Semiclassical theory of the photogalvanic effect in non-centrosymmetric systems

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We develop a semiclassical theory of nonlinear transport and the photogalvanic effect in non-centrosymmetric media. We show that terms in semiclassical kinetic equations for electron motion which are associated with the Berry curvature and side jumps give rise to a dc current quadratic in the amplitude of the ac electric field. We demonstrate that the circular photogalvanic effect is governed by these terms in contrast to the linear photogalvanic effect and nonlinear I-V characteristics which are governed mainly by the skew scattering mechanism. In addition, the Berry curvature contribution to the magnetic-field induced photogalvanic effect is calculated.

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I. INTRODUCTION AND PHENOMENOLOGICAL DESCRIPTION

Recently, considerable progress was made in the semiclassical theory of the anomalous Hall effect (see the review article, Ref. [1]). The renewed theory based on the generalized semiclassical Boltzmann equation removes the previous controversies surrounded this effect. This theory takes into account the following mechanisms of the anomalous Hall effect: (i) the skew scattering contribution which appears on going beyond the Born approximation, (ii) the contribution to the electron velocity due to the Berry curvature in the electron Bloch wavefunction and the contributions of side-jumps, i.e., coordinate shifts at the scattering events, (iii) to the electron velocity and (iv) to a change of the electron energy upon a scattering in the presence of an external electric field. In the light of this advance in linear transport physics, the semiclassical approach to nonlinear transport should be revised as the next natural step. In the present paper we develop a semiclassical theory of a dc electric-current response quadratic in an external dc or ac electric field. The former response is a nonlinear contribution to the I-V characteristics while the latter can be interpreted as a low-frequency photogalvanic effect. The microscopic theories of photogalvanic effects induced by light waves of sufficiently high frequencies lying in the terahertz, infrared or visible regions are based on quantum-mechanical calculations. They make allowance for both the skew and side-jump mechanisms (usually called respectively the ballistic and the shift contributions) and have no need for the introduction of the Berry curvature, for details see the books Refs. [10][11]. On the other hand, the existing theories of static currents quadratic in the electric field regard the skew contributions only, see Refs. [1][15]. The shift contribution to the I-V characteristics has been considered in the regime of streaming achieved in strong electric fields. As for the Berry-curvature mechanism of nonlinear transport, to the best of our knowledge, it has not been analyzed yet.

Phenomenologically, the electric current density \( j \) which is quadratic in the electric field is described by

\[
j_{\lambda} = \Lambda_{\lambda \mu \nu} E_{\mu} E_{\nu}^* + T_{\lambda \mu \nu \eta} E_{\mu} E_{\eta}^* q, \tag{1}\]

where \( \Lambda \) and \( T \) are material tensors, \( E \) and \( q \) are the electric-field amplitude and the wave vector of the ac electromagnetic wave, \( \lambda, \mu, \nu \) and \( \eta \) are the Cartesian coordinates. In the particular limit of a static field (the wave frequency \( \omega \rightarrow 0 \)) the amplitude \( E \) reduces to the dc electric field, and \( q \rightarrow 0 \). The second term in the right-hand side of Eq. (1) describes the photon drag effect. It was first observed and described as the high-frequency Hall effect by Barlow. In the quantum-mechanical description of optical transitions, the drag effect arises due to momentum transfer from photons to charge carriers. This current can be induced in both centro- and non-centrosymmetric systems. The semiclassical theory of this effect is well developed, see, e.g., Refs. [20][21] and the review Ref. [22], and here we focus completely on the first term in the right-hand side of Eq. (1). It can be conveniently decomposed as

\[
j_{\lambda} = \chi_{\lambda \mu \nu} \{E_{\mu} E_{\nu}^*\}_s + \gamma_{\lambda \mu} (E \times E^*)_{\mu}, \tag{2}\]

where \( \{E_{\mu} E_{\nu}^*\}_s \) is the symmetrized product \( (E_{\mu} E_{\nu}^* + E_{\nu}^* E_{\mu})/2 \). The two contributions in Eq. (2) describe, respectively, the linear and circular photogalvanic effects, or LPGE and CPGE. At \( \omega \rightarrow 0 \), the vector \( E \) is certainly real and the CPGE vanishes while the LPGE reduces to the quadratic conductivity. Since the current density is a polar vector and, therefore, odd under the operation of space inversion, while the square of the electric field is even, the CPGE and LPGE are allowed only in materials that lack inversion symmetry, respectively, in gyrotropic media and piezoelectrics.

