Pure spin photocurrents

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Abstract
The pure spin currents, i.e., the counterflow of particles with opposite spin orientations, can be optically injected in semiconductors. Here, we develop a phenomenological theory, which describes the polarization dependences of spin currents excited by linearly polarized light in bulk semiconductors and quantum well structures of various symmetries. We present microscopic descriptions of the pure spin photocurrents for interband optical transitions in undoped quantum wells as well as for direct intersubband and indirect intrasubband (Drude-like) transitions in n-doped quantum well structures. We also demonstrate that pure spin currents can be generated in structures of sufficiently low symmetries by simple electron gas heating. The theoretical results are compared with recent experimental observations.

1. Introduction
By definition, the pure spin current of free carriers, electrons or holes, is a spin flux without an electric current. It can be conceived as formed by opposing equivalent flows of spin-up and spin-down particles. This non-equilibrium distribution of carriers in the wave vector and spin spaces is characterized by zero charge current, because electric currents contributed by spin-up and spin-down particles cancel each other, but leads to the accumulation of opposite spins at the opposite edges of the sample. In particular, a pure spin current is produced in a direction perpendicular to an applied electric field due to spin-dependent skew or side-jump scattering of electrons by impurities or phonons. This effect known as the spin Hall effect was predicted in early theoretical studies [1, 2] and observed in recent years, see [3–5].

In this study, we present the theory of pure spin currents generated under the light absorption in an unbiased semiconductor structure. In spin-dependent optical spectroscopy, the efforts were mostly directed toward (i) the photogeneration of non-equilibrium spin polarization of carriers, the effect known as the optical orientation (see, e.g., [6, 7]), and (ii) the generation of spin-sensitive electric currents known as the circular photogalvanic, spin-galvanic and magneto-gyrotropic effects [7, 8]. In contrast, the free carriers participating in a pure spin current neither have a net spin polarization nor produce a net charge current. A spatial separation of electron spins caused by the spin photocurrent was first observed by using two-color optical coherence control techniques, due to quantum interference of one- and two-photon absorption of two orthogonally-polarized overlapping laser pulses with frequencies ω and 2ω [9–11]. Then, Bhat et al [12] and Tarasenko and Ivchenko [13] showed that merely one-photon absorption of linearly polarized light should produce pure spin currents in noncentrosymmetric bulk semiconductors and quantum well (QW) structures: in this case there is no net motion of charge but spin-up and spin-down photoelectrons travel in opposite directions. The theoretical prediction was followed by an observation of pure spin currents induced by a single linearly polarized optical pulse in (1 1 0)-oriented GaAs QWs [14].

Theoretically, the two-color generation and control of spin currents have been extensively analyzed [12, 15–17]. Here we will consider, in order, the one-photon generation of pure spin currents in unbiased structures under interband, intersubband and intraband absorption of linearly polarized or unpolarized light and derive equations for the corresponding currents. Particularly, we compare different mechanisms of pure spin currents and show difference in the behavior of their contributions as a function of the light frequency and the polarization direction with regard to the crystallographic axes. We also show that pure spin currents emerge in QW structures as soon as the electron gas is simply driven out of thermal equilibrium with the crystal lattice. Finally, we will discuss a new phenomenon which can be called the pure valley-orbit current and observed in many-valley semiconductors. The role of spin-up and spin-down states in pure spin currents is replaced in the valley-orbit current by the index of the conduction-band valleys: the valleys are equally populated, there is no net charge current, but the electrons in different valleys travel in different directions.
2. Phenomenology

Phenomenologically, the spin flux, or, in general, the flux of angular momentum, is described by a second-rank pseudotensor $\mathbf{J}$ whose components $J_{\mu}^\nu$ stand for the flow in the $\beta$-direction of spins oriented along $\alpha$, with $\alpha$ and $\beta$ being the Cartesian coordinates. Nonzero components of the photoinduced spin current $\mathbf{J}$ are determined by the light polarization and the explicit form of spin–orbit interaction governed by the structure symmetry. They can be revealed from the symmetry analysis which requires no knowledge about microscopical mechanisms of the spin current generation. Indeed, in the regime of linear dependence of $\mathbf{J}$ on the light intensity $I$, the spin-photocurrent components are phenomenologically related by

$$J_{\mu}^\nu = I \sum_{y} Q_{a\beta y} e_{y} e_{\mu}^{\ast}$$

(1)

to the light-polarization unit vector $\mathbf{e}$ and the complex conjugate vector $e^{\ast}$. Equation (1) represents the most general form of the spin-photocurrent description because the set of quadratic terms $e_{y} e_{\mu}^{\ast}$ fully determines the light-polarization state.

Equation (1) can be usefully rewritten in the equivalent form

$$J_{\mu}^\nu = I \sum_{y} L_{a\beta y} e_{y} e_{\mu}^{\ast} + e_{\mu}^{\ast} e_{\nu} + I \sum_{y} C_{a\beta y} \mathbf{[e \times e^{\ast}]}_\mu,$$

(2)

where $L_{a\beta y} = (Q_{a\beta y} + Q_{b\alpha y})/2$ is a fourth-rank pseudotensor symmetric in the two indices, $C_{a\beta y} = \sum_{\gamma} Q_{a\gamma y} e_{\gamma} e_{\beta} / (2I)$ is a third-rank tensor and $e_{y} e_{\beta}$ is the completely antisymmetric third-rank pseudotensor or the Levi-Civita tensor. The pseudotensor $L$ describes spin photocurrents which are independent of the sign of light circular polarization for elliptically polarized light and can be conveniently measured for the linearly polarized radiation. In contrast, the tensor $C$ stands for helicity-sensitive spin photocurrents which reverse their polarity upon switching the sign of circular polarization. This occurs because the cross product $\mathbf{[e \times e^{\ast}]}$ is zero for linearly polarized light and proportional to the light helicity for elliptical or circular polarization. Usually, the absorption of circularly polarized light results in a considerable spin photocurrent.

