Kinetic equations for hopping processes of small polarons under spin-orbit coupling

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We treat the hopping model of small polarons which are coupled by the Rashba spin-orbit interaction. The kinetic equations for the density matrix are derived and solved for a number of different charge- and spin-transport phenomena. We focus on the mutual coupling between the spin and charge degrees-of-freedom and its dependence on an applied electric field. Specific results are obtained for: (1) the electric-field-induced spin accumulation, (2) the relaxation of an initial homogeneous spin-magnetic moment, (3) the anomalous Hall effect of charge carriers, (4) the frequency response of the spin-Hall current, (5) the spatial charge- and spin-distribution near the boundary, and (6) the creation of a spin-magnetic moment due to particle diffusion. Although the polaron model has its own peculiarities, many results are reminiscent of corresponding findings in the band-transport theory, which recently has received considerable interest in the literature.

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I. INTRODUCTION

The generation of magnetic moments in semiconductors with spin-orbit interaction can be controlled by an electric field[1,2] or by optical means.[3,4] This effect is considered to be a particularly promising tool for spin-based device applications. As a consequence, the theoretical and experimental study of spin-related phenomena in the electronic transport of such systems has recently attracted intense attention. Most important is the interplay between the spin and charge degree-of-freedom as well as the antisymmetric scattering in the absence of any magnetic field, which gives rise to the spin-Hall effect[5] and the spin accumulation.[6]

The majority of theoretical studies is devoted to the linear transport regime of extended electronic states subjected to pure elastic scattering (see, for example, Refs. 7–11). In contrast, there are only few papers referring to the alternative hopping transport regime in systems with spin-orbit interaction.[12–16] Some spin-related effects have a different character in systems with extended and localized eigenstates. As an example, we mention the anomalous Hall effect, which is due to asymmetric scattering (quantum interference) for itinerant (localized) electrons. In the theory of hopping transport, one restricts the approach usually to two-site transitions. In the presence of a magnetic field, this approximation, which neglects the influence of the magnetic field on scattering probabilities, becomes completely insufficient. In order to account for the field-induced quantum interference, it is necessary to treat at least three-site hopping processes too.[17] The associated magnetic flux depends on how the carriers circle the triangle of the three hopping sites. Consequently, the carrier motion proceeds along a direction, which differs, in general, from that one of the electric field (Hall effect). Note that the three-site hopping model is only the simplest approximation that takes into account the quantum interference. For crystals, this approximation is only applicable for hexagonal symmetry. Otherwise (for instance for cubic symmetry), the more complicated four-site hopping model has to be taken into account. As the spin-orbit coupling (in the absence of an external magnetic field) leads to similar effects as an effective magnetic field, the consideration of its influence on the hopping transport also requires a treatment beyond the two-site approximation.[12,13]

In this paper, a general approach is presented that accounts for combined spin-charge effects in the hopping transport of localized carriers. We focus on a model for a crystal with hexagonal symmetry, in which localized eigenstates are due to strong electron-phonon interaction (model for small polarons). Our main task is the derivation of a coupled set of rate equations for the charge and spin densities. In addition, we present solutions of these kinetic equations for a number of experimentally relevant phenomena such as the spin accumulation, the anomalous Hall effect, the stationary spatial distribution of carriers and spins at contacts, and the decay of an initial packet of charge carriers and spins due to diffusion processes.

The derived rate equations are sufficiently general so that they are expected to be also applicable for transport phenomena due to extended states, when the model parameters (drift and Hall mobilities, diffusion coefficient, relaxation time) are appropriately redefined. However, it will be shown that this analogy holds only for the case of sufficiently weak spin-orbit coupling. Otherwise, the rate equations become non-Markovian so that the experimentally relevant quantities exhibit a nontrivial frequency dispersion under the influence of a time-dependent electric field.

II. MODEL AND BASIC EQUATIONS

Let us treat a model of a two-dimensional electron gas (2DEG) with Rashba spin-orbit interaction[18] that gives rise

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to the following dispersion relation for a system with nearest-neighbor coupling and with a narrow energy band

$$\varepsilon(\boldsymbol{k}-[\boldsymbol{\sigma}\times\boldsymbol{K}])=J_g\cos[(\boldsymbol{k}-[\boldsymbol{\sigma}\times\boldsymbol{K}])\boldsymbol{g}], \tag{1}$$

where $\boldsymbol{g}$ denotes the vector between nearest neighbors and J_g the resonance integral between them. $\boldsymbol{\sigma}$ is the vector of Pauli matrices and $\boldsymbol{k}=(k_x,k_y,0)$ denotes the in-plane momentum. The spin-orbit coupling is given by the wave vector $\boldsymbol{K}$, which is oriented perpendicular to the 2DEG (along the z axis). Most authors used the Rashba coupling constant $\alpha=\hbar K/m^*$, which has the dimension of a velocity (m^* denotes the effective mass). In addition, the model is characterized by the presence of strong inelastic electron-phonon couplings, for the description of which the polaron theory is utilized.[19] In this approach, the well-known polaron canonical transformation of the Hamiltonian leads to the result

$$H=-e\boldsymbol{E}\sum_{m,\lambda}\boldsymbol{R}_m a^\dagger_{m\lambda}a_{m\lambda}+\sum_{\boldsymbol{q}}\hbar\omega_{\boldsymbol{q}}\left(b^\dagger_{\boldsymbol{q}}b_{\boldsymbol{q}}+\frac{1}{2}\right)$$
$$+\sum_{m,\lambda}\sum_{m',\lambda'}J^{\lambda'\lambda}_{m'm}\hat{\Phi}_{m'm}a^\dagger_{m'\lambda'}a_{m\lambda}, \tag{2}$$