We assume that, in addition to the dc or ac electric field, a static magnetic field \( B \) is applied to the sample, so that the coefficients \( \chi_{\lambda \mu \nu} \) and \( \gamma_{\lambda \mu} \) are \( B \)-dependent. In the present work we keep only zero- and first-order terms in \( B \), in which case one has

\[
\begin{align*}
\chi_{\lambda \mu \nu}(B) &= \chi_{\lambda \mu \nu}^{(0)} + \beta_{\lambda \mu \nu \eta} B_\eta, \\
\gamma_{\lambda \mu}(B) &= \gamma_{\lambda \mu}^{(0)} + \beta_{\lambda \mu \eta \nu} B_\eta. \tag{3}
\end{align*}
\]
Since the homogeneous magnetic field is invariant under space inversion the tensors $\beta^\perp$ and $\beta^C$ are also nonzero only in non-centrosymmetric media.

II. GENERALIZED BOLTZMANN EQUATION

Since the photon drag current is neglected we assume the $ac$ electric field $E(t)$ to be homogeneous and ignore effects related to the magnetic field of the electromagnetic wave. This means that the electron distribution function $f_P(t)$ is independent of the spatial coordinate $r$. Since in this paper we ignore the spin-orbit interaction this function is also spin-independent. The kinetic equation for $f_P(t)$ has the standard form

$$\frac{\partial f_P}{\partial t} + \dot{p} \cdot \frac{\partial f_P}{\partial p} = I_P\{f\},$$

(4)

where $\dot{p}$ is the time derivative of the momentum of the center of mass of the electron wavepacket, and $I_P\{f\}$ is the collision term. The latter consists of two terms, $I_p^{(el)}\{f\}$ and $I_p^{(inel)}\{f\}$, representing the elastic scattering by static defects or disorder and inelastic scattering by phonons. For simplicity we assume the typical elastic-scattering time to be much shorter than that for inelastic scattering, $\tau_{inel}$. In this case $I_p^{(el)}\{f\}$ and $I_p^{(inel)}\{f\}$ describe, respectively, the electron momentum and energy relaxation. Taking into account the Berry curvature, side-jumps under electron scattering, and skew scattering, the equations for $\dot{p}$ and $I_P\{f\}$ are modified as compared to the classical Boltzmann equation. Following Refs. 3, 12 (see also Ref. 1) we expand the term $I_p^{(el)}\{f\}$ into powers of the scattering potential $V(r)$ and write it as a sum of the scattering rate found in the lowest, second-order, approximation

$$I_p^{(el,2)}\{f\} = \sum_{p'} W^{(2)}_{p'p}(f_{p'} - f_p),$$

(5)

$$W^{(2)}_{p'p} = \frac{2\pi}{\hbar} \left| \langle W_{p'p} \rangle \right|^2 \delta[\varepsilon_{p'} - \varepsilon_p - eE(t) \cdot r_{p'p}],$$

(6)

and the antisymmetric part of the scattering rate (the skew contribution)

$$I_p^{(el,a)}\{f\} = \sum_{p'} W^{(a)}_{p'p}(f_{p'} - f_p)$$

(7)

calculated beyond the Born approximation (third- and higher-order corrections): $W^{(a)} = -W^{(a)}_{p'p}$. Here $V_{p'p}$ is the transition matrix element, $\varepsilon_p$ is the electron energy, and the angle brackets indicate averaging over the random scattering potential. One can see that the $\delta$-function in Eq. (4) contains a change of energy of the scattered electron under the side-jump $r_{p'p}$ in the presence of an external electric field $E$, see Ref. 8. Note that the inelastic collision term $I_p^{(inel)}\{f\}$ also contains an asymmetric part. Moreover, in the presence of a magnetic field the asymmetric scattering is allowed even in the Born approximation.$^{23}$

A. Zero magnetic field

In the absence of an external magnetic field one has $\dot{p} = eE(t)$, where $e$ is the electron charge. The electric current density reads

$$j = \frac{2e}{V_d} \sum_p \dot{r}_p f_p.$$  

(7)

Here $V_d$ is the macroscopic volume of the $d$-dimensional sample, $d = 3, 2, 1$ for a bulk sample, quantum well and quantum wire, respectively, and the factor 2 takes into account the electron spin degeneracy. The velocity of the electron wavepacket is given by

$$\dot{r}_p = v_p - \dot{p} \times F_p + \delta\dot{r}_p,$$  

(8)

where $v_p = \partial\varepsilon_p/\partial p$ is the electron conventional velocity. The first two terms in Eq. (8) were derived in Refs. 3, 4, 5 while the third term was derived in Refs. 8, 24 (see also Ref. 11 for a review). The Berry curvature $F_p$ is defined by $^{3, 4, 5}$

$$F_p = \frac{\partial}{\partial p} \times \Omega(p), \quad \Omega(p) = i \left( u_k |\partial u_k/\partial k| \right)$$  