Here, $I$ is the light intensity, the index $\alpha$ runs over the cubic axes $x\parallel[1 0 0]$, $y\parallel[0 1 0]$ and $z\parallel[0 0 1]$ and the index $\alpha + 1$ is obtained by the cyclic permutation of the indices $x$, $y$, $z$. Note that nonzero values of the phenomenological parameters $L_{1}$ and $L_{2}$ in equation (3) are allowed in noncentrosymmetric crystals of $T_d$ symmetry and forbidden for diamond-type centrosymmetric crystals. The symmetry of (001)-oriented QWs grown from zincblende-type semiconductors reduces to the point group $D_{2d}$ in symmetrical structures and $C_{3v}$ in asymmetrical structures. For the latter, the spin current components photoinduced in the $(xy)$-plane are described by ten linearly independent constants as follows:

$$J_{x}^{y} / I = L_{x}^y e_{x} e_{y}, \quad J_{y}^{z} / I = L_{y}^z e_{y} e_{z},$$

$$J_{z}^{x} / I = -L_{z}^x e_{z} e_{x}, \quad J_{x}^{z} / I = -L_{x}^z e_{x} e_{z},$$

(4)

$$J_{x}^{y} / I = -L_{x}^y e_{x} e_{y}, \quad J_{y}^{z} / I = L_{y}^z e_{y} e_{z},$$

$$J_{z}^{x} / I = L_{z}^x e_{z} e_{x}, \quad J_{x}^{y} / I = L_{x}^y e_{x} e_{y}.$$}

Here, the superscript $B$ marks those coefficients which are allowed in QWs of $D_{2d}$ symmetry and can be related to bulk inversion asymmetry (BIA) of the host crystal and/or anisotropy of the chemical bonds at the QW interfaces, while the superscript $S$ marks the contributions which appear because of structure inversion asymmetry (SIA) only. Therefore, in symmetrical (001)-grown QWs, the coefficients $L_{y}^z$ vanish and the polarization dependences of the spin current components are completely determined by the terms proportional to $L_{x}^y$. In the opposite limit, where the SIA predominates and the QW structure can effectively be described by the axial point group $C_{\infty v}$, the spin photocurrent is given by equation (4) with the BIA-related terms being disregarded and the coefficients $L_{y}^z$ satisfying the relation $L_{y}^z = L_{x}^y + L_{z}^x$. It follows from equation (4) that, under normal incidence on a (001)-grown QW, the linearly polarized light can excite fluxes of electron spins oriented only in the interface plane. To create the spin photocurrent components, which can cause the spatial profile of the spin density $J_{S}$, one has to irradiate the QW in the oblique-incidence geometry. This is in contrast to QWs grown along low-symmetry crystallographic axes, where the normally-incident light can induce fluxes of both the in-plane and out-of-plane components of the spin polarization.

As an example of such low-symmetry structures, we consider QWs grown on (110)-oriented substrates and use the $(x', y', z')$ coordinate frame with $z'$ along the growth direction and the in-plane axes $x'\parallel[1 1 0]$ and $y'\parallel[0 0 1]$. Asymmetry-grown (110)-oriented QWs have the point group $C_{4v}$ and contain only two symmetry elements: the identity and a mirror plane. In this particular case, the components of the spin current excited by normally-incident light are phenomenologically given by

$$J_{x}^{y} / I = L_{x}^y e_{x} e_{y}, \quad J_{y}^{z} / I = L_{y}^z e_{y} e_{z},$$

$$J_{z}^{x} / I = -L_{z}^x e_{z} e_{x}, \quad J_{x}^{y} / I = -L_{x}^y e_{x} e_{y},$$

(5)

$$J_{x}^{y} / I = L_{x}^y e_{x} e_{y}, \quad J_{y}^{z} / I = L_{y}^z e_{y} e_{z},$$

$$J_{z}^{x} / I = -L_{z}^x e_{z} e_{x}, \quad J_{x}^{y} / I = -L_{x}^y e_{x} e_{y}.$$
Symmetrical (1 1 0)-grown QWs contain an additional mirror plane \( m_2 = (1 1 0) \) perpendicular to the \( z' \)-axis. Reflection in the plane \( m_2 \) changes the sign of the \( J_{\beta}^{\sigma} \) and \( J_{\beta}^{\sigma'} \) (\( \beta = x', y' \)) components of the spin current but does not modify \( J_\beta^+ \) as well as the in-plane components of the polarization vector \( \mathbf{e} \). Therefore, in symmetrical (1 1 0)-grown QWs, the parameters \( L_1' \ldots L_6' \) vanish and the spin photocurrent is solely described by the last line of equation (5).

Microscopically, the emergence of a pure spin current under light absorption is related to spin–orbit interaction coupling spin states and spatial motion of charge carriers, the latter being directly affected by the electric field of the light. In terms of the kinetic theory, the \( J_{\beta}^{\sigma} \) component of the spin photocurrent in the conduction band is contributed by a non-equilibrium correction \( \alpha \sigma_\alpha k_x \) to the electron spin density matrix, where \( \sigma_\alpha \) is the Pauli matrix and \( \mathbf{k} \) is the wave vector. In general, the concept of spins is uncertain in systems with spin–orbit interaction, since the spin and spin-dependent velocity cannot be determined simultaneously (see, e.g., [18]). Mathematically, it is caused by the fact that the Pauli matrices and the velocity operator do not commute. However, this problem of the spin current definition emerges in high orders in the spin–orbit interaction only and vanishes for the cases where spin currents are directly proportional to the constant of spin–orbit coupling. To the first order in the spin–orbit coupling and within the relaxation time approximation, the components of the pure spin current photoinduced in the conduction band are given by

\[
J_{\beta}^{\sigma} = \sum_k \tau_e \text{Tr} \left[ \frac{\sigma_\alpha}{2} \varphi_{\beta}(\mathbf{k})G(\mathbf{k}) \right],
\]

with the spin-dependent corrections being taken into account either in the velocity operator \( \mathbf{v}(\mathbf{k}) \) or in the photogeneration rate of the spin density matrix \( G(\mathbf{k}) \). Here, \( \tau_e \) is the relaxation time of the spin current which can differ from the conventional momentum relaxation time that governs the electron mobility. Electron–electron collisions between particles of opposite spins, which do not affect the mobility, contribute to the relaxation of pure spin currents reducing the time \( \tau_e \) (see the spin Coulomb drag [19, 20] and the effect of the electron–electron interaction on spin relaxation [21] and references therein).