where $\boldsymbol{E}$ and $\boldsymbol{R}_m$ denote the external electric field and the radius vector of the lattice site m, respectively. Furthermore, the annihilation operators for electrons at the site m with spin $\lambda=1,2$ and for phonons with the wave vector $\boldsymbol{q}$ and frequency $\omega_{\boldsymbol{q}}$ are expressed by $a_{m\lambda}$ and $b_{\boldsymbol{q}}$, respectively. The polaron multiphonon operator $\hat{\Phi}_{m'm}$ is defined, e.g., in Refs. 14 and 19. The only difference between the Hamiltonian in Eq. (2) and its standard form in the polaron theory is due to the spin dependence of the resonance integral $J_{m'm}\to J^{\lambda'\lambda}_{m'm}$ given by

$$J^{\lambda'\lambda}_{m'm}=J_{m'm}\exp(i\boldsymbol{\sigma}\cdot[\boldsymbol{K}\times\boldsymbol{R}_{m'm}])_{\lambda'\lambda}. \tag{3}$$

Therefore, the diagram technique developed to account for scattering processes of small polarons can be applied with slight modifications (cf., for instance, Ref. 19).

For the treatment of spin-dependent hopping effects, we need the density matrix

$$\rho^\lambda_{\lambda'}(m,t)=\langle a^\dagger_{m\lambda'}a_{m\lambda}\rangle_t, \tag{4}$$

which is calculated from balance equations

$$\frac{d\rho^\lambda_{\lambda'}(m,t)}{dt}=\sum_{m'}\sum_{\lambda_1,\lambda_2}\rho^{\lambda_1}_{\lambda_2}(m',t)W^{\lambda_1\lambda}_{\lambda_2\lambda'}(m',m), \tag{5}$$

in which the transition probabilities W satisfy the sum rule

$$\sum_{m,\lambda}W^{\lambda_1\lambda}_{\lambda_2\lambda}(m',m)=0. \tag{6}$$

Due to the conservation of the particle number, we obtain

$$S^{-1}\sum_{m,\lambda}\rho^\lambda_\lambda(m,t)=n, \tag{7}$$

where S denotes the area of the 2DEG and n is the electron density per unit area. Note that Eq. (5) was obtained in the one-electron approximation so that all results derived below apply only to Boltzmann statistics. In addition, non-Markovian effects are excluded from our consideration.

In this paper, we treat a crystalline system, for which the transition probabilities $W^{\lambda_1\lambda'}_{\lambda_2\lambda}(m',m)$ depend only on the difference of the site numbers $m'-m$ so that the balance equation (5) becomes diagonal after performing a Fourier transformation

$$\rho^\lambda_{\lambda'}(\boldsymbol{\kappa},t)=\sum_m\rho^\lambda_{\lambda'}(m,t)\exp(i\boldsymbol{\kappa}\cdot\boldsymbol{R}_m), \tag{8}$$

$$\frac{d\rho^\lambda_{\lambda'}(\boldsymbol{\kappa},t)}{dt}=\sum_{\lambda_1,\lambda_2}\rho^{\lambda_1}_{\lambda_2}(\boldsymbol{\kappa},t)W^{\lambda_1\lambda}_{\lambda_2\lambda'}(\boldsymbol{\kappa}). \tag{9}$$

For disordered systems, the influence of the spin-orbit interaction on localized states was treated in Ref. 16.

To proceed further, it is expedient to collect from the four components of the density matrix $\rho^\lambda_{\lambda'}$ the particle ρ and spin $\boldsymbol{\rho}=(\rho_x,\rho_y,\rho_z)$ contributions defined by

$$\rho=\sum_\lambda\rho^\lambda_\lambda,\quad \boldsymbol{\rho}=\sum_{\lambda,\lambda'}\rho^\lambda_{\lambda'}\boldsymbol{\sigma}_{\lambda\lambda'}. \tag{10}$$

The kinetic equations for these quantities have the form

$$\frac{d\rho(\boldsymbol{\kappa},t)}{dt}=\sum_{\lambda_1,\lambda_2}\sum_\lambda\rho^{\lambda_1}_{\lambda_2}(\boldsymbol{\kappa},t)W^{\lambda_1\lambda}_{\lambda_2\lambda}(\boldsymbol{\kappa}), \tag{11}$$

$$\frac{d\boldsymbol{\rho}(\boldsymbol{\kappa},t)}{dt}=\sum_{\lambda_1,\lambda_2}\sum_{\lambda,\lambda'}\rho^{\lambda_1}_{\lambda_2}(\boldsymbol{\kappa},t)W^{\lambda_1\lambda}_{\lambda_2\lambda'}(\boldsymbol{\kappa})\boldsymbol{\sigma}_{\lambda\lambda'}, \tag{12}$$

where, as usual in the theory of small polarons, the hopping probabilities W are expanded into a series of the resonance integral J. The lowest-order two-site approximation of the Eqs. (11) and (12) has already been studied in Ref. 13. This approach provides the following contributions to the right-hand sides of Eqs. (11) and (12), which are linear in the electric field $\boldsymbol{E}$ and linear and quadratic in $\boldsymbol{\kappa}$:

$$-[D\boldsymbol{\kappa}^2+iu(\boldsymbol{\kappa}\cdot\boldsymbol{E})]\rho, \tag{11a}$$

$$-\left[D\boldsymbol{\kappa}^2+iu(\boldsymbol{\kappa}\cdot\boldsymbol{E})+\frac{\hat{A}}{\tau}\right]\boldsymbol{\rho}$$
$$-4D\left\{\left[\boldsymbol{K}\times\left(i\boldsymbol{\kappa}-\frac{e\boldsymbol{E}}{2k_BT}\right)\right]\times\boldsymbol{\rho}\right\}. \tag{12a}$$

In these equations, the drift mobility of small polarons

$$u=u_0\frac{\sqrt{\pi}J^2}{E_a^{1/2}(k_BT)^{3/2}}\exp\left(-\frac{E_a}{k_BT}\right) \tag{13}$$

has been introduced. The parameters are: E_a—an activation energy (cf. Ref. 19), $D=uk_BT/e$—the diffusion coefficient, a—the lattice constant, $u_0=ea^2/\hbar$—a mobility parameter, $\tau=(4DK^2)^{-1}$—the spin-relaxation time, and $\hat{A}$—a diagonal tensor with the components $A_{xx}=A_{yy}=1$, $A_{zz}=2$.