(9)

with $k = p/\hbar$, and $u_k(r)$ being the periodic amplitude of the Bloch wavefunction. The spatial shift of an electron under the scattering $p \rightarrow p'$ has the form$^{5, 24}$

$$r_{p'p} = -\frac{\text{Im} \langle W_{p'p}^* \hat{D}_{p'p} V_{p'p} \rangle}{\langle |V_{p'p}|^2 \rangle} + \Omega(p') - \Omega(p),$$  

(10)

where

$$\hat{D}_{p'p} = \hbar \left( \frac{\partial}{\partial p} + \frac{\partial}{\partial p'} \right).$$

B. Nonzero static magnetic field

The equation for $\dot{p}$ is modified in a magnetic field $B$ into$^{15}$

$$\dot{p} = e \left[ E(t) + \frac{\dot{r}_p}{c} \times B \right],$$  

(11)

where $\dot{r}_p$ is defined by Eq. (8).

Taking into account the Berry-curvature-induced renormalization of phase-space volume$^{22}$ the equations
for the electron and electric current densities, \( N_d \) and \( j \), are also modified as follows

\[
N_d = \frac{2}{V_d} \sum_p \left( 1 - \frac{e}{c} B \cdot F_p \right) f_p ,
\]

\[
j = \frac{2e}{V_d} \sum_p \left( 1 - \frac{e}{c} B \cdot F_p \right) \dot{r}_p f_p .
\]

(12)

Due to the same reason, in Eq. (5) one should perform the replacement

\[
\sum_{p'} \to \sum_{p'} \left( 1 - \frac{e}{c} B \cdot F_{p'} \right) .
\]

(13)

Below we take into account contributions to the current \([7]\) up to the first order in \( F_p \) and \( r_{p'} p \). This allows one to approximate the \( \delta \)-function in Eq. (8) by

\[
\delta[\varepsilon_{p'} - \varepsilon_p - eE(t) \cdot r_{p'} p] = \delta(\varepsilon_{p'} - \varepsilon_p) + eE(t) \cdot r_{p'} p \frac{\partial}{\partial \varepsilon_p} \delta(\varepsilon_{p'} - \varepsilon_p) .
\]

(14)

Expanding \( \dot{r}_p \) and \( \dot{p} \) up to the first order in \( F_p \) we can present the solution of the coupled equations \([8]\) and \([11]\) in the following form

\[
\dot{r}_p = \dot{v}_p \left( 1 + \frac{e}{c} B \cdot F_p \right) - eE(t) \times F_p - \frac{e}{c} (\dot{v}_p \cdot F_p) B + \dot{\delta}_p ,
\]

\[
\dot{p} = e \left[ E(t) \left( 1 + \frac{e}{c} B \cdot F_p \right) + \frac{\dot{v}_p + \delta \dot{r}_p}{c} \times B - \frac{e}{c} (E(t) \cdot B) F_p \right] .
\]

(15)

### III. NONLINEAR TRANSPORT IN THE ABSENCE OF MAGNETIC FIELD

The electron distribution function \( f_p \) is expanded in powers of the electric-field amplitude \( E \) defined by

\[
E(t) = E e^{i\omega t} + E^* e^{-i\omega t} .
\]

(16)

For the classical Boltzmann equation in the relaxation-time and effective-temperature approximations one has

\[
f_p^{(1)}(t) = e^{-i\omega t} f_{p\omega}^{(1)} + \text{c.c.} , \quad f_{p\omega}^{(1)} = -e\tau_\omega (E \cdot v_p) f_p^{(0)}
\]

(17)

for the linear correction, and

\[
f_p^{(2)} = f_0(\varepsilon_p, \Theta) - f_0(\varepsilon_p) + f_p^{(2)}(p) ,
\]

\[
f_p^{(2)}(p) = 2e^2 \tau \text{Re}(\tau_\omega) f_0^n(v_{p\mu} v_{p\nu} - v_{p\mu} \delta_{\mu\nu}) \{ E_\mu E^*_\mu \} ,
\]

(18)

for the time-independent second-order correction. Here the momentum relaxation time \( \tau \) is defined by

\[
I_p^{(2)} \{ f \} = -\frac{f_p - f_p^{(0)}}{\tau} ,
\]

(19)

where the bar means averaging over the directions of \( p \), hereafter for simplicity we neglect the dependence of \( \tau \) on the electron energy \( \varepsilon_p \), \( f_0(\varepsilon_p, \Theta) \) is the Fermi-Dirac distribution function at the effective temperature \( \Theta \) different from the bath temperature \( T \) due to the heating of the electron gas, \( f_0(\varepsilon_p) = f_0(\varepsilon_p, T) \), a prime means derivative over \( \varepsilon_p \): \( f'_0 = \partial f_0(\varepsilon_p)/\partial \varepsilon_p \), and

\[
\tau_\omega = \frac{\tau}{1 - i\omega \tau} .
\]

