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Orbital mechanism of the circular photogalvanic effect in quantum wells

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It is shown that the free-carrier (Drude) absorption of circularly polarized radiation in quantum well structures leads to an electric current flow. The photocurrent reverses its direction upon switching the light helicity. A pure orbital mechanism of such a circular photogalvanic effect is proposed that is based on interference of different pathways contributing to the light absorption. Calculation shows that the magnitude of the helicity dependent photocurrent in n-doped quantum well structures corresponds to recent experimental observations.

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1. Introduction. The absorption of circularly polarized light in semiconductor structures may lead to generation of an electric current which reverses its direction upon switching the light polarization from right-handed to left-handed and vice versa. Such a circular photogalvanic effect (CPGE) caused by asymmetry of elementary processes of photoexcitation requires structures of the appropriate symmetry. It is possible only in media which belong to one of the gyrotropic classes [1, 2].

Recently, CPGE has attracted a great deal of attention. It was observed in low-dimensional structures: in zinc-blende- and diamond-type quantum wells (QWs), which are also belong to the gyrotropic media [3]. Circular photocurrents were studied for a variety of optical ranges, including interband transitions in QWs [4 – 6], direct intersubband [7] and indirect intrasubband (Drude-like) [8] transitions in n-doped QW structures.

So far, the microscopic theory of CPGE in zinc-blende-type structures has been developed for the direct interband or intersubband optical transitions. It has been shown that the effect originates from the complicated band structure of the semiconductor compounds, mostly, from spin-orbit splitting of the electron or hole states, which is odd in the wave vector $\mathbf{k}$ [9, 10, 7], and linear-in-$\mathbf{k}$ terms in the matrix elements of interband optical transitions [9, 11]. Here we address CPGE caused by the free-carrier absorption of circularly polarized radiation in n-doped QW structures. We show that a pure orbital mechanism of the effect can predominate in this spectral range, which is related to neither spin-orbit coupling nor spin-sensitive selection rules for the optical transitions. This mechanism is based on interference of different pathways contributing to the light absorption. A similar CPGE model for the optical transitions between quantum subbands was proposed in Ref. [12]. However, the orbital mechanism is especially efficient for the free-carrier absorption, which is contributed by indirect optical transitions.

We consider CPGE in (001)-oriented QWs grown from zinc-blende-type semiconductors. Symmetry analysis shows that the helicity dependent photocurrent $\mathbf{j}$ in such structures can be induced only at oblique incidence of the radiation [2, 3] and is described by two linearly independent constants γ_1 and γ_2 as follows:

$$\begin{aligned} j_x &= [\gamma_2\, l_x - \gamma_1\, l_y]\, I P_{\rm cicr}, \\ j_y &= [\gamma_1\, l_x - \gamma_2\, l_y]\, I P_{\rm cicr}, \end{aligned} \qquad (1)$$

where $\mathbf{l} = \mathbf{q}/q$ is the unit vector pointing in the light propagation direction, $\mathbf{q}$ and I are the wave vector and intensity of the light inside the structure, respectively; $P_{\rm circ}$ is the radiation helicity ranging from -1 (for the left-handed circular polarization) to $+1$ (for the right-handed circular polarization); $x \parallel [100]$, $y \parallel [010]$, and $z \parallel [001]$ are the cubic crystallographic axes. Phenomenologically, the constant γ_1 is related to the heterostructure asymmetry, i.e., nonequivalence of the z and $-z$ directions, while the constant γ_2 originates from lack of an inversion center in the host crystal.

At normal incidence of the light the helicity dependent photocurrent can be induced only in quantum wells grown along one of the low-symmetric crystallographic directions, such as (110)- or (113)-oriented structures. In this geometry CPGE is described by

$$j_{x'} = \gamma_3\, l_{z'} I P_{\rm cicr}, \qquad (2)$$

where x' is an in-plane axis, which is pointed along $[1\bar{1}0]$ for (110)- and (113)-oriented QWs, and z' is the growth direction. The phenomenological constant γ_3 is entirely

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related to absence of an inversion center in the host crystal, similarly to γ_2 in Eq. (1).

Below we present a microscopic theory for the orbital mechanism of CPGE and show that this mechanism contributes to γ_1 as well as γ_2 and γ_3 constants.