As mentioned above, a due treatment of the spin-orbit coupling in a system of small polarons requires the consideration of at least three-site hopping transitions. The situation is reminiscent of the calculation of the charge-Hall effect in the presence of a magnetic field,[19] where the three-site transitions give rise to a magnetic flux and associated quantum interferences.[17] For hopping systems with spin-orbit coupling, three-site hops were already treated in Ref. 13. However, in this paper only contributions up to the order K^2 were taken into account. Unfortunately, this simplification does not allow the treatment of such interesting phenomena as the electric-field-induced spin accumulation. The calculation of the three-site hopping contributions to the right-hand sides of Eqs. (11) and (12) is somewhat tedious but straightforward (and similar to calculations in Ref. 13). The final result has the form

$$-4iD\frac{u_H}{u_K}\left\{\frac{e(\boldsymbol{\kappa}[\boldsymbol{K}\times\boldsymbol{E}])(K\cdot\boldsymbol{\rho})}{2k_BTK^2}+(\boldsymbol{\kappa}\cdot[K\times\boldsymbol{\rho}])\right\}, \tag{11b}$$

$$-4D\frac{u_H}{u_K}\left\{ie\frac{(\boldsymbol{\kappa}[\boldsymbol{K}\times\boldsymbol{E}])}{2k_BTK^2}\boldsymbol{K}+\left[\boldsymbol{K}\times\left(i\boldsymbol{\kappa}-\frac{e\boldsymbol{E}}{k_BT}\right)\right]\right\}\rho, \tag{12b}$$

where a spin-related mobility $u_K=e/(\hbar K^2)$ has been introduced. In addition to the results in Ref. 13, there are contributions proportional to K^3. The Hall mobility

$$u_H=u_0\frac{\sqrt{\pi}J}{\sqrt{E_ak_BT}}\exp\left(-\frac{E_a}{3k_BT}\right), \tag{14}$$

of small polarons in hexagonal crystals drastically differs from the drift mobility given in Eq. (13).[19]

Note that there are three-site contributions that are due to virtual hopping transitions and which are given by the imaginary part of the scattering probability.[13] Similar virtual transitions also occur in the spin-mediated band transport.[20] Although the significance of these contributions is not sufficiently clear up to now, we can safely drop their consideration for not too strong spin-orbit couplings.

Taking into account Eqs. (11a) and (11b) together with (12a) and (12b), we obtain from Eqs. (11) and (12) the final set of kinetic equations

$$\frac{d\rho}{dt}=-[D\boldsymbol{\kappa}^2+iu(\boldsymbol{\kappa}\cdot\boldsymbol{E})]\rho-4iD\frac{u_H}{u_K}\left\{\frac{e(\boldsymbol{\kappa}\cdot[\boldsymbol{K}\times\boldsymbol{E}])(K\cdot\boldsymbol{\rho})}{2k_BTK^2}+(\boldsymbol{\kappa}\cdot[K\times\boldsymbol{\rho}])\right\}, \tag{15}$$

$$\frac{d\boldsymbol{\rho}}{dt}=-\left[D\boldsymbol{\kappa}^2+iu(\boldsymbol{\kappa}\cdot\boldsymbol{E})+\frac{\hat{A}}{\tau}\right]\boldsymbol{\rho}-4D\left\{\left[\boldsymbol{K}\times\left(i\boldsymbol{\kappa}-\frac{e\boldsymbol{E}}{2k_BT}\right)\right]\times\boldsymbol{\rho}\right\}-4D\frac{u_H}{u_K}\left\{ie\frac{(\boldsymbol{\kappa}[\boldsymbol{K}\times\boldsymbol{E}])}{2k_BTK^2}\boldsymbol{K}+\left[\boldsymbol{K}\times\left(i\boldsymbol{\kappa}-\frac{e\boldsymbol{E}}{k_BT}\right)\right]\right\}\rho. \tag{16}$$

These rate equations for the charge (15) and spin (16) degrees-of-freedom are the main results in this paper. In the literature, many authors[9,21–25] derived similar equations for extended states subject to elastic scattering. As in these studies energy dissipation is not included, the kinetic equations refer to spectral densities, which are integrated over the energy at the end of the calculation. In contrast, Eqs. (15) and (16), which do not have a spectral character, refer to the hopping regime with strong inelastic scattering with the phonon heat bath. When the electric field is switched off, the energy relaxation does not play any significant role and from a phenomenological point of view, Eqs. (15) and (16) agree with corresponding results in Refs. 9 and 22, when model parameters like u, u_H, D, and τ are appropriately redefined.