In the following subsections we will take into account skew-scattering, Berry-curvature, and side-jump effects, find corrections to the electron distribution function given by the sum of \([17]\) and \([18]\), substitute these corrections into Eq. \([7]\) and find the electric current proportional to bilinear products \( E_p E^*_p \) in accordance with the phenomenological equation \([2]\). It should be mentioned that, for an \( ac \) electric field, the current \([2]\) or \([7]\) is defined as the time average. Taking into account the definition \([10]\) of the field amplitude the tensor components \( \chi_{\mu\nu}(\omega) \) in Eq. \([2]\) found in the limit \( \omega \to +0 \) and those for the static electric field differ by a factor of 2.

#### A. Skew-scattering contribution

In this subsection we set all vectors \( r_{p'p} \) and \( F_p \) to zero so that the nonlinear current appears only due to the asymmetrical parts of elastic and inelastic scattering rates, respectively, \( I_{p}^{(el,a)} \{ f \} \) and \( I_{p}^{(inel,a)} \{ f \} \) and one has

\[
j = \frac{2e\tau}{V_d} \sum_p v_p \left[ I_{p}^{(el,a)} \{ f^{(2)} \} + I_{p}^{(inel,a)} \{ f_0(\varepsilon_p, \Theta) \} \right] .
\]

(20)

The second term contributes to the current only in systems possessing a polar axis.

Equation \([20]\) demonstrates that the skew-scattering processes give rise only to LPGE. The current proportional to the degree of circular polarization of the electric field does not appear due to skew scattering.

#### B. Berry-curvature related current

Let us now ignore both the asymmetry of collision rate and side-jumps at the scattering and analyze the effect of
the Berry curvature on the nonlinear current. Note that in centrosymmetric systems $F_p = F_{-p}$, while in systems with time reversal symmetry $F_p = -F_{-p}$. Thus if the system is both time- and centrosymmetric the Berry curvature vanishes, $F_p \equiv 0$.

The nonlinear current is obtained by taking into account linear-in-$E$ corrections to the velocity $\mathbf{v}_p$ and to the distribution function, see Eqs. (15) and (17), and time averaging the sum

$$ j = \frac{2e^2}{V_d} \sum_p F_p \times E(t) f_p^{(1)}(t). $$

The result written in terms of the tensors $\chi^{(0)}_{\lambda \mu \nu}$ and $\gamma^{(0)}_{\lambda \mu}$ reads

$$ \chi^{(0)}_{\lambda \mu \nu} = \frac{2e^3}{V_d} \text{Re} (\tau_\omega) \sum_p f_p^{(0)} (\epsilon_{\lambda \mu \eta} C_{\nu \eta} + \epsilon_{\lambda \eta \mu} C_{\nu \eta}), $$

$$ \gamma^{(0)}_{\lambda \mu} = \frac{2e^3}{V_d} \text{Im} (\tau_\omega) \sum_p f_p^{(0)} C_{\lambda \mu}, $$

where $\epsilon_{\lambda \mu \eta}$ is the antisymmetric unit third-rank tensor and

$$ C_{\lambda \mu}(\epsilon_p) = v_{\lambda \mu} F_{\mu} p. $$

While deriving the equation for $\gamma^{(0)}_{\lambda \mu}$ we took into account that the sum $\sum_p C_{\lambda \mu}$ makes no contribution to the current because $\nabla_p \cdot (\nabla_p \times \Omega(p)) \equiv 0$.

In the particular case of $d$-dimensional degenerate electron gas with a parabolic dispersion $\epsilon_p = p^2/2m^*$ ($m^*$ is the electron effective mass) where $k_B T \ll E_F$ and $f_p^{(0)} \approx -\delta(\epsilon_p - E_F)$ with $E_F$ being the Fermi energy, Eqs. (21) are reduced to

$$ \chi^{(0)}_{\lambda \mu \nu} = \frac{em^* d}{2E_F} (\epsilon_{\lambda \mu \eta} C_{\nu \eta} + \epsilon_{\lambda \eta \mu} C_{\nu \eta}) \text{Re} (\sigma_\omega), $$

$$ \gamma^{(0)}_{\lambda \mu} = \frac{-em^* d}{2E_F} C_{\lambda \mu} \text{Im} (\sigma_\omega), $$

where $\sigma_\omega$ is the linear Drude conductivity,

$$ \sigma_\omega = \frac{N_d e^2 \tau_\omega}{m^*}, $$

and the value of $C_{\lambda \mu}(\epsilon_p)$ is taken at the Fermi energy.

