Formally, the KKM effect in the hopping regime can be derived from Eq. (5b) by taking the sum over all sites:

$$\boldsymbol{s} = -\frac{2\tau_s m}{n\hbar^2}\boldsymbol{e}_z \times \hat{\boldsymbol{\beta}}\mathcal{J}, \tag{10}$$

where the spin current is defined as [29,51]

$$\mathcal{J} = \frac{1}{2}\sum_{ij}\boldsymbol{R}_{ij}I^{s,z}_{ij}, \tag{11}$$

and $\boldsymbol{e}_z$ is a unit vector along the z axis.

The spin-galvanic effect can be treated in a similar way, see Fig. 3. Spin polarization S_y due to the spin-orbit interaction leads to the spin current $\mathcal{J}$ (inverse KKM effect). In turn, the spin current induces the electric current j_x due to the inverse spin-Hall effect (ISHE). Therefore, both CISP and SGE are intimately related to the spin current and can be decomposed into two steps, SHE + KKM and inverse KKM + inverse SHE, respectively. In fact, CISP and SGE are reciprocal to each other due to time reversal symmetry [52].

The spin current defined by Eq. (11) consists of two contributions [53]: diffusion spin current $\mathcal{J}_{\mathrm{diff}}$ and drift spin current $\mathcal{J}_{\mathrm{dr}}$, which correspond to the two terms of Eq. (6). Since the system under study is strongly inhomogeneous, the drift spin current leads to spin separation in the steady state. This, in turn, induces the diffusion spin current in the opposite direction as shown in Fig. 2(a). Neglecting the spin relaxation, these two contributions completely cancel each other, so $\mathcal{J} = \mathcal{J}_{\mathrm{diff}} + \mathcal{J}_{\mathrm{dr}}$ is zero. The on-site hyperfine-induced spin relaxation diminishes spin separation and upsets the balance; therefore,

$$\mathcal{J} = \frac{1}{\tau_s}\sum_i \boldsymbol{R}_i S^z_i. \tag{12}$$

This expression shows that in the limit of infinite nuclei-induced spin relaxation time, the total spin current

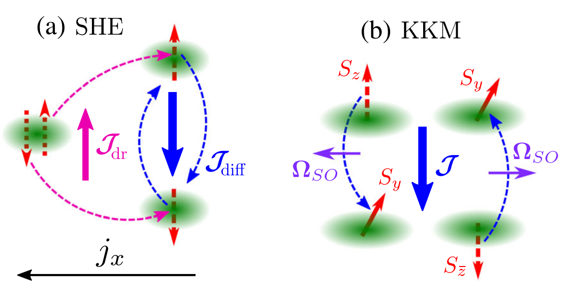

FIG. 2. Microscopic mechanism of CISP. (a) Electric current flow leads to the drift spin current $\mathcal{J}_{\mathrm{dr}}$ due to the SHE. In the strongly inhomogeneous system under study, it is partially compensated by the diffusion spin current $\mathcal{J}_{\mathrm{diff}}$. (b) Spin current $\mathcal{J}$, accompanied by the spin precession in spin-orbit field $\boldsymbol{\Omega}_{SO}$, results in electron spin polarization S_y due to the KKM effect.

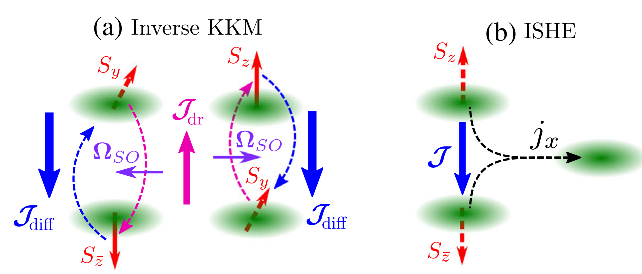

FIG. 3. Mechanism of SGE. (a) Spin polarization leads to the drift spin current $\mathcal{J}_{\mathrm{dr}}$ due to the inverse KKM effect. It is partially compensated by the diffusion spin current, $\mathcal{J}_{\mathrm{diff}}$. (b) Spin current $\mathcal{J}$ results in the electric current j_x due to ISHE.

vanishes. However, the CISP is proportional to the product of the spin relaxation time and the spin current, see Eq. (10), so it always has a finite value. The interplay between drift and diffusion spin currents is illustrated in Fig. 4. Usually, τ_s is much longer than the characteristic hopping time τ_{ij} so the total spin current is less than both $\mathcal{J}_{\rm dr}$ and $\mathcal{J}_{\rm diff}$.