2. Microscopic theory. The free-carrier absorption of radiation is always accompanied by electron scattering from static defects, acoustic or optical phonons, etc., because of the need for energy and momentum conservation. Such indirect optical transitions are treated as second-order processes, which involve electron-photon interaction and electron scattering, via virtual intermediate states. The intermediate states can be those within the same quantum subband, $e1$ in our case, or in other conduction or valence subbands. The dominant pathway determining the QW absorbance involve intermediate states within the subband $e1$ (see Fig. 1). The matrix element of such kind of processes has the form [2]

$$M_{\mathbf{k}'\mathbf{k}}^{(e1)} = \frac{eA}{c\omega m^*}\,\mathbf{e}\cdot(\mathbf{k}'-\mathbf{k})\,V_{11}. \tag{3}$$

Here $\mathbf{k}$ and $\mathbf{k}'$ are the initial and final electron wave vectors, e is the electron charge, A is the amplitude of the electromagnetic wave related to the light intensity by $I = A^2\omega^2 n_\omega/2\pi c$, ω is the light frequency, m^* is the effective electron mass, n_ω is the refractive index of the medium, $\mathbf{e}$ is the (complex) unit vector of the light polarization, and V_{11} is the matrix element of intrasubband scattering. As follows from Eq. (3), optical transitions with intermediate states in the subband $e1$ can be induced only by radiation with nonzero in-plane component of the polarization vector $\mathbf{e}$. They do not lead to any circular photocurrent because the square of the matrix element (3), which determines the transition rate, is independent of the light helicity.

The helicity dependent photocurrent arises if one takes into account interference of the processes depicted in Fig. 1 and those with virtual intermediate states in other subbands.

2.1. *Contribution to* γ_1. The quantum subband, which is the closest to $e1$, is the electron subband $e2$. Optical transitions with intermediate states in the subband $e2$ contributing to the free-carrier absorption are sketched in Fig.2. The matrix element of these indirect processes has the form

$$M_{\mathbf{k}'\mathbf{k}}^{(e2)} = i\frac{eA}{c\hbar}\left(\frac{\varepsilon_{21}}{\varepsilon_{21}-\hbar\omega} - \frac{\varepsilon_{21}}{\varepsilon_{21}+\hbar\omega}\right) z_{21}\, e_z V_{21}. \tag{4}$$

Here ε_{21} is the energy separation between the subbands $e2$ and $e1$, z_{21} is the coordinate matrix element between the envelope functions in the subbands, $\varphi_1(z)$ and $\varphi_2(z)$,

$$z_{21} = \int_{-\infty}^{+\infty} \varphi_2(z)z\,\varphi_1(z)\,dz, \tag{5}$$

and V_{21} is the matrix element of *inter*-subband scattering. In contrast to Eq. (3), the matrix element (4) is nonzero only for radiation with nonvanishing out-of-plane component of the polarization vector $\mathbf{e}$ and contains the imaginary unit as a prefactor.

Light absorption by free carriers is contributed by processes with all possible intermediate states. Making allowance for the transitions via the subbands $e1$ and $e2$ one can write for the photocurrent

$$\mathbf{j} = e\frac{4\pi}{\hbar}\sum_{\mathbf{k},\mathbf{k}'}[\tau_p(\varepsilon_{\mathbf{k}'})\,\mathbf{v}_{\mathbf{k}'} - \tau_p(\varepsilon_{\mathbf{k}})\,\mathbf{v}_{\mathbf{k}}]\times$$
$$\times\left|M_{\mathbf{k}'\mathbf{k}}^{(e1)} + M_{\mathbf{k}'\mathbf{k}}^{(e2)}\right|^2 (f_{\mathbf{k}} - f_{\mathbf{k}'})\,\delta(\varepsilon_{\mathbf{k}'} - \varepsilon_{\mathbf{k}} - \hbar\omega), \tag{6}$$

where $\mathbf{v}_{\mathbf{k}} = \hbar\mathbf{k}/m^*$ and $\varepsilon_{\mathbf{k}} = \hbar^2k^2/(2m^*)$ are the electron velocity and kinetic energy, respectively, $\tau_p(\varepsilon_{\mathbf{k}})$ is the momentum relaxation time, which may depend on the electron energy, $f_{\mathbf{k}}$ is the function of equilibrium carrier distribution in the subband $e1$, and the factor 4 in Eq. (6) accounts for the spin degeneracy.