### 3. Interband transitions in QWs

Among microscopic mechanisms of the pure spin photocurrent we first discuss that related to \( \mathbf{k} \)-linear spin–orbit splitting of quantum subbands [13], in the following the split-subband-related mechanism. It is most easily conceivable for direct transitions between the heavy-hole valence subband \( hh1 \) and conduction subband \( e1 \) in (1 1 0)-grown QWs. In such structures, the spin component along the QW normal \( z' \) is coupled with the in-plane electron wave vector due to the terms proportional to \( \sigma_\alpha k_x \) and \( J_x k_y \) in the conduction and valence bands, respectively, where \( J_x \) is the \( 4 \times 4 \) matrix of the angular momentum 3/2 [7]. This leads to the \( \mathbf{k} \)-linear spin splitting of both the electron subband \( e1 \) and the valence subband \( hh1 \) into branches with the spin projection \( \pm 1/2 \) and \( \pm 3/2 \), respectively, as sketched in figure 1(a). The corresponding dispersions in the subbands at small in-plane wave vector are given by

\[
E_{k_x \pm 1/2}^{(e1)} = \frac{\hbar^2 (k_x^2 + k_y^2)}{2m_e} \pm \gamma_{e}^{(e1)} k_x, \quad E_{k_x \pm 3/2}^{(hh1)} = \frac{\hbar^2 (k_x^2 + k_y^2)}{2m_h} \pm \gamma_{hh}^{(hh1)} k_x,
\]

where \( m_e \) and \( m_h \) are the electron and hole effective masses in the QW plane. Note that the spin splitting of the conduction subband is relativistic and, therefore, small as compared to the nonrelativistic term \( J_x k_y \) describing the splitting of heavy-hole states in (1 1 0)-grown structures.

Due to the selection rules, the allowed direct optical transitions from the valence subband \( hh1 \) to the conduction subband \( e1 \) are \( |+3/2 \rangle \rightarrow |+1/2 \rangle \) and \( |-3/2 \rangle \rightarrow |-1/2 \rangle \) [6], as illustrated in figure 1(a) by vertical lines. Under excitation with linearly polarized or unpolarized light the rates of both transitions are equal. In the presence of spin splitting, the optical transitions induced by photons of the fixed energy \( \hbar \omega \) occur at opposite points of the \( \mathbf{k} \) space for the spin branches \( s_z = \pm 1/2 \). Such an asymmetry of photoexcitation results in a flow of electrons within each spin branch. The corresponding fluxes \( i_{1/2} \) and \( i_{-1/2} \) are of equal strength but of opposite direction. Thus, this non-equilibrium electron distribution is characterized by the nonzero spin current \( (1/2) (i_{1/2} - i_{-1/2}) \) but a vanishing charge current, \( e (i_{1/2} + i_{-1/2}) = 0 \).

To calculate the spin current, we note that the points of optical transitions in the \( \mathbf{k} \) space are determined by the energy and quasi-momentum conservation which reads

\[
E_{g}^{QW} + \frac{\hbar^2 (k_x^2 + k_y^2)}{2\mu} + 2s_z (\gamma_{e}^{(e1)} - \gamma_{hh}^{(hh1)}) k_x = \hbar \omega, \quad (8)
\]

where \( E_{g}^{QW} \) is the QW band gap at \( k = 0 \) and \( \mu = m_m m_h / (m_e + m_h) \) is the reduced effective mass. Owing to spin splitting of the \( e1 \) and \( hh1 \) subbands, electrons are photoexcited into the spin branches \( s_z = \pm 1/2 \) with the average velocities

\[
\langle v_{s_z} \rangle = \frac{\hbar}{m_e} (k_x^0) + 2s_z \gamma_{e}^{(e1)} \hbar = 2s_z \mu \left( \frac{\gamma_{e}^{(e1)} + \gamma_{hh}^{(hh1)}}{m_h / m_e} \right). \quad (9)
\]
The opposite motion of spin-up and spin-down electrons decays within the relaxation time $\tau_e$. However, under the steady-state excitation the electron generation is continuous resulting in the spin current

$$J'_e = \frac{\mu \tau_e}{2\hbar} \left( \frac{Y^{(e)}_{\gamma' e'}(1)}{m_h} + \frac{Y^{(hh)}_{\gamma' e'}}{m_e} \right) \frac{\eta_{\text{CV}}}{\hbar c} I,$$

where $\eta_{\text{CV}}$ is the spin absorbance.