Symmetry analysis allows one to find linearly independent coefficients in the expansion of the Berry curvature $F_p$ in powers of the momentum $p$. To illustrate, below we give examples of such analysis for the bulk crystal symmetries $T_d$ (zinc-blende lattice) and $C_{6v}$ (wurtzite) and the point symmetry $C_2$ of a quantum-well structure grown along the low-symmetry axis $[hhl]$ different from [001], [111] or [110].

In the $T_d$ point group the components of the polar vector $v_p \propto p$ and axial vector $F_p$ transform according nonequivalent irreducible representations $F_2$ and $F_1$. Since the direct product of the representations $F_2$ and $F_1$ does not contain the identity representation $A_1$ the average of the product $v_{\lambda \mu} F_{\mu} \overline{p}$ over the directions of $p$ vanishes, and the Berry-curvature related current is forbidden for the $T_d$ symmetry. The zero value of $\gamma^{(0)}_{\lambda \mu}$ agrees with the fact that the $T_d$ crystal class is not gyrotropic and forbids the circular PGE.

In crystals of the $C_{6v}$ symmetry with $z$ being the polar axis the components $v_{px}$, $v_{py}$ and $-F_{px}$ transform according to the representation $E_2$ and the nonzero averages $\chi_{\lambda \mu}$ and $\gamma_{\lambda \mu}$ are $C_{zy} = -C_{yz}$. It follows then that nonzero components of the photogalvanic tensors are $\chi^{(0)}_{xzy} = \chi^{(0)}_{xyz} = \chi^{(0)}_{zxy}$ and $\gamma^{(0)}_{xzy} = -\gamma^{(0)}_{zyx}$, and they can be expressed via $C_{xy}(\epsilon_p)$ by Eqs. (21). Expanding $F_p$ to first order in $p$ we have

$$ F_p = A \mathbf{\hat{c}} \times p, $$

where $\mathbf{\hat{c}}$ is a unit vector along the polar axis $z$ and $A$ is a constant. Note that a similar equation follows for the continuous group $C_{6\infty}$. From Eq. (23) we obtain

$$ C_{xy}(\epsilon_p) = \frac{2}{3} A \epsilon_p, $$

and using Eqs. (21) we get for arbitrary degeneracy of the electron gas

$$ \chi^{(0)}_{xxy} = A e m^* \text{Re} (\sigma_\omega), \quad \gamma^{(0)}_{xxy} = -A e m^* \text{Im} (\sigma_\omega). $$

In the band-structure model of a wurtzite-type semiconductor including the conduction band $\Gamma_{1c}$, and the valence bands $\Gamma_{0v}, \Gamma_{1v}$, we obtain the following estimation for the constant in Eq. (23)

$$ A = \frac{4 \hbar P \lambda P |Q|}{E_g \Delta_c (E_g + \Delta_c)}. $$

Here

$$ P_{\pm} = \frac{-i}{m_0} \langle S|p_x|X \rangle = \frac{-i}{m_0} \langle S|p_y|Y \rangle, \quad P_{||} = \frac{-i}{m_0} \langle S|p_z|Z \rangle, $$

$$ Q = \frac{-i}{m_0} \langle Z|p_x|X \rangle = \frac{-i}{m_0} \langle Z|p_y|Y \rangle, $$

$m_0$ is the free electron mass, $p_x = -i \hbar \partial / \partial x$, $E_g$ is the fundamental energy gap, $\Delta_c$ is the crystal splitting, and $S$, $(X,Y)$ and $Z$ are the $\Gamma$-point Bloch functions in the $\Gamma_{1c}$, $\Gamma_{0v}$ and $\Gamma_{1v}$ bands, respectively. Note that the polar crystal symmetry is fixed by a nonzero matrix element $Q$.

In a two-dimensional system of the $C_2$ symmetry with the interface plane normal to $z$ and the mirror reflection plane $(yz)$ a single nonzero component of $F$ is $F_{p,z}$. The first-order term is given by

$$ F_{p,z} = A_s p_x, $$

where $A_s$ is a constant. As a result one has

$$ C_{\lambda \mu}(\epsilon_p) = A_s \epsilon_p \delta_{\lambda x} \delta_{\mu z} $$

and one can readily use Eqs. (21) to relate the photogalvanic tensors to the real and imaginary parts of the two-dimensional conductivity $\sigma_\omega$. 