The hopping amplitude exponentially decreases with the increase of a distance between the sites, $J_{ij} \sim \exp(-R_{ij}/a_b)$, where the localization radius a_b is assumed to be the same for all sites. This gives an opportunity to make a quantitative analysis of the spin effects in the hopping regime where $n_s a_b^2 \ll 1$. To that end, we extend the percolation theory [31] to account for spin degrees of freedom. The electric current in the hopping regime flows only in the so-called percolation cluster, where the distances between the sites are the smallest and the potential energies are close to each other. The current-induced spin polarization takes place only in the vicinity of this path. The interference between the hopping paths also drops rapidly down at the distances larger than a_b. Since the electric current is the same in the whole cluster, the main contribution to spin generation is given by the smallest triads of sites having the size $\sim a_b$ [54]. Therefore, the CISP conductivity can be presented as [34]

$$\hat{\boldsymbol{\sigma}}_{\rm CISP} = \tau_s {\rm Tr}(\hat{\boldsymbol{\beta}}^2)\hat{\boldsymbol{\beta}}^T \mathcal{P} f(n_s, \tau_s). \tag{13}$$

Here, $\mathcal{P} = (ma_b/\hbar^2)^3 \hbar n_s a_b/(enJ_0\tau_0\rho)$, ρ is the resistivity, J_0 and τ_0 are the characteristic hopping integral and time for the distance $\sim a_b$, and $f(n_s, \tau_s)$ is a dimensionless function which tends to a finite value as n_s goes to zero.

The spin-galvanic current can be similarly obtained from the kinetic equation (5a). It is generated also in small triads of sites and flows mainly in the percolation cluster. The calculation yields the following result for the SGE response [34]:

$$\hat{\boldsymbol{\sigma}}_{\rm SGE} = 4{\rm Tr}(\hat{\boldsymbol{\beta}}^2)\hat{\boldsymbol{\beta}}^T \mathcal{P} k_B T n f(n_s, \tau_s). \tag{14}$$

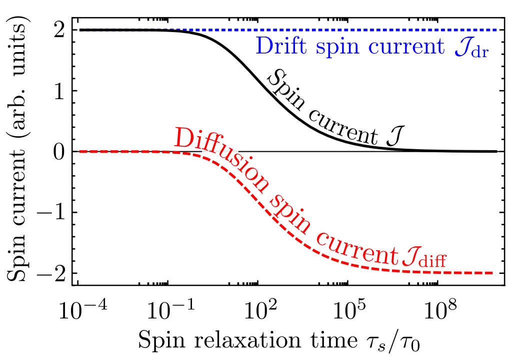

FIG. 4. Drift, diffusion, and total spin currents (in arbitrary units) as functions of hyperfine-induced spin relaxation time for $n_s a_b^2 = 0.01$. The black curve shows the function $f(n_s, \tau_s)$ in absolute units.

Here, the function f coincides with that for CISP, Eq. (13), see Supplemental Material [34]. This coincidence comes from the Onsager relation taking place for CISP and SGE susceptibilities due to reciprocity of these two effects [43,44]. We have analytically calculated the function $f(n_s, \tau_s)$ for the model of a regular triangle [34].