Another contribution to the spin photocurrent may come from k-linear terms in the matrix elements of the interband optical transitions [22], hereafter referred to as the matrix-element-related mechanism. Taking into account $k \cdot p$ admixture of the remote conduction band $\Gamma'_1$ with the valence-band and conduction-band states, $X_k$, $Y_k$, $Z_k$ and $S_k$, respectively, one derives the interband matrix elements of the velocity operator for bulk zinc-blende-type semiconductors [23, 24]:

$$i \langle S_k | e \cdot v | X_k \rangle = (\hbar / m)(e_x + i\beta(e_x k_z + e_y k_y)),$$

$$i \langle S_k | e \cdot v | Y_k \rangle = (\hbar / m)(e_x + i\beta(e_x k_z + e_y k_y)),$$

$$i \langle S_k | e \cdot v | Z_k \rangle = (\hbar / m)(e_x + i\beta(e_x k_z + e_y k_y)),$$

where $P = (\hbar / m_0)(S \cdot p, |Z\rangle$, $P' = i(\hbar / m_0)(S \cdot p, |Z\rangle$, and $Q = i(\hbar / m_0)(X \cdot p, |Z\rangle$ are the interband matrix elements at the $\Gamma$ point of the Brillouin zone, $X'$, $Y'$, $Z'$ are the Bloch functions of the $\Gamma'_1$ band, $m_0$ is the free electron mass, $\beta = QP'P(Q + P'P)$ is a material parameter, $E_x$ is the fundamental band gap, and $E'_x$ is the energy separation between conduction bands $\Gamma'_1$ and $\Gamma'_1$ at the $\Gamma$ point. For GaAs, the coefficient $\beta$ can be estimated as $0.2–1 \AA$ depending on the band parameters used [25, 26].

The k-linear terms in equation (11) do not modify the selection rules for optical transitions from the heavy-hole valence subband to the conduction band. As before, the only allowed transitions are $|+3/2 \rangle \rightarrow |+1/2 \rangle$ and $|-3/2 \rangle \rightarrow |-1/2 \rangle$. However, the rates of the above transitions become dependent of the in-plane wave vector. Particularly, for the linearly polarized light normally incident upon a (1 1 0)-grown QW, the squared moduli of the matrix elements, which determine the optical transition rates, assume the following form in the linear-in-$\beta$ approximation:

$$\langle |+1/2 \rangle | e \cdot v | +3/2 \rangle|^2 = P^2 / (2\hbar^2) \left[ 1 + 2\beta k_x e_x e_y 
- 2\beta k_y (e_y^2 - e_z^2) \right].$$

$$\langle |-3/2 \rangle | e \cdot v | -3/2 \rangle|^2 = P^2 / (2\hbar^2) \left[ 1 - 2\beta k_x e_x e_y 
+ 2\beta k_y (e_y^2 - e_z^2) \right].$$

It follows from equation (12) that, for a fixed light polarization, the spin-up and spin-down electrons are predominantly photoexcited at opposite points in the k space. This is illustrated in figure 1(b) for the light polarized along the $y'$-axis, where electrons with the spin $+1/2$ are generated at a higher rate into states with positive values of $k_y$, whereas electrons with the spin $-1/2$ are mainly generated into states with $k_y < 0$. The difference in rates is shown by vertical lines of different thicknesses. We note that here the spin-orbit splitting of the subbands $h1$ and $l$ is unimportant and, therefore, not shown in figure 1(b) for simplicity. The spin-dependent asymmetry of optical excitation also leads to the pure spin current. Calculation shows that, in (1 1 0)-grown QWs, the components of the spin photocurrent caused by k-linear terms in the matrix elements of optical transitions have the form

$$J'_e^i = \beta(e_x^2 - e_y^2) \frac{\tau_e \eta_{\text{CV}} I}{\hbar c}, \quad J'_e^l = \beta(e_x e_y) \frac{\tau_e \eta_{\text{CV}} I}{\hbar c},$$

where $\varepsilon = (\hbar \omega - E_{gQW})/m_e$ is the kinetic energy of photoexcited electrons. In contrast to equation (10), this contribution depends on the polarization plane of the incident light and vanishes for unpolarized light. From comparison of equations (10) and (13) one can see that, depending on the value of $\hbar \omega - E_{gQW}$, the two contributions to $J'_e$ can be comparable or one of them can dominate over the other. We also note that both the spin current contributions are caused by bulk inversion asymmetry and do not vanish in symmetrically-grown (1 1 0)-QWs.

In (0 0 1)-grown QWs, the absorption of linearly polarized or unpolarized light results in an in-plane flow of electron spins, see equation (4). In contrast to the low-symmetry QWs considered above, in the (0 0 1)-QW structures the linear-in-k terms in the matrix elements of optical transitions from the heavy-hole subband vanish at the normal incidence. Since, in addition, the k-linear spin splitting of the heavy-hole subband is suppressed in (0 0 1)-grown structures [27, 28], we conclude that the spin photocurrents are entirely related to the spin-orbit splitting of the conduction subband. Assuming parabolic spin-independent dispersion in the $h1$ subband and taking into account the spin-dependent contribution

$$H'_{\text{so}} = \sum_{\alpha \beta} \gamma_{\alpha \beta}^{(1)} \sigma_\alpha k_\beta,$$

to the electron effective Hamiltonian in the subband $e1$, the components of the pure spin current generated in the subband $e1$ are derived to be

$$J'^{e1}_\alpha = \gamma_{\alpha \beta}^{(1)} \tau_e \hbar \eta_{\text{CV}} \frac{\hbar c}{2m_h \hbar \omega} I.$$

For the interband transitions from the light-hole subband or the spin-split band $\Gamma_1$, both the split-subband-related and the matrix-element-related mechanisms lead to polarization-dependent pure spin photocurrents. The analysis shows that, in the geometry of normal incidence, the optical excitation from the light-hole subband in (0 0 1)-grown QWs leads to the spin current described by equation (4) where the phenomenological coefficients satisfy the relations $L^h_{\Gamma_1} = L^h_{\Gamma_1} - L^h_{\Gamma_1}$ and $L^h_{\Gamma_1} = L^h_{\Gamma_1} - L^h_{\Gamma_1}$. If the spin photocurrent is solely caused by k-linear terms in the matrix elements of optical transitions then, in addition, $L^h_{\Gamma_1} = 0$ and $L^h_{\Gamma_1} = L^h_{\Gamma_1} = 0$. In the opposite case, when the spin current is mainly contributed by the split-subband-related mechanism, the coefficients are interconnected by $L^h_{\Gamma_1} = \pm L^h_{\Gamma_1}$ and $L^h_{\Gamma_1} = \pm L^h_{\Gamma_1}$ with the sign ‘+’ or ‘-‘ depending on, respectively, whether the spin splitting of the subbands $e1$ or $lh1$ predominates, [28].