C. Shift currents

In the semiclassical theory, the shift current consists of two contributions, the first one is due to the scattering-induced correction to the electron velocity, see Eq. [8], and has the form

$$j^{(\text{vl})} = \frac{2e}{V_d} \sum_p \delta f_p J^{(2)}_p (p),$$

(27)

while the other is related to the field-induced correction to the collision term and can be presented by the time average of

$$j^{(sc)} = \frac{2e}{V_d} \sum_p v_p \delta f_p,$$

where

$$\delta f_p = -2\varepsilon \sum_{p'} [j_p^{(1)}(t) - f_p^{(1)}(t)] \begin{pmatrix} E(t) \cdot r_{p'p} \end{pmatrix}$$

$$\times \frac{2\pi}{\hbar} \langle |V_{p'p}|^2 \rangle \frac{\partial}{\partial \varepsilon_p} \delta (\varepsilon_p - \varepsilon_{p'}) .$$

(28)

Taking into account the identity

$$v_p \frac{\partial S(\varepsilon_p)}{\partial \varepsilon_p} = \frac{\partial S(\varepsilon_p)}{\partial p}$$

the second contribution may be reduced into

$$j^{(sc)} = \frac{4\pi e^2\tau}{hV_d^2} \sum_{p'p''} \delta (\varepsilon_p - \varepsilon_{p'})$$

$$\times \frac{\partial}{\partial p} \left[ (f_{p''}^{(1)} - f_{p''}^{(1)}) \langle |V_{p'p}|^2 \rangle \begin{pmatrix} E^* \cdot r_{p'p} \end{pmatrix} \right] + \text{c.c.} .$$

(29)

We ignore the shift-induced corrections to antisymmetric part of the scattering rate $W^{(0)}$ because the combined effect of the Berry curvature and side-jumps is negligible.

In order to obtain simple analytical equations for the photogalvanic tensors, we simplify the model and assume that the scattering matrix element $V_{p'p}$ is the sum of a main term $V_0$ independent of the electron momenta, and an additional term $\delta V_{p'p}$ dependent on the momenta, governing the spatial shift [10], but making no influence on the relaxation time $\tau$. In this model the current $j^{(sc)}$ can be written as

$$j^{(sc)} = \frac{2e^3}{V_d^2 \tau \omega} \sum_{pp'} W^{(2)}_{p'p}$$

$$\times \frac{\partial}{\partial p} \left[ \begin{pmatrix} E \cdot \left( \frac{\partial f_0(\varepsilon_p)}{\partial p} - \frac{\partial f_0(\varepsilon_{p'})}{\partial p'} \right) \end{pmatrix} \begin{pmatrix} E^* \cdot r_{p'p} \end{pmatrix} \right] + \text{c.c.} .$$

(30)

The further simplifications become possible if we expand the shift [10] in powers of $p', p$ and retain quadratic terms. The latter are the lowest nonvanishing terms taking into account the following properties of the polar vector $r_{p'p'}$: spatial shifts under electron scattering are symmetrical functions of the momenta, $r_{-p'} = r_{p'p}$, and reverse under the interchange of $p'$ and $p$, $r_{p'p} = -r_{p'p}$.

For bulk $T_d$ and $C_{6v}$-symmetry crystals the second-order terms are

$$r_{p'p} = \eta \left( p_z p_x - p_x p_z \right) \left( C_{6v} \right),$$

$$r_{p'p; x} = \eta \left( p_x p_y - p_y p_x \right) \left( T_d \right),$$

(31)

and two other components are obtained by cyclic permutation.

For a quantum well of the $C_s$ symmetry, the elementary spatial shift has the form

$$r_{p'p} = \left[ \eta_1 \left( p_x^2 - p_y^2 \right) + \eta_2 \left( p_y^2 - p_z^2 \right) \right] \hat{y}$$

$$+ \eta_3 \left( p_x p_y - p_y p_x \right) \hat{x} ,$$

(32)

where $\hat{x}$ and $\hat{y}$ are the unit vectors along the $Ox$ and $Oy$ axes, respectively. By using Eqs. [27], [31] and [32] we obtain the following nonzero tensor components for a heterostructure of the $C_s$ symmetry

$$\chi^{(0)}_{xzx}(\text{vl}) = -\chi^{(0)}_{yzy}(\text{vl}) = 2(\eta_1 - \eta_2) e^* \text{Re}(\sigma_\omega) ,$$

$$\chi^{(0)}_{xzy}(\text{vl}) = \eta_3 e^* \text{Re}(\sigma_\omega) ,$$

$$\chi^{(0)}_{yzy}(\text{sc}) = 2 \chi^{(0)}_{xzx}(\text{sc}) = 2(\eta_1 + \eta_2) e^* \text{Re}(\sigma_\omega) ,$$

$$\chi^{(0)}_{xzy}(\text{sc}) = (\eta_1 + \eta_2) e^* \text{Im}(\sigma_\omega) .$$

(33)

Since the tensor $\chi^{(0)}_{\lambda\mu\nu}$ is symmetrical with respect to interchange of the second and third indices the components $\chi^{(0)}_{xzx}(\text{vl})$ and $\chi^{(0)}_{xzy}(\text{sc})$ are also nonzero.