The spin-Hall conductivity can be deduced from Eqs. (10) and (13):

$$\hat{\boldsymbol{\sigma}}_{\rm SHE} = \hat{\boldsymbol{\beta}}^T(\boldsymbol{e}_z \times \hat{\boldsymbol{\beta}})\frac{\hbar^2 n\mathcal{P}}{m} f(n_s, \tau_s). \tag{15}$$

We stress that, in strongly inhomogeneous systems, the drift spin current is always accompanied by the diffusion spin current, and therefore, the spin-Hall conductivity relates the applied electric field to the *total* spin current $\mathcal{J}$. This conductivity vanishes in the absence of hyperfine-induced spin relaxation. However, the spin separation is caused only by the drift spin current, which does not depend on spin relaxation, and therefore, can be found as

$$\mathcal{J}_{\rm dr} = -\hat{\boldsymbol{\beta}}^T(\boldsymbol{e}_z \times \hat{\boldsymbol{\beta}}\boldsymbol{E})\frac{\hbar^2 n\mathcal{P}}{m} f(n_s, 0). \tag{16}$$

The possibility of intrinsic spin current and spin separation for free electrons was intensively debated, for review see, e.g., Refs. [48,55]. It was found that the intrinsic spin-Hall effect is possible at the edges of the sample [56,57] or in mesoscopic systems [58]. In the hopping regime, the electric current flows in a narrow quasi-one-dimensional cluster. Therefore, the intrinsic spin current is expected to be nonzero in the strongly inhomogeneous system under study.

The extrinsic spin-orbit coupling can also lead to the spin orientation [50] and spin relaxation [53] in analogy with Elliott-Yafet mechanism for free electrons. Extrinsic spin-orbit coupling also contributes to the spin-Hall conductivity similarly to the hyperfine interaction, see Eq. (12). However, the extrinsic spin-orbit coupling is parametrically small in the hopping regime since it is determined by three-center integrals [23,59].

In order to make an estimation of CISP by Eq. (13), we present the hopping resistivity as $\rho = \rho_0 \exp(2l_c/a_b)$, where l_c is the maximum distance between neighboring sites in the percolation cluster. We adopt the model where the hopping amplitudes are $J_{ij} = J_0 \exp(-R_{ij}/a_b)$ and $\tau_{ij} = \tau_0 \exp(2R_{ij}/a_b)$. Under these assumptions, $l_c \approx 1.2/\sqrt{n_s}$. The numerical simulation of spin dynamics in the hopping regime was performed on the square sample with 5×10^5 sites [34].

All of the three effects under study are described by a single dimensionless function f and obey the common dependence on the site concentration and on the spin relaxation time. The spin current as a function of τ_0/τ_s is shown in Fig. 4 for the small concentration $n_s a_b^2 = 0.01$. The drift and diffusion contributions to the spin current are shown separately. As expected, in the limit of slow spin relaxation, the drift and diffusion currents completely cancel each other out, so the total spin current is zero. We stress that the spin separation is still present in this limit and represents the intrinsic SHE. As the spin relaxation rate increases, the diffusion spin current diminishes, and in the limit $\tau_s = 0$, only the drift spin current survives in agreement with Eq. (16). For small concentrations $n_s a_b^2 < 0.02$, we find a finite value $f(n_s, 0) \approx 2.0$, so the drift spin current is independent of n_s in this limit.

For typical parameters, $n_s = 20n = 2 \times 10^{11}$ cm^{-2}, $a_b = 10$ nm, $J_0 = 10$ meV, $\beta_{xy} = \beta_{yx} = 10$ meV Å, $m = 0.1 m_0$, $E = 1$ kV/cm, $\rho_0 = 50$ kOhm [60], $\tau_0 = 10$ ps, and $\tau_s = 100$ ns, we obtain a small value $s \sim 3 \times 10^{-5}$. However, increase of n_s results in drastic enhancement of the electrical spin polarization. At the crossover from hopping to the diffusion conductivity, we get a relatively large value of CISP $s \sim 1\%$ easily detectable in experiments.

Conclusion.—We have proposed a unified description of CISP, SGE, and SHE in the hopping regime. Based on numerical simulations and percolation theory, we made estimations of the corresponding susceptibilities. Because of the suppression of spin relaxation in the hopping conductivity regime, the spin effects are underlined; in particular, the degree of current-induced spin polarization for real structures can be large.

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