The injection of pure spin currents in (1 1 0)-oriented GaAs QWs at room temperature by one-photon absorption of a linearly polarized optical pulse was demonstrated by Zhao et al [14]. Spatially resolved pump–probe technique was used which enabled the authors to obtain signatures of the pure spin currents by measuring the resulting spin separations of
1–4 nm. The pump pulse excited electrons from the valence to the conduction band with an excess energy of ~148 meV. The probe was tuned near the band edge. It was observed that the spin current resulting in separation of the spin density $S_z$ along the $[1\ 1\ 0]$-axis reversed its direction when the polarization of the pump pulse was switched from $\varepsilon_1 \parallel x'$ to $\varepsilon_1 \parallel y'$. This indicates that, for the photon energy used in the experiment, the polarization-dependent contribution dominates over the polarization-independent term.

### 4. Intersubband transitions in n-doped QWs

The intersubband light absorption in n-doped QW structures is a resonant process which becomes possible if the photon energy $\hbar \omega$ is tuned to the intersubband energy separation. In the simple one-band model, direct optical transitions between the electron subbands $e_1$ and $e_2$ conserve spin and are induced only by radiation with a nonzero normal component $e_\perp$ of the polarization vector. If the spin degeneracy of the quantum subbands is lifted, such spin-conserving optical transitions give rise to a pure spin current [13, 29]. This mechanism is illustrated in figure 2, where the intersubband transitions $(e_1, +1/2) \rightarrow (e_2, +1/2)$ and $(e_1, -1/2) \rightarrow (e_2, -1/2)$ are shown by vertical solid lines. Due to $k$-linear spin splitting of the subbands together with the energy and quasi-momentum conservation, the optical transitions induced by light of a fixed frequency occur only at certain values of $k_x$, denoted by $k_{+1/2}$ and $k_{-1/2}$ for the spin states $\pm 1/2$, respectively, where the photon energy $\hbar \omega$ matches the energy spacing between the subbands. As is evident from figure 2(a), these $k_x$-points are of opposite signs for transitions from the spin branches $\pm 1/2$. Similarly to the interband light absorption considered in section 3, such spin-dependent asymmetry of photoexcitation gives rise to pure spin currents in both $e_1$ and $e_2$ subbands.

An interesting feature of the pure spin photocurrent caused by $k$-linear splitting of the subbands is its spectral response. Figures 2(a) and (b) show what happens if the photon energy $\hbar \omega$ crosses the resonance varying from $\hbar \omega < E_{21}$ to $\hbar \omega > E_{21}$, where $E_{21}$ is the energy separation between the subbands at $k = 0$. For photon energy below $E_{21}$ (see figure 2(a)), the optical transitions $(e_1, +1/2) \rightarrow (e_2, +1/2)$ occur at negative values of $k_x$, leading to a flow of spin-up electrons in the subband $e_1$ in the $x'$-direction. With increasing light frequency, the point of optical transitions $k_x = k_{+1/2}$ at which the energy and quasi-momentum conservation laws are met moves toward positive values of $k_x$ (see figure 2(b)). This results in an inversion of the spin current.

The explicit spectral dependence of the spin photocurrent in an ideal QW drastically depends on the fine structure of the energy spectrum. In real QW structures, the spectral width of the intersubband resonance is substantially broadened. Allowance for the broadening can be made assuming, e.g., that the energy separation $E_{21}$ between the subbands varies in the QW plane [30, 31]. Then, to the first order in the spin–orbit coupling, the spin current contributions in the subbands are given by

$$J_{\beta}^{(e_1)} = \frac{\tau_{e_1} e_{\beta}}{2\hbar} \left( y_{e_\beta}^{(e_2)} - y_{e_\beta}^{(e_1)} \right) \frac{d\eta_{21}(\hbar \omega)}{d\hbar \omega} \frac{I}{\hbar \omega}$$

$$J_{\beta}^{(e_2)} = \frac{\tau_{e_2} e_{\beta}}{2\hbar} \left( y_{e_\beta}^{(e_2)} - y_{e_\beta}^{(e_1)} \right) \times \left[ \eta_{21}(\hbar \omega) - \frac{\hbar \omega}{d\eta_{21}(\hbar \omega)/d\hbar \omega} \right] \frac{I}{\hbar \omega},$$

where $\tau_{e_1}$ and $\tau_{e_2}$ are the spin current relaxation times in the subbands $e_1$ and $e_2$, respectively, $y_{e_\beta}^{(e_1)}$ and $y_{e_\beta}^{(e_2)}$ are the constants of $k$-linear spin–orbit coupling in the subbands, see equation (14), $\eta_{21}(\hbar \omega)$ is the intersubband absorbance for radiation polarized along the QW normal with the inhomogeneous broadening being taken into account and $\bar{E}$ is the mean value of the electron kinetic energy. The energy $\bar{E}$ is equal to $E_F/2$ for a two-dimensional degenerate gas with the Fermi energy $E_F$ and $k_B T$ for a non-degenerate gas at the temperature $T$.

The spin photocurrents (16) and (17) are contributed by spin-conserving optical transitions and, therefore, are proportional to the difference of subband splitting constants. The spectral behavior of the pure spin currents in both subbands repeats the derivative of the light absorption spectrum $d\eta_{21}(\hbar \omega)/d\hbar \omega$ provided the intersubband absorption line is narrow enough. Close to the absorption maximum the spin photocurrents reverse their directions with varying light frequency. We also note that the contribution $J_{\beta}^{(e_1)}$ can considerably exceed $J_{\beta}^{(e_2)}$ since the relaxation time in the excited subband $\tau_{e_2}$ may be quite short even at low temperatures due to the effective channel of relaxation by emission of an optical photon.