III. SPATIALLY HOMOGENEOUS SYSTEMS

First, let us solve the basic kinetic Eqs. (15) and (16) for spatially homogeneous systems by omitting any boundary effects. In this case, one can treat the field-induced homogeneous spin accumulation and the relaxation of an initial homogeneous nonequilibrium magnetic moment. For $\boldsymbol{\kappa}=0$, the Laplace transformed kinetic Eqs. (15) and (16) take the simple form

$$\rho^{(0)}=n/s, \tag{17}$$

$$\left(s+\frac{\hat{A}}{\tau}\right)\boldsymbol{\rho}^{(0)}-2u\{[\boldsymbol{K}\times\boldsymbol{E}]\times\boldsymbol{\rho}^{(0)}\}-4\frac{u_Hu}{u_K}[\boldsymbol{K}\times\boldsymbol{E}]\rho^{(0)}=\boldsymbol{\rho}_0, \tag{18}$$

where s is the Laplace variable, which replaces the time t. $\boldsymbol{\rho}_0$ denotes the initial homogeneous spin moment and the upper index (0) refers to spatially homogeneous quantities. Solving these equations and returning to the time representation, we obtain

$$\boldsymbol{\rho}(t)=\boldsymbol{\rho}_{ac}(t)+\boldsymbol{\rho}_r(t), \tag{19}$$

where the field-induced spin accumulation

$$\boldsymbol{\rho}_{ac}(t)=\boldsymbol{\rho}_\infty[1-\exp(-t/\tau)],$$

approaches the steady-state value

$$\boldsymbol{\rho}_\infty=\frac{\hbar u_H}{k_BT}(\boldsymbol{K}\times\boldsymbol{E})n=\hbar u_H(\boldsymbol{K}\times\boldsymbol{E})\frac{dn}{d\varepsilon_F}. \tag{20}$$

Here, we conveniently employed $dn/d\varepsilon_F=nk_BT$ (with ε_F being the Fermi energy), which is satisfied for the Boltzmann statistics. The generation of a magnetic moment by an electric field is the well-known magnetoelectric effect of the electrodynamics. Its appearance in systems with spin-orbit interaction was predicted by Edelstein[6] many years ago. Note that Eq. (20) completely agrees with Edelstein's result,[6] although he treated extended states and Fermi statistics (in this model, the Hall and drift mobilities agree $u_H=u=e\tau_p/m^*$, with τ_p being the momentum relaxation time). This similarity stresses the universal character of Eq. (20), which seems to be independent of the transport mechanism and the carrier statistics. An ac electric field $\boldsymbol{E}(t)=\boldsymbol{E}\exp(-i\omega t)$ induces a frequency dispersion of the spin accumulation, which exhibits the standard Drude form

$$\boldsymbol{\rho}_{ac}(\omega) = \frac{\boldsymbol{\rho}_\infty}{1 - i\omega\tau} \tag{21}$$

for the considered hopping regime. For extended states, the frequency dependence of the spin accumulation is in general more complicated[20] and takes the Drude form only for weak spin-orbit coupling, when $\nu_F K \tau_p = Kl \ll 1$ (where ν_F denotes the Fermi velocity and l the length of the mean free path). Under the condition $Kl \gg 1$, the spin accumulation of itinerant electrons behaves differently and exhibits a resonance at the frequency $\omega = 2\nu_F K$. Therefore, we conclude that the universal character of Eqs. (15) and (16) is only expected for weak spin-orbit coupling. In general, the time dependence of the spin accumulation has a non-Markovian character.

Next, let us study the spin contribution $\boldsymbol{\rho}_r(t)$ in Eq. (19), which describes the relaxation of a homogeneous spin moment. For an electric field oriented along the x axis, we have

$$\rho_{ry}(t) = \rho_{0y} \exp\left(-\frac{t}{\tau}\right), \tag{22}$$

$$\rho_{rx}(t) = \exp\left(-\frac{3t}{2\tau}\right) \times \frac{\rho_{0x}\cos(t\sqrt{\zeta^2-1}/2\tau - \Phi) + \rho_{0z}\sin(t\sqrt{\zeta^2-1}/2\tau)}{\cos\Phi}, \tag{23}$$

$$\rho_{rz}(t) = \exp\left(-\frac{3t}{2\tau}\right) \times \frac{\rho_{0z}\cos(t\sqrt{\zeta^2-1}/2\tau + \Phi) - \rho_{0x}\sin(t\sqrt{\zeta^2-1}/2\tau)}{\cos\Phi}, \tag{24}$$

where the phase Φ is calculated from $\sin\Phi = \zeta^{-1}$, with ζ being the following field parameter

$$\zeta = E/E_c, \quad E_c = (4u\tau K)^{-1} = DK/u = Kk_BT/e. \tag{25}$$

According to this solution, an initial nonequilibrium magnetic moment in the x-z plane performs spin rotations in this plane with the frequency $\sqrt{\zeta^2-1}/(2\tau)$, when the electric field is larger than the critical value E_c. The spin rotation was predicted for a magnetic moment in the hopping regime in Refs. 12 and 13 and later also for the band-transport model.[21,24] The experimental verification of these damped oscillations requires strong electric fields $\zeta \gg 1$. The eigenmodes can be excited by interference effects with photogenerated electrons or by an external magnetic field.[14] We call these eigenmodes spin remagnitization waves in analogy to the well known trap recharging waves (space charge waves) in semiconductors,[26,27] which are experimentally detected by the frequency dependence of the current or by the study of the Bragg diffraction pattern in photorefractive crystals.

IV. HALL CURRENT

The current of charge carriers is defined by the time derivative of the dipole moment $\boldsymbol{j} = d\boldsymbol{D}/dt$, the Fourier transformed version of which has the form

$$\boldsymbol{j}(t) = ie\boldsymbol{\nabla}_{\boldsymbol{\kappa}} \left.\frac{d}{dt}\rho(\boldsymbol{\kappa},t)\right|_{\boldsymbol{\kappa}=0}. \tag{26}$$

The $\boldsymbol{\kappa}$ derivative of Eq. (15) is easily calculated and we obtain

$$\boldsymbol{j}(t) = eun\boldsymbol{E} + \frac{\hbar u_H}{\tau}\left\{\frac{e[\boldsymbol{K}\times\boldsymbol{E}]}{2k_BTK^2}(\boldsymbol{K}\cdot\boldsymbol{\rho}^{(0)}(t)) + [\boldsymbol{K}\times\boldsymbol{\rho}^{(0)}(t)]\right\}, \tag{27}$$

in which the spatially homogeneous spin density $\boldsymbol{\rho}^{(0)}(t)$ is given by Eqs. (19), (20), (22), and (24). When initially there is no magnetic moment $\boldsymbol{\rho}_0 = \boldsymbol{0}$, the ac response of the charge current has the form

$$\boldsymbol{j}(\omega) = \left\{1 - \frac{(2u_H/u_K)^2}{1 - i\omega\tau}\right\} enu\boldsymbol{E}. \tag{28}$$