The square of the matrix element sum in Eq. (6) contains the interference term $2\mathrm{Re}[M_{\mathbf{k}'\mathbf{k}}^{(e1)} M_{\mathbf{k}'\mathbf{k}}^{(e2)*}]$, which is odd in the wave vector. Therefore, it leads to an asymmetrical distribution of the photoexcited carriers in $\mathbf{k}$-space, i.e., to an electric current. Moreover, the interference term is proportional to components of $i[\mathbf{e}\times\mathbf{e}^*]$. It vanishes for the linearly polarized light and is proportional to the light helicity P_{circ} for the elliptical or circular polarization, because $i[\mathbf{e}\times\mathbf{e}^*] = \mathbf{l}P_{\mathrm{circ}}$. Thus, the sign of the interference term is determined by the light helicity, and the photocurrent reverses its direction by changing the light polarization from right-handed to left-handed and vice versa.

Physically, the orbital mechanism of the circular photogalvanic effect can also be interpreted as follows. Under excitation with circularly polarized light the processes depicted in Fig.1 and Fig.2 are added constructively for transitions to positive k_x' (or negative k_x', depending on the light helicity) and destructively for transitions to $-k_x'$. It means that the transition rates to the states k_x' and $-k_x'$ are different, resulting in an electric current of the photoexcited carriers. Direction of the photocurrent with respect to the crystallographic axes is determined by the light polarization and the ex-

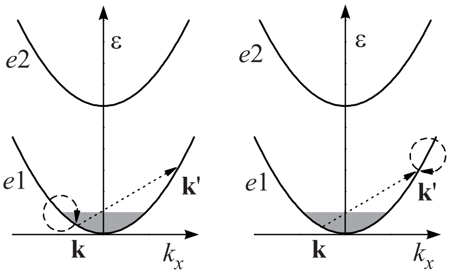

Fig.1. Intrasubband optical transitions $(e1,\mathbf{k}) \to (e1,\mathbf{k}')$ with intermediate states in the subband $e1$. Dashed circles and dotted lines represent electron-photon interaction and electron scattering, respectively. Panels correspond to two possible processes: electron-photon interaction followed by electron scattering (left panel) and electron scattering followed by electron-photon interaction (right panel)

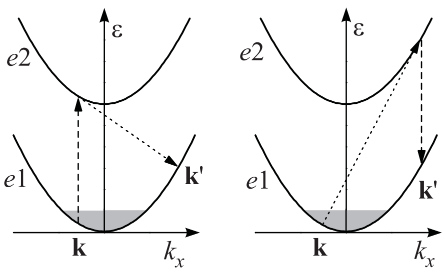

Fig.2. Intrasubband optical transitions $(e1,\mathbf{k}) \to (e1,\mathbf{k}')$ with intermediate states in the subband $e2$. Dashed and dotted lines represent electron-photon interaction and electron scattering, respectively. Panels correspond to two possible processes: electron-photon interaction followed by electron scattering (left panel) and electron scattering followed by electron-photon interaction (right panel)

plicit form of the matrix elements of intrasubband and intersubband scattering.

The matrix element (4) is proportional to out-of-plane component of the polarization vector. Therefore, the term caused by interference of the optical processes via the subband $e1$ and $e2$ can contribute to CPGE at oblique incidence only. We consider the effect for (001)-grown quantum wells. In such structures electron scattering by static defects, acoustic or optical phonons is usually treated as central. Under this assumption the matrix elements of intrasubband and intersubband scattering, V_{11} and V_{21}, respectively, take no account of the lattice symmetry of the host semiconductor and depend on $|\mathbf{k}'-\mathbf{k}|$ only. Then, the helicity dependent photocurrent flows in the direction perpendicular to the light incidence plane. This contribution corresponds to the terms described by the phenomenological constant γ_1 in Eq. (1). Calculation after Eq. (6) shows that, in the case of elastic scattering and provided $\hbar\omega \ll \varepsilon_{21}$, the constant is given by

$$\gamma_1 = -2e\tau_p \frac{\omega z_{21}}{\varepsilon_{21}}\, \xi \eta_{\parallel}, \tag{7}$$