A contribution to the pure spin currents may also come from linear-in-$k$ spin-dependent terms in the matrix elements of optical transitions. While in the one-band approximation the intersubband absorption can only be induced by the $e_\perp$ component of the polarization vector, in a multi-band model optical transitions between the electron subbands $e_1$ and $e_2$ are allowed for any polarization [32]. Moreover, $k \cdot p$ admixture of the valence-band and remote conduction-band states to the electron wavefunctions adds both spin-dependent [33] and $k$-linear terms to the matrix elements of the optical transitions.
Taking into account these contributions, the $2 \times 2$ spin matrix $M_{21}$ describing the intersubband transitions assumes the form
\[
M_{21} = M_{21}^{(0)} \times \left( e_\perp + i \sum_{a \beta} \lambda_{a \beta}^\perp \sigma_a e_\beta + i \sum_{a \beta} \lambda_{a \beta}^\perp k_a e_\beta + \sum_{a \beta} \lambda_{a \beta}^\perp k_a e_\beta \right),
\]
where $M_{21}^{(0)}$ is the matrix element calculated in the one-band approximation for radiation polarized along the QW normal. The tensor $\lambda$ is responsible for the intersubband optical orientation of electron spins [33], while $\lambda' \lambda''$ describe the optical alignment of electron momenta. Taking into account $k$-linear terms in equation (18), we derive the contributions to pure spin currents excited in the $e_1$ and $e_2$ subbands as
\[
J_{\beta}^{(e_1)} = \left( \sum_{y} \lambda_{y \beta}^\perp k_y e_\gamma + \sum_{y} \lambda_{y \beta}^{\perp} e_\gamma \right) \tau_{\alpha} \frac{\tilde{E} I_{\alpha e} (\hbar \omega)}{\hbar \omega},
\]
\[
J_{\beta}^{(e_2)} = - (\tau_{\alpha} / \tau_{e}) J_{\beta}^{(e_1)}.
\]
In contrast to equation (16), the spectral response of the contribution (19) repeats the light absorption spectrum.

5. Free-carrier absorption in n-doped QWs

The light absorption by free carriers, or Drude-like absorption, occurs in doped semiconductor structures when the photon energy $\hbar \omega$ is smaller than the band gap as well as the energy spacing between the subbands. Such an intersubband excitation of carriers with linearly polarized light also gives rise to a pure spin current. However, in contrast to the direct transitions considered in sections 3 and 4, the subband splitting leads to no essential contribution to the spin current induced by intersubband optical excitation. The more important contribution comes from the spin-dependent asymmetry of electron scattering [13, 34]. Indeed, the free-carrier absorption is always accompanied by electron scattering from acoustic or optical phonons, static defects, etc., because of the need for energy and momentum conservation. In systems with a spin–orbit interaction, processes involving change of the particle wave vector are spin dependent. In particular, in the QW structures the matrix element of electron scattering $V_{k' k}$ contains, in addition to the main contribution $V_0$, asymmetric spin-dependent terms [33, 35]
\[
V_{k' k} = V_0 + \sum_{a \beta} V_{a \beta} \sigma_a (k_x k_x'),
\]
where $k$ and $k'$ are the initial and scattered in-plane wave vectors, respectively. This leads in turn to $k$-linear spin-dependent contribution to the scattering rate, which is determined by the matrix element squared. Microscopically, such terms in the scattering rate originate from structure and/or bulk inversion asymmetries similar to $k$-linear Rashba and Dresselhaus spin splitting of the electron subbands.

Due to the spin-dependent asymmetry of scattering, electrons photoexcited from the subband bottom are scattered in preferred directions depending on their spin states. This is illustrated in figure 3(a), where the free-carrier absorption is shown as a combined two-stage process involving the electron–photon interaction (solid vertical lines) and the electron scattering (dashed horizontal lines). The scattering asymmetry is shown by dashed lines of different thicknesses: electrons with the spin $+1/2$ are preferably scattered into the states with $k_x' > 0$, while electrons with the spin $-1/2$ are predominantly scattered into the states with $k_x' < 0$. Obviously, such an asymmetry of photoexcitation in the $k$ space leads to a pure spin current, where the spin-up and spin-down electrons counterflow and the charge current vanishes.

In the perturbation theory approach, the indirect optical transitions are treated as second-order virtual processes involving intermediate states. To the first order in spin–orbit interaction, the compound matrix element of the intersubband transitions accompanied by the electron scattering from short-range potentials has the form [36]
\[
M_{k k} = \frac{e A}{c m c} e \cdot (k' - k) V_{k k} - 2 \frac{e A}{c h} \sum_{a \beta} V_{a \beta} \sigma_a e_\beta,
\]
where $e$ is the electron charge, $A$ is the vector potential of the electromagnetic wave and $c$ is the light velocity. The first term on the right-hand side of equation (21) describes transitions $(e_1, k) \rightarrow (e_1, k')$ with intermediate states in the conduction subband $e_1$, the second term corresponds to the transitions via intermediate states in other bands. We assume that the electron scattering is elastic and consider the geometry of normal incidence of the light so that the polarization vector $e$ lies in the $(xy)$-plane. Then, the polarization dependences of spin current components are given by
\[
J_{i}^{\mu} = \tau_e \frac{\langle V_0 V_{\alpha' \gamma} e_\gamma^2 \rangle}{\langle V_{\alpha' \gamma} \rangle} e_i^2 + \frac{\langle V_0 V_{\alpha' \gamma} e_\gamma \rangle}{\langle V_{\alpha' \gamma} \rangle} e_i e_\gamma.
\]
Here, the angle brackets $(\cdot \cdot \cdot)$ stand for averaging over the spatial distribution of scatterers and $\eta_{e 1}$ for the radiation absorbance in this spectral range. The components $J_{i}^{\mu}$ can be obtained from equation (22) by the replacement $x \leftrightarrow y$.