IV. NONLINEAR TRANSPORT IN THE PRESENCE OF MAGNETIC FIELD

Here we outline the derivation of the Berry-curvature related photocurrent linear in a static magnetic field $B$. In the presence of a magnetic field the kinetic equation has the form

$$\frac{\partial f_p}{\partial t} + \vec{p} \cdot \frac{\partial f_p}{\partial \vec{p}} = \sum_{p'} \left( 1 - \frac{e}{c} \vec{B} \cdot \vec{F}_{p'} \right) W^{(2)}_{p'p}(f_{p'} - f_p),$$

(34)

where $\vec{p}$ is given by Eq. [10]. It is instructive to introduce the auxiliary function

$$\phi_p = \left( 1 - \frac{e}{c} \vec{B} \cdot \vec{F}_p \right) f_p .$$

Then in the linear-in-$B$ approximation we have the following kinetic equation for this function:

$$\frac{\partial \phi_p}{\partial t} + \frac{e^2}{c^2} E \cdot \frac{\partial}{\partial \vec{p}} \left[ \vec{B} \cdot \vec{F}_p \phi_p \right]$$

$$+ e \left[ E + \frac{\vec{p} \times \vec{B}}{c} - \frac{e}{c} (E \cdot \vec{B}) \vec{F}_p \right] \cdot \frac{\partial \phi_p}{\partial \vec{p}}$$

$$= - \frac{1}{\tau} \left[ \phi_p - \phi_0 + \frac{e}{c} (\vec{B} \cdot \vec{F}_p) \phi_0 \right] .$$

(35)
Let us introduce the notations for corrections $\phi_p^{(E^2B^2)}$ to the auxiliary function \( m = 0, 1, 2; n = 0, 1, \ldots \), \( \phi_p^{(E)}, \phi_p^{(B)}, \phi_k^{(EB)} \) etc. From Eq. (12) we find that the following expression for the electric current quadratic-in-\( E \) and linear-in-\( B \)

\[
j^{(E^2B)} = \frac{2e}{V_d} \sum_p \left[ v_p \phi_p^{(E^2B)} \right] - e(E \times F_p)\phi_p^{(EB)} + \frac{e}{c} F_p \times (v_p \times B) \phi_p^{(E^2)}
\]

As an example we further consider a 2D system of \( C_s \) symmetry and take \( F_p \) in the form \( F_p \parallel z, \beta \parallel z, \hat{A}_s \parallel z \). Solving the kinetic equation (35) one can obtain the linear- and quadratic-in-\( E \) corrections at \( B = 0 \) given by Eqs. (17) and (18) and linear-in-\( B \) corrections in the form

\[
\phi_p^{(B)} = -\frac{e}{c} B F_{p, z} f_0(\varepsilon_p)
\]

\[
\phi_p^{(EB)} = \frac{e^3}{c} \tau_0 B_{, z} E_{, z} \cdot \frac{\partial}{\partial p} [F_{p, z} (E \cdot v_p) f_0] + c.c.
\]

In the above expressions, we neglected terms which do not contribute to the current. For the particular case of \( C_s \) symmetry nonzero components of the magnetophotogalvanic tensors \( \beta_{\mu \nu \eta}^l \) and \( \beta_{\nu \mu \eta}^l \) defined according to Eq. (3) become

\[
\beta_{xxxz}^l = \frac{4\beta_0}{1 + (\omega \tau)^2}, \quad \beta_{xyyz}^l = -\frac{4\beta_0}{1 + (\omega \tau)^2}, \quad \beta_{yyxz}^l = 4\beta_0 \frac{2 + (\omega \tau)^2}{1 + (\omega \tau)^2}, \quad \beta_{xzzx}^l = -\beta_{yzyz}^l = 2\beta_0 \frac{1 - (\omega \tau)^2}{1 + (\omega \tau)^2}, \quad \beta_{yxzy}^l = -\beta_{yzyz}^l = \frac{\omega \tau}{1 + (\omega \tau)^2},
\]

where \( \beta_0 = A_s N_2 D E^2/m^* c \).