As $\boldsymbol{j}(\omega)$ has the same direction as the electric field $\boldsymbol{E}$, the Hall current of charge carriers disappears in this case. The spin-orbit interaction leads only to a small correction $\sim K^4$, which, however, exhibits a frequency dispersion that makes its experimental detection in principle feasible. Note that the frequency dependence in Eq. (28) has no counterpart in the polaron theory of particles without spin. For small polarons, the frequency dependence of the conductivity sets in at very high frequencies $\omega \approx E_a/\hbar$, which typically lies in the visible spectral range.[19] However, such a frequency dependence as that of Eq. (28) is also observed in the conductivity of extended electronic states coupled by spin-orbit interaction.[20,28] In general, the resonance denominator in the expression for the spin accumulation is more complicated in this case.[20]

If an initial magnetic moment exists, then a charge current is induced, which is calculated from

$$\boldsymbol{j}_r(t) = \frac{\hbar u_H}{\tau}\left\{\frac{e[\boldsymbol{K}\times\boldsymbol{E}]}{2k_BTK^2}(\boldsymbol{K}\cdot\boldsymbol{\rho}_r(t)) + [\boldsymbol{K}\times\boldsymbol{\rho}_r(t)]\right\}, \tag{29}$$

where the spin-relaxation density $\boldsymbol{\rho}_r(t)$ is given by Eqs. (22)–(24). We point out that this current does not disappear at zero electric field $\boldsymbol{E}=0$. According to the time-reversal invariance of the Hamiltonian, such a current is exclusively due to the initial macroscopic spin moment $\boldsymbol{\rho}_0$. Inserting Eqs. (22)–(24) into Eq. (29), we obtain for the relaxation components of the charge current

$$j_{rx}(t) = -uKk_BT\frac{u_H}{u_K}\exp\left(-\frac{t}{\tau}\right)\rho_{0y}, \tag{30}$$

$$j_{ry}(t) = 2eE\frac{u_H}{u_K}u\exp\left(-\frac{3t}{2\tau}\right) \times \frac{\rho_{0z}\cos(t\sqrt{\zeta^2-1}/2\tau+\Phi) - \rho_{0x}\sin(t\sqrt{\zeta^2-1}/2\tau)}{\cos\Phi} + k_BTK\frac{u_H}{u_K}u\exp\left(-\frac{3t}{2\tau}\right) \times \frac{\rho_{0x}\cos(t\sqrt{\zeta^2-1}/2\tau-\Phi) + \rho_{0z}\sin(t\sqrt{\zeta^2-1}/2\tau)}{\cos\Phi}. \quad (31)$$

The Hall contribution in Eq. (31), the principle existence of which was already predicted in Refs. 29 and 30, disappears in the steady-state due to spin relaxation. This result is not surprising because the system does not obey the magnetic symmetry. A stationary Hall current of charge carriers exists in systems with spin-orbit coupling only under additional conditions, as, for instance, the presence of an external magnetic field perpendicular to the 2DEG or ferromagnetic contacts.

We now turn to the spin-Hall current, which was originally treated by Hirsch.[5] Most theoretical studies in this field refer to the band-transport model. Shortly after the interest in this field had grown rapidly, the subject led to a controversial discussion and to contradicting results. The main problem consists in the definition of the proper spin current. In contrast to the charge current, the spin current is not a measurable quantity and does not occur in Maxwell's equations. In the experiment, only the induced magnetic moments at the sample boundaries can be detected, which might be due to a spin current perpendicular to the electric field,[31–33] quite similar to the Hall voltage, which results from the Hall current of charge carriers. The question, how the proper spin current has to be defined was thoroughly treated in the literature.[20,34,35] Here, in analogy to Eq. (26), we identify the spin current with the time derivative of the spin displacement

$$j^i_{(s)\alpha}(t) = \left.\frac{i}{2}\frac{d}{dt}\frac{\partial}{\partial\kappa_\alpha}\rho_i(t)\right|_{\boldsymbol{\kappa}=\mathbf{0}}, \quad (32)$$

where $i=x,y,z$ denotes the components of the spin-density matrix $\boldsymbol{\rho}$ and $\alpha=x,y$ the axis of the 2DEG. According to this definition, the spin current is given by the time derivative of the center of mass velocity of the spin packet multiplied by the total spin of the system. Contrary to the charge current, which is based on particle-number conservation, the spin moment finally relaxes to zero so that it is expected that the spin current in Eq. (32) disappears in the steady state.

By calculating the $\boldsymbol{\kappa}$ derivative of Eq. (16) and putting $\boldsymbol{\kappa}=\mathbf{0}$ afterwards, we obtain for the spin-Hall current

$$j^i_{(s)\alpha}(t) = j^i_{(s0)\alpha}(t) + j^i_{(sE)\alpha}(t), \quad (33)$$

where the first contribution

$$j^x_{(s0)y}(t) = -K\frac{\hbar u_H}{2e\tau}n\exp\left(-\frac{t}{\tau}\right), \quad (34)$$

is independent of the electric field. The y component of this spin current is zero $j^y_{(s0)y}=0$. A field-independent spin-Hall current was already obtained in Refs. 13 and 15 for the hopping regime and in Ref. 20 for the band-transport model. The origin of this field-independent spin current is the initial time evolution of the spin accumulation, which is related to this spin current by the universal relation

$$\frac{d\rho^y_{ac}(t)}{dt}n = -2eEj^x_{(s0)y}(t)\frac{dn}{d\varepsilon_F} \quad (35)$$

that expresses the phenomenological concept of an effective electrochemical potential (a similar relationship has been obtained for the band transport[20]).