Equation (22) shows that pure spin currents can be injected in QWs by the elastic-scattering-assisted photoexcitation with linearly polarized light but vanish for the normally-incidence unpolarized radiation, when $e_i^2 = e_\gamma^2 = 1/2$, $e_i e_\gamma = 0$. The
nonzero components of the spin current are determined by
the explicit form of the matrix element of scattering and the light
polarization plane. In QWs grown on (1 1 0)-oriented substrates, the scattering rate contains the term proportional
to \( \langle V_\alpha V_{\alpha'} \rangle \sigma_\alpha (k_{\alpha'} + k_{\alpha}) \) giving rise to the components \( J^{\alpha}_{\perp} \propto (e_{\alpha} - e_{\alpha'}) \), \( J^{\alpha}_{\parallel} \propto e_{\alpha} e_{\alpha'} \), which are in accordance with the phenomenological equation (5). In (0 0 1)-grown structures, the nonzero coefficients are \( \langle V_\alpha V_{\alpha'} \rangle = -\langle V_\alpha V_{\alpha'} \rangle \) and \( \langle V_{\alpha'} V_{\alpha} \rangle = -\langle V_{\alpha'} V_{\alpha} \rangle \), and the normally-incident radiation can excite fluxes of the in-plane spin components only.

The pure spin current caused by the free-carrier absorption can be converted into an electric current by polarizing electron spins, e.g., by the application of an external magnetic field, as was shown by Ganichev et al. [34], see also [8]. Indeed, in the case of intrasubband absorption, the fluxes of the spin-up and spin-down carriers, \( i_{1/2} \) and \( i_{-1/2} \), are proportional to the electron densities in the spin subbands, \( n_{1/2} \) and \( n_{-1/2} \), respectively. In a spin-polarized system, where \( n_{1/2} \neq n_{-1/2} \), the fluxes \( i_{1/2} \) and \( i_{-1/2} \) no longer compensate each other yielding a net electric current

\[
j_{\beta} = 4e \sum_{\alpha} S_{\alpha} J^{\alpha}_{\beta},
\]

where \( S = \) the average electron spin with \( |S| = (1/2)(n_{1/2} - n_{-1/2})/(n_{1/2} + n_{-1/2}) \).

6. Pure spin currents caused by electron heating

In addition to the free-carrier absorption, the spin-dependent asymmetry of the electron scattering by phonons gives rise to a pure spin current if the electron gas is simply driven out of thermal equilibrium with the crystal lattice (see [8, 34, 37]). In such a relaxational mechanism, the spin current is generated in the process of energy relaxation of electrons no matter how the thermal equilibrium between the electron and phonon subsystems was initially disturbed.

The relaxational mechanism of the spin current generation is illustrated in figure 3(b), where the processes of energy relaxation of hot electrons by emitting phonons are shown by dashed curves. Due to the spin-dependent asymmetry of the electron–phonon interaction, electrons with the spin \( +1/2 \) relax faster from the high-energy states with positive \( k_{\perp} \), while electrons with the spin \( -1/2 \) predominantly vacate the high-energy states with negative \( k_{\perp} \). This leads to an asymmetrical distribution, where the spin-up carriers mainly occupy the left-hand branch of the dispersion curve (carriers with the opposite spin orientation have gone to the subband bottom), while the spin-down carriers mainly occupy the right-hand branch. Such a spin-dependent imbalance of electrons between the positive and negative \( k_{\perp} \) yields a pure spin current.

We consider the energy relaxation of electrons confined in a QW by bulk acoustic phonons. Taking into account \( k \)-linear contributions to the electron–phonon interaction, the matrix element of the electron scattering by phonons can be modeled by

\[
V_{\alpha k}(q) = V_\alpha(q_{\perp}) + \sum_{\alpha \beta} V_{\alpha \beta}(q_{\perp}) \sigma_\alpha (k_{\beta} + k_{\alpha}^\perp),
\]

where \( V_\alpha(q_{\perp}) \) and \( V_{\alpha \beta}(q_{\perp}) \) are functions of \( q_{\perp} \), their forms dependent on the QW design, and \( q = \pm(k - k', q_{\perp}) \) is the three-dimensional wave vector of the phonon involved. We assume that both electrons and phonons obey the Boltzmann statistics, but the electron temperature \( T_e \) differs from the lattice temperature \( T_0 \). Then, the rates of phonon emission and absorption become nonequal leading to a spin current

\[
j^e_{\perp} = \frac{n_e}{\tau_e} \frac{\hbar c^2}{2} \frac{T_e - T_0}{T_e} \sum_{\alpha \beta} V_{\alpha \beta} \Re [V_\alpha(q_{\perp}) V_{\alpha \beta}(q_{\perp})] q_{\perp} |dq_{\perp}| \int_{-\infty}^{\infty} |V_\alpha(q_{\perp})|^2/|q_{\perp}| dq_{\perp}^2,
\]

(25)

where \( N_e \) is the carrier density, \( \tau_\text{ph} \) is the momentum relaxation time governed by the electron–phonon interaction and \( c_s \) is the sound velocity in the crystal.

As a more detailed example, we consider the (1 1 0)-grown QWs. In this case, the dominant spin-dependent contribution to the Hamiltonian of the electron–phonon interaction in the deformation-potential model is proportional to \( \sigma_\alpha (k_{\perp} + k_{\alpha}^\perp) \), and the corresponding Hamiltonian has the form [38]

\[
H_{\text{el-phon}}(k', k) = \Xi \sum_a u_{aa} + \xi \Xi_{\text{cv}} u_{e \text{cv}} \sigma_\alpha (k_{\perp} + k_{\alpha}^\perp)/2.
\]

Here, \( \Xi \), and \( \Xi_{\text{cv}} \) are the intraband and interband constants of the deformation potential, \( u_{aa} \) are the phonon-induced strain tensor components, \( \xi = F \Delta_{\text{ao}}/(3 E_{\text{f}} [E_\perp + \Delta_{\text{ao}}]) \), and \( \Delta_{\text{ao}} \) is the spin–orbit splitting of the valence band. The interband constant \( \Xi_{\text{cv}} \) originates from the lack of an inversion center in zinc-blende-type crystals and vanishes in centrosymmetric semiconductors [6]. Assuming that electrons are confined in a rectangular quantum well of width \( a \), we derive for the spin current

\[
j^e_{\perp} = -\frac{\pi^2 \xi}{3a^2} \frac{\tau_e}{\tau_\text{ph} k_{\beta} T_0} \Xi_{\text{cv}} \frac{T_e - T_0}{T_e} N_e.
\]

(27)

Equation (27) shows that the spin current component \( j^e_{\perp} \) strongly depends on the QW width.