It follows from the above equations that in the static limit \( \omega = 0 \) the magnetic-field induced corrections are given by

\[
\delta_j = 2A_s e m^* \sigma_H (E_x^2 - E_y^2), \quad \delta_j = 4A_s e m^* \sigma_H E_x E_y
\]

at \( B \parallel z \), and

\[
\delta_j = A_s e m^* \sigma_H E_y E_z, \quad \delta_j = -A_s e m^* \sigma_H E_x E_z
\]

at \( B \parallel y \). Here \( \sigma_H \) is the Hall conductivity equal to \( \omega_c \sigma_0 \), where \( \omega_c \) is the cyclotron frequency and \( \sigma_0 \) is the Drude conductivity at \( \omega = 0 \).

### V. DISCUSSION AND CONCLUSION

We have developed a semiclassical theory of nonlinear transport and the photogalvanic effects in non-centrosymmetric media. It has been shown that the terms in semiclassical kinetic equations describing the electron motion associated with the Berry curvature and elementary shifts in the real space contribute to the linear and circular photogalvanic effects as well as to the quadratic I-V characteristics (as a static limit of the LPGE). Previously Bloch et al.\[14,15\] studied the skew scattering mechanism of the LPGE. However, since it makes no contribution to the CPGE the Berry-curvature and shift related mechanisms certainly dominate the current sensitive to the chirality of the electromagnetic wave.

It should be noted that we have considered linear-in-electric-field corrections to the collision integral due to the quantum coordinate shifts, see the term \( -eE(\xi) \cdot r_{j\prime}^p \) in the \( \delta \)-function of Eq. (6). A comparable contribution to the tensors \( \chi \) and \( \gamma \) comes from linear-in-\( E \) corrections to the squared matrix element \( \langle |V_{j\prime\prime}^p|^2 \rangle \) allowed in non-centrosymmetric systems.\[25\]

The following estimations summarize different contributions and allow one to compare their relative role in the formation of a \( dc \) current as a quadratic response to an \( ac \) electric field. The skew scattering contribution can be estimated as

\[
J_{sk} \sim e N_d \bar{v} \xi_{sk}^{(imp)} \frac{f(2)}{f_0}
\]

or

\[
\chi_{sk} \sim \bar{\xi}_{sk}^{(imp)} \text{Re}(\sigma \omega \bar{e}/\bar{\xi}),
\]

where \( \bar{v} \) and \( \bar{\xi} \) are the characteristic electron velocity and energy \( (\bar{\varepsilon} \gg \hbar) \), \( l = \bar{\varepsilon} \tau \) is the mean free path, and the dimensionless parameter of the scattering asymmetry is given by, see Eq. (20),

\[
\xi_{sk}^{(imp)} = \frac{\bar{v}_{p, l} \Phi_p}{|v_p|^2} \left\{ \frac{v_p v_p}{|v_p|^2} \right\},
\]

where \( \bar{v}_{p, l} \) is the electron momentum relaxation time due to scattering by acoustic phonons, \( \xi_{sk}^{(imp)} \) is the dimensionless asymmetry parameter of the electron-phonon interaction including the deformation-potential and piezoelectric mechanisms, see, e.g., Ref.\[20\].

The Berry-curvature and shift-related contributions to LPGE, \( \chi_B \) and \( \chi_{sh} \), are given by the constants \( A \) and \( \eta \),
respectively, see Eqs. (24) and (33). In semiconducting pyroelectric materials one can use a general estimation \( A \sim \eta \sim \xi_{\text{piro}} a_0/(m^*E_g) \), where \( a_0 \) is lattice constant and \( \xi_{\text{piro}} \) is the dimensionless asymmetry parameter. As a result, we have
\[
\chi_B \sim \chi_{sh} \sim \xi_{\text{piro}} \text{Re}(\sigma_\omega) \frac{e a_0}{E_g}.
\] (45)
For a wurzite-type semiconductor one has from Eq. (20)
\[
\xi_{\text{piro}} a_0 \sim \hbar Q/\Delta_c.
\]
It follows from Eqs. (13) and (15) that, for comparable asymmetry parameters \( \xi_{\text{sk}} \) and \( \xi_{\text{piro}} \), the skew-scattering contribution to the LPGE prevails over the Berry-curvature and shift related contributions.

On the other hand, the Boltzmann kinetic equation yields no contribution to the CPGE even if skew scattering is included: \( \gamma_{sk} = 0 \). The circular nonlinear current appears only if \( F_p \) and/or \( r_{p'p} \) are nonzero in which case one has
\[
\gamma_B \sim Aem^* \text{Im}(\sigma_\omega), \quad \gamma_{sh} \sim \eta m^* \text{Im}(\sigma_\omega).
\]
In the static limit both values \( \gamma_B \) and \( \gamma_{sh} \) as expected vanish. At finite frequencies, however, the Berry-curvature and shift-related effects provide the main contribution to the CPGE.

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