The second contribution to the spin-Hall current in Eq. (33) is induced by an electric field and has the following explicit form in the time domain

$$j^x_{(sE)y}(t) = K\frac{3\hbar u_H}{2e\tau}n\left\{e^{-t/\tau} - e^{-3t/(2\tau)}\left[\frac{1+\zeta^2/6}{\sqrt{\zeta^2-1}}\sin\left(\frac{t}{2\tau}\sqrt{\zeta^2-1}\right) + \cos\left(\frac{t}{2\tau}\sqrt{\zeta^2-1}\right)\right]\right\}, \quad (36)$$

$$j^z_{(sE)y}(t) = -K\frac{\hbar u_H n}{2e\tau}\frac{\zeta}{2}e^{-3t/(2\tau)}\left\{\frac{7}{\sqrt{\zeta^2-1}}\sin\left(\frac{t}{2\tau}\sqrt{\zeta^2-1}\right) - \cos\left(\frac{t}{2\tau}\sqrt{\zeta^2-1}\right)\right\}. \quad (37)$$

In analogy to the spin accumulation, the relaxation of the spin-Hall current proceeds by a rotation of the magnetic moment in the x-z plane with the frequency $\sqrt{\zeta^2-1}/(2\tau)$ (when $E > E_c$). All previous theoretical studies of the spin-Hall current are restricted to linear electric field effects. In that approximation, there is no rotation of the magnetic moment so that the interest of the authors focused on the frequency dependence of the spin-Hall current, which is due to an oscillating electric field of the form $\boldsymbol{E}\exp(-i\omega t)$. In the linear field regime, the frequency dispersion of the spin-Hall current is obtained from Eq. (37) by a Fourier transformation. The result

$$j^z_{(sE)y}(\omega) = \frac{\hbar u_H En}{4k_BT}\frac{i\omega(2+i\omega\tau)}{(1-i\omega\tau)(2-i\omega\tau)} \quad (38)$$

describes a spin-Hall current that disappears proportional to ω^2 in the limit $\omega\to 0$ (cf. also Ref. 15) and reaches for $\omega\tau\to\infty$ the plateau $\hbar u_H En/(4k_BT\tau)$. The transition between these two regimes occurs at $\omega\approx\tau^{-1}=4DK^2$. A similar frequency dispersion of the spin-Hall current is also characteristic for the band-transport model, where, however, the ballistic spin-Hall conductivity of a pure sample remains finite in the limit $\omega\to 0$ (cf., for instances, Refs. 20 and 36–38).

V. EFFECTS AT CONTACTS

The basic rate Eqs. (15) and (16) also allow the calculation of the stationary spatial distribution of charge carriers and spin at the sample boundaries. We restrict to a simple sample geometry namely the half plane defined by $y>0$, for which the kinetic Eqs. (15) and (16) simplify to

$$\rho'' + 2\frac{u_H}{u_K}\rho_x' + \frac{u_H}{u_K}\zeta\rho_z' = 0, \tag{39}$$

$$\rho_x'' - \rho_x + \frac{\zeta}{2}\rho_z - 2\frac{u_H}{u_K}\rho' = 0, \tag{40}$$

$$\rho_y'' - \rho_y + 2\rho_z' + \frac{u_H}{u_K}\zeta\rho = 0, \tag{41}$$

$$\rho_z'' - 2\rho_z - 2\rho_y' + \frac{u_H}{u_K}\zeta\rho' - \frac{\zeta}{2}\rho_x = 0, \tag{42}$$

where ρ' expresses the derivative of ρ with respect to $\xi=y/\sqrt{D\tau}=2Ky$. The solution of these equations has a rather complex form so that we derive only results for weak spin-orbit coupling $u_H \ll u_K$. In this case, the carrier density is homogeneous $\rho=n$, where n denotes the equilibrium particle density. For the spin-density matrix $\boldsymbol{\rho}$, we obtain

$$\rho_x = c\,\exp(-\xi) + \zeta\,\mathrm{Re}[c_1 \exp(-\lambda\xi)], \tag{43}$$

$$\rho_y = -c\frac{\zeta}{4}\exp(-\xi) + 4\,\mathrm{Re}[c_1\lambda\exp(-\lambda\xi)] + \frac{u_H}{u_K}\zeta n, \tag{44}$$

$$\rho_z = -2\,\mathrm{Re}[c_1(\lambda^2-1)\exp(-\lambda\xi)], \tag{45}$$

where the real c and complex c_1 constants are determined from the boundary condition whereas λ is given by

$$\lambda = \frac{1}{2}\left(\sqrt{\sqrt{8+\zeta^2}-1} + i\sqrt{\sqrt{8+\zeta^2}+1}\right) = i\sqrt{\frac{1}{2}(1+i\sqrt{7+\zeta^2})}. \tag{46}$$

The last term on the right-hand side of Eq. (44) provides the field-induced spin accumulation [cf. Eq. (20)].

Assuming a finite magnetic moment ρ_z at the boundary $y=0$ (e.g., realized by a ferromagnetic contact), an electric field induces all three components of the spin moment, which experience damped oscillations with the wavelength $2\pi K^{-1}(\sqrt{8+\zeta^2}+1)^{-1/2}$ up to a depth of $K^{-1}(\sqrt{8+\zeta^2}-1)^{-1/2}$. When $E=0$, the spin component ρ_x is zero (or vanishes at a distance $(2K)^{-1}$ from the boundary when $\rho_x(y=0)\neq 0$). For weak spin-orbit coupling $\nu_F K\tau_p \ll 1$, the characteristic diffusion length calculated for the band-transport model at $E=0$ in Ref. 22 completely agrees with the result in Eq. (46).