where ξ is a dimensionless parameter, which depends on the structure design and mechanisms of scattering, and $\eta_{\parallel}$ is the QW absorbance for radiation polarized in the QW plane. We note that both the absorbance $\eta_{\parallel}$ and the momentum relaxation time τ_p are governed by the matrix element of intrasubband scattering V_{11}. Thus, the product $\tau_p \eta_{\parallel}$ is independent of the scattering rate. For quasi-elastic scattering from acoustic phonons or short-range static defects, the product is given by

$$\tau_p\, \eta_{\parallel} = \frac{2\pi\alpha}{n_\omega} \frac{\kappa\hbar}{m^*\omega^2} N_e, \tag{8}$$

where $\alpha = e^2/\hbar c \approx 1/137$ is the fine-structure constant, N_e is the carrier density, and κ is a parameter depending on the carrier distribution. The latter is equal to 1 and 2 for the cases $\hbar\omega \gg \bar{\varepsilon}$ and $\hbar\omega \ll \bar{\varepsilon}$, respectively, with $\bar{\varepsilon}$ being the mean kinetic energy of equilibrium carriers ($\bar{\varepsilon} = E_F/2$ for a degenerate two-dimensional gas and $\bar{\varepsilon} = k_B T$ for a non-degenerate two-dimensional gas, where E_F is the Fermi energy, k_B is the Boltzmann constant, and T is the temperature).

At low temperatures the free-carrier absorption is dominated by processes, which involve elastic scattering from static defects such as impurities, imperfections of the QW interfaces, etc. For electron scattering from short-range defects the parameter ξ has the form

$$\xi = \int\limits_{-\infty}^{+\infty} \varphi_1^3(z)\varphi_2(z)w(z)dz \Bigg/ \int\limits_{-\infty}^{+\infty} \varphi_1^4(z)w(z)dz, \tag{9}$$

where $w(z)$ is the distribution function of the scatterers along the growth direction. At higher temperatures electron-phonon interaction predominates over the electron scattering by static defects. In the case of quasi-elastic scattering from acoustic phonons the parameter ξ is also given by Eq. (9) where $w(z)$ is set to be a constant.

In accordance with general symmetry arguments, the helicity dependent photocurrent corresponding to the phenomenological constant γ_1 is related to inversion asymmetry of the heterostructure. This follows also from Eqs. (7) and (9), which demonstrate that the sign and magnitude of γ_1 is determined by asymmetry of the confinement potential and the doping profile. In particular, $\gamma_1 \equiv 0$ for the absolutely symmetrical structures, where $w(z)$ and $\varphi_1(z)$ are even functions while $\varphi_2(z)$ is an odd function with respect to the QW center.

2.2. *Contribution to γ_2 and γ_3.* To obtain the circular photocurrents described by the phenomenological constants γ_2 in Eq. (1) and γ_3 in Eq. (2) one has to take into account the lattice symmetry of the QW host semiconductor. This can be done considering interference of optical transitions with intermediate states in the subband $e1$ and those via the valence-band states.

To calculate this contribution to CPGE we neglect spin-orbit splitting of the valence band for simplicity and assume that the effective hole masses in the QW plane are larger than the effective electron mass. Then, the matrix element of the intrasubband optical transitions via the valence-band states can be presented as

$$M^{(v)}_{\mathbf{k}'\mathbf{k}} = i\frac{eA}{c\hbar}\frac{\hbar\omega}{E_g^2}P\sum_j e_j V_{SR_j}. \tag{10}$$

Here E_g is the band gap energy, $P = i(\hbar/m_0)\langle S|p_z|R_z\rangle$ is the Kane matrix element, m_0 is the free electron mass, S and R_j $(j = x, y, z)$ are the Bloch functions of the conduction and valence bands at Γ-point of the Brillouin zone, respectively, and V_{SR_j} are the matrix elements of *inter*-band scattering. For scattering from acoustic phonons the matrix elements V_{SR_j} have the form [13, 14]

$$\begin{aligned} V_{SR_x} &= \Xi_{cv}\langle\mathbf{k}'|u_{yz}|\mathbf{k}\rangle, \\ V_{SR_y} &= \Xi_{cv}\langle\mathbf{k}'|u_{xz}|\mathbf{k}\rangle, \\ V_{SR_z} &= \Xi_{cv}\langle\mathbf{k}'|u_{xy}|\mathbf{k}\rangle, \end{aligned} \tag{11}$$