7. Pure valley-orbit currents

In addition to the spin, free carriers in solid states can be characterized by other internal properties, e.g., by a well number in multiple QW structures or a valley index \( \nu \) in many-valley semiconductors. In the latter case, one can consider pure orbit-valley currents, where partial electron fluxes in valleys \( i \), are nonzero but the net electric current \( e \sum_i i \) vanishes [13]. Here, the role of spin-up and spin-down states is replaced by the valley index: there is no net charge current, but the electrons in different valleys travel in different directions.

To elaborate the concept of pure orbit-valley currents, we consider silicon-based quantum wells grown on a (1 1 1)-oriented surface. In Si QWs, the conduction-band subbands are formed by six equivalent valleys \( X, X', Y, Y', Z, Z' \) located at the \( \Delta \) points of the Brillouin zone of the bulk crystal. All the valleys retain their equivalence in (1 1 1)-grown structures because the angles between the growth direction and the valley principal axes are the same. Figure 4 sketches the valley positions and orientations in the two-dimensional \( k \).
space in the QW plane. In asymmetrical (1 1 1)-grown QWs, each valley has the \(C_3\) point-group symmetry allowing for the generation of a partial in-plane flux \(\mathbf{i}_j\) at normal incident light. Under excitation with unpolarized light, the fluxes \(\mathbf{i}_j\) are directed along the in-plane projections of the valley principal axes (see figure 4(a)). Since the structure is invariant with respect to the rotation by 120° along the growth direction, the total charge current \(\sum_{\nu} \mathbf{i}_j\) vanishes. Thus, such an electron distribution can be referred to as an optically injected pure valley-orbit current.

In addition to the polarization-independent photocurrent, the excitation of a Si (1 1 1)-grown QW with circularly polarized light at normal incidence results, in each valley, in a flux component \(\mathbf{i}_j^c\) which reverses its direction upon switching the light polarization from right-handed to left-handed circular polarization. Such helicity-dependent components \(\mathbf{i}_j^c\) flow perpendicularly to the valley principal axes (see figure 4(b)) and also contribute to the pure valley-orbit current. We note that the absence of a total photocurrent under illumination with unpolarized or circularly polarized light is related to the overall \(C_{3v}\) symmetry of the QWs which, however, allows for a net electric current induced by linearly polarized light. In this particular case, the partial fluxes in valleys become nonequal and do not compensate each other.

Pure valley-orbit currents can also be optically injected in bulk multi-valley noncentrosymmetrical crystals such as AlAs, AISb, GaP, etc. In these compounds, the conduction-band minima are located at the \(X\) points at the Brillouin-zone edge. Each of three equivalent valleys \(v = X, Y, Z\) has the \(D_{3d}\) symmetry allowing for the helicity-dependent electron flux \(\mathbf{i}_j^c\):

\[
\begin{align*}
\mathbf{i}_x &= \mathcal{P}i(0, -\mathbf{e}_y, \mathbf{e}_z), \\
\mathbf{i}_y &= \mathcal{P}i(-\mathbf{e}_x, 0, \mathbf{e}_z), \\
\mathbf{i}_z &= \mathcal{P}i(\mathbf{e}_x, -\mathbf{e}_y, 0),
\end{align*}
\] (28)

where \(\mathbf{e}_\alpha (\alpha = x, y, z)\) are components of the vector \(i[(\mathbf{e} \times \mathbf{e}^\dagger)] = P_{\text{circ}}q/q, \mathbf{q}\) is the light wave vector and \(P_{\text{circ}}\) is the light helicity ranging from \(-1\) to \(+1\). In accordance with the overall \(T_{3d}\) point-group symmetry of the zinc-blende-type crystals, the total current vanishes for homogeneous illumination with circularly polarized light. In an external magnetic field, each contribution \(\mathbf{i}_j\) varies due to the Lorentz force acting upon electrons. This action is, however, different for different valleys due to the energy spectrum anisotropy in valleys. As a result, the magnetic field causes an imbalance of the valley-orbit current giving rise to a nonzero net electric current

\[
\mathbf{j} \propto (\mathbf{e}_x B_y + \mathbf{e}_y B_x, \mathbf{e}_y B_z + \mathbf{e}_z B_y + \mathbf{e}_z B_x).
\]

For the particular geometry \(\mathbf{q}[[1 1 \overline{1}]\) and \(\mathbf{B}[0 1 0]\), the magnetic-field-induced photocurrent \(\mathbf{j}\) appears in the \([1 0 1]\)-direction.

### 8. Conclusion

We have shown that pure spin currents of free carriers can readily be created in semiconductor structures by optical excitation with linearly polarized or even unpolarized light. The pure spin currents lead to spatial separation of the spin-up and spin-down particles and accumulation of the opposite spins at the opposite edges of the sample. We have presented the microscopic theory of pure spin photocurrents for all main types of optical transitions ranging from the fundamental interband to the free-carrier absorption. In the present paper, we have focused on the spin photocurrents contributed by charge carriers. In addition, spin fluxes (or, in general, angular-momentum fluxes) can also be formed by neutral particles or excitations lacking electric charge such as photons [39, 40], excitons or exciton polaritons [41, 42], and even phonons and magnons. The study of spin currents is naturally inscribed in the physics of spin-related phenomena and opens up new opportunities for the realization of novel device concepts.

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