Let us now turn to the treatment of the spin-induced charge accumulation at the boundary $y=0$ for $E=0$, when Eqs. (39)–(42) decouple into independent sets of equations for ρ,ρ_x and ρ_y,ρ_z. For $\zeta=0$, the Eqs. (39)–(42) have the solution

$$\rho = n + \rho_x^{(0)}\frac{\exp(-\sqrt{1-(2u_H/u_K)^2}\xi)}{\sqrt{1-(u_K/2u_H)^2}}, \tag{47}$$

$$\rho_x = \rho_x^{(0)}\exp(-\sqrt{1-(2u_H/u_K)^2}\xi), \tag{48}$$

$$\rho_y = \mathrm{Re}\left[c\frac{1-\lambda^2}{2\lambda}\exp(-\lambda\xi)\right], \tag{49}$$

$$\rho_z = \mathrm{Re}[c\,\exp(-\lambda\xi)], \tag{50}$$

where λ is given by Eq. (46) for $\zeta=0$. The solutions in Eqs. (47) and (48) are applicable under the condition $2u_H<u_K$, which is very well satisfied in low-mobility samples, where $u_H<1\ \mathrm{cm^2/Vs}$ but $u_K\approx 10^3\ \mathrm{cm^2/Vs}$ for $K^{-1}\approx 10^{-6}$ cm. Note, however, that Eq. (47) gives only a rough estimate of the spin-mediated charge accumulation at the boundary because the approach does not fully capture the basic physics of the problem. The redistribution of charges induces an internal electric field that has to be self-consistently treated by the Poisson equation. Such a self-consistent approach, which becomes especially important for $2u_H>u_K$ (when oscillatory solutions exist), subsequently leads to a coupling between all components of the density matrix.

The solutions in Eqs. (49) and (50) for the spin components ρ_y and ρ_z describe the generation of a stacked magnetic moment near the interface, which occurs, when at least one of the densities ρ_y and ρ_z does not vanish at $y=0$. Such long-lived spin-coherent oscillations have been identified by a recent numerical study, too.[39]

The hard-wall boundary condition deserves particular attention, when at $y=0$ both the charge and spin currents across the boundary have to disappear. Just under these conditions, the experimental study of the spin-Hall effect in GaAs has been performed.[31–33] Two competing mechanisms determine the formation of a spin moment at the boundary namely (i) the supply of polarized spins by the spin-Hall current and (ii) the relaxation of the created excess magnetic moment. Taking into account Eq. (39), the condition for the disappearance of the charge current at the boundary is formulated by

$$\rho' + \frac{u_H}{u_K}(2\rho_x + \zeta\rho_z) = 0. \tag{51}$$

To obtain a similar boundary condition for the spin current, one needs a reliable definition of the spin current under spatial dispersion. In Ref. 13, the following hard-wall boundary conditions have been applied to solve Eqs. (40)–(42)

$$\rho_x' - \frac{2u_H}{u_K}\rho = 0, \tag{52}$$

$$\rho_y' + 2\rho_z = 0, \tag{53}$$

$$\rho_z' - 2\rho_y + \frac{u_H}{u_K}\zeta\rho = 0. \tag{54}$$

Under these conditions, all three components of the spin moment exist at the interface. Moreover, the approach predicts the appearance of a magnetic field,[13] the quantitative experimental identification of which, however, seems to be difficult at present.

According to the boundary conditions (52)–(54), a finite inhomogeneous charge and spin distribution exist at the interface even at $E=0$. In contradiction to this result, arguments are put forward in the literature[40,41] that favor the disappearance of the spin moment in the whole sample (including the interface) under the condition of thermodynamic equilibrium.

VI. EFFECTS OF SPIN DIFFUSION

Finally, we demonstrate that the basic kinetic Eqs. (15) and (16) also allow a detailed analysis of diffusion phenomena in systems with spin-orbit interaction. Here, our primary concern is the treatment of the diffusion of an initial point source ($\rho=1$, $\boldsymbol{\rho}=\mathbf{0}$), which is described by the Laplace-transformed kinetic Eqs. (15) and (16):

$$[s+D\boldsymbol{\kappa}^2+iu(\boldsymbol{\kappa}\cdot\boldsymbol{E})]\rho+4iD\frac{u_H}{u_K}\left\{\frac{e(\boldsymbol{\kappa}[\boldsymbol{K}\times\boldsymbol{E}])(\boldsymbol{K}\cdot\boldsymbol{\rho})}{2k_BTK^2}+(\boldsymbol{\kappa}\cdot[\boldsymbol{K}\times\boldsymbol{\rho}])\right\}=1, \tag{55}$$

$$\left[s+D\boldsymbol{\kappa}^2+iu(\boldsymbol{\kappa}\cdot\boldsymbol{E})+\frac{\hat{A}}{\tau}\right]\boldsymbol{\rho}+4D\left\{\left[\boldsymbol{K}\times\left(i\boldsymbol{\kappa}-\frac{e\boldsymbol{E}}{2k_BT}\right)\right]\times\boldsymbol{\rho}\right\}+4D\frac{u_H}{u_K}\left\{ie\frac{(\boldsymbol{\kappa}[\boldsymbol{K}\times\boldsymbol{E}])}{2k_BTK^2}\boldsymbol{K}+\left[\boldsymbol{K}\times\left(i\boldsymbol{\kappa}-\frac{e\boldsymbol{E}}{k_BT}\right)\right]\right\}\rho=0. \tag{56}$$

Again, s denotes the Laplace variable that replaces the time t. When an electric field is absent, these equations decouple into a set of equations for the quantities ρ and $[\boldsymbol{\kappa}\times\boldsymbol{\rho}]_z$ as well as ρ_z and $(\boldsymbol{\kappa}\cdot\boldsymbol{\rho})$. Accordingly, a spin moment may develop in the system, whose divergence and consequently also its "magnetic charge" vanish. For the Rashba model, this feature has an universal character that appears also in the band-transport model.[42]

To derive analytical results for the spin and particle diffusion, we consider a system with weak spin-orbit coupling, when the spin and charge degrees-of-freedom completely decouple in the lowest-order approximation so that we obtain

$$\rho(\boldsymbol{\kappa},s)=[s+D\boldsymbol{\kappa}^2+iu(\boldsymbol{\kappa}\cdot\boldsymbol{E})]^{-1},\quad \boldsymbol{\rho}(\boldsymbol{\kappa},s)=\mathbf{0}.$$

A finite correction to the spin diffusion arises from the first-order approximation of Eq. (56), which has the form

$$\boldsymbol{\rho}(\boldsymbol{\kappa},s)=-\frac{\hbar u_H}{\tau[s+D\boldsymbol{\kappa}^2+iu(\boldsymbol{\kappa}\cdot\boldsymbol{E})+\hat{A}/\tau][s+D\boldsymbol{\kappa}^2+iu(\boldsymbol{\kappa}\cdot\boldsymbol{E})]}\times\left\{i\frac{(\boldsymbol{\kappa}[\boldsymbol{K}\times\boldsymbol{E}])\boldsymbol{K}}{2k_BTK^2}+\left[\boldsymbol{K}\times\left(\frac{i}{e}\boldsymbol{\kappa}-\frac{\boldsymbol{E}}{k_BT}\right)\right]\right\}. \tag{57}$$

Applying an inverse Fourier and Laplace transformation, we obtain

$$\boldsymbol{\rho}_\perp(\boldsymbol{r},t)=\left[1-\exp\left(-\frac{t}{\tau}\right)\right]\hbar u_H\left[\frac{\boldsymbol{K}\times(\boldsymbol{r}+u\boldsymbol{E}t)}{2eDt}\right]\rho(\boldsymbol{r},t), \tag{58}$$

$$\rho_z(\boldsymbol{r},t)=\left[1-\exp\left(-\frac{2t}{\tau}\right)\right]\frac{\hbar u_H}{8Dtk_BT}[\boldsymbol{E}\times\boldsymbol{r}]_z\rho(\boldsymbol{r},t), \tag{59}$$

with $\boldsymbol{\rho}_\perp=(\rho_x,\rho_y)$ and the well-known particle-diffusion function

$$\rho(\boldsymbol{r},t)=\frac{1}{4\pi Dt}\exp\left(-\frac{(\boldsymbol{r}-u\boldsymbol{E}t)^2}{4Dt}\right).$$

The Eqs. (58) and (59) describe the evolution of an initially absent spin moment that is created by the particle diffusion. The dynamics of both the spin and particle diffusion have a similar character. At $\boldsymbol{E}=\mathbf{0}$, the vector of the induced spin moment lies in the plane of the 2DEG and its divergence vanishes. Note that the generation of a magnetic moment by the charge-carrier diffusion is closely related to the spin-Hall effect.

VII. SUMMARY

The main results in this paper represents the set of kinetic Eqs. (15) and (16) for the four components of the density matrix that refer to a hopping system with localized states subject to spin-orbit interaction. These equations are similar to corresponding ones in the theory of band transport, especially for zero electric field. However, in contrast to the band model, the Eqs. (15) and (16) have been derived by considering strong inelastic scattering mediated by a phonon heat bath so that they do not have a spectral character. Nevertheless, from a phenomenological point of view, the kinetic equations for the hopping and band transport phenomena agree for weak spin-orbit coupling ($Kl\ll 1$), when physical parameters as the drift and Hall mobilities are adequately redefined. For small polarons, both parameters are given by Eqs. (13) and (14), whereas for the band transport, we have $u=u_H=e\tau_p/m^*$. Independent of the transport mechanism, the diffusion coefficient obeys the Einstein relation $nu=eDdn/d\varepsilon_F$. The situation changes drastically for strong spin-orbit interaction ($Kl\gg 1$). In this case, the non-Markovian behavior becomes essential so that the effective diffusion coefficient varies with time, and relaxation processes are no longer simply exponential. Physical effects in this interesting regime will be treated in a forthcoming paper.

Based on the kinetic Eqs. (15) and (16), a number of different spin-related problems in the model of small polarons were considered:

(i) The field-induced spin accumulation in the model of small polarons was treated in response to both a dc and ac electric field [Eqs. (20) and (21)]. In addition, we considered the temporal relaxation of an initial nonequilibrium magnetic moment and its field-mediated rotation [Eqs. (22)–(24)].

(ii) The anomalous Hall effect of charge carriers was analyzed, which exists, when an initial homogeneous magnetic moment exists [Eqs. (30) and (31)].

(iii) The frequency-dependent spin-Hall current was calculated [Eq. (38)] and compared with previous results.

(iv) The steady-state spatial distribution of the magnetic moment near the boundary was considered. For weak electric fields, the spin moment executes damped oscillations up to a depth of the order of K^{-1}. This penetration depth decreases by the factor $E_c/(KE)$ with increasing electric field strength [cf., Eq. (46)].

(v) It was demonstrated that the diffusion of a particle packet generates a spin-magnetic moment. For zero electric field, the induced magnetic moment lies in the plane of the 2DEG and its divergence vanishes. Under the influence of an electric field, all components of the spin moment are nonzero and the magnetic charge does not vanish [Eqs. (58) and (59)].

In spite of these theoretical findings, an experimental study of spin-related phenomena in the hopping transport regime would be worthwhile. Recently, transport mechanisms via polaron hopping have been identified in the hexaboride compounds $Eu_{1-x}Ca_xB_6$.[43]

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