where Ξ_{cv} is the interband deformation-potential constant, $\langle\mathbf{k}'|u_{\alpha\beta}|\mathbf{k}\rangle$ are the matrix elements of coupling between the electron states $\mathbf{k}$ and $\mathbf{k}'$ in the subband $e1$ caused by the deformation, and $u_{\alpha\beta}$ $(\alpha \neq \beta)$ are the phonon-induced off-diagonal components of the strain tensor. The interband constant Ξ_{cv} originates from lack of an inversion symmetry in zinc-blende-type crystals (T_d point group) and vanishes in centrosymmetric semiconductors [13].

Calculation shows that in (001)-oriented QWs the matrix element $M^{(v)}_{\mathbf{k}'\mathbf{k}}$ contains a term proportional to $(k_x' - k_x)(k_y' - k_y)e_z$. Interference of this term and $M^{(e1)}_{\mathbf{k}'\mathbf{k}}$ gives rise to the circular photocurrent described by γ_2 in Eq. (1). For the free-carrier absorption assisted by quasi-elastic scattering from acoustic phonons the constant γ_2 has the form

$$\gamma_2 = -2e\tau_p\frac{\Xi_{cv}}{\Xi_c}\frac{\omega P}{E_g^2}\zeta\eta_\parallel, \tag{12}$$

where Ξ_c is the intraband deformation-potential constant, which determines the light absorbance $\eta_\parallel$, and ζ is a parameter depending on the QW width, the carrier distribution and the photon energy. For a rectangular quantum well with the infinitely high barriers, $\zeta = 8\pi\bar{k}a/3$ if the photon energy is much less than the mean electron energy and $\zeta = k_\omega a/12$ in the opposite limiting case. Here $\bar{k}$ is the mean value of $|\mathbf{k}|$ ($\bar{k} = 2k_F/3$ for a degenerate two-dimensional gas and $\bar{k} = \sqrt{\pi m^* k_B T/2\hbar^2}$ for a non-degenerate two-dimensional gas, where k_F is the Fermi wave vector), $k_\omega = \sqrt{2m^*\omega/\hbar}$, and a is the QW width.

In (110)-oriented QWs interference of the optical transitions with intermediate states in the subband $e1$ and in the valence band leads to the helicity dependent photocurrent even at normal incidence of the light. Calculation shows that for this particular mechanism the constant γ_3 has the form

$$\gamma_3 = -\gamma_2/4\zeta. \tag{13}$$

It is independent of the QW width and is determined solely by the carrier density, the scattering mechanism and the band parameters.

As is mentioned above, the phenomenological constants γ_2 and γ_3 are related to lack of an inversion symmetry in the bulk semiconductor rather than to the structure asymmetry. This follows also from Eqs. (12) and (13), which show that both γ_2 and γ_3 do not vanish even in symmetrical QWs grown from zinc-blende-type compounds.

Another orbital contribution to γ_2 can originate from interference of the processes via the subbands $e1$ and $e2$ (depicted in Figs.1 and 2, respectively) in the presence of scattering anisotropy. Indeed, the T_d point group of zinc-blende-type semiconductors does not contain the space inversion and allows for an antisymmetric term in the matrix element of scattering, which changes the sign upon the replacement $\mathbf{k} \to -\mathbf{k}$ and $\mathbf{k}' \to -\mathbf{k}'$. Such an anisotropy in scattering can be modeled, e.g., by impurity potential of the form [15]

$$\begin{aligned} U_i(\mathbf{r}) = U_0(|\mathbf{r} - \mathbf{r}_i|) + \\ + U_3(|\mathbf{r} - \mathbf{r}_i|)(x - x_i)(y - y_i)(z - z_i), \end{aligned} \tag{14}$$

where $\mathbf{r}_i$ is the impurity position. The antisymmetric part of the impurity potential does not violate inherent isotropy of electrical properties of bulk cubic materials and, therefore, can not be easily detected [16]. However, in (001)-oriented QWs it adds a term to the matrix element of electron scattering, which is proportional to $(k_x' - k_x)(k_y' - k_y)$ and, therefore, contributes to γ_2. We assume that the impurity range is much shorter than the QW width and the inequality $\hbar\omega \ll \varepsilon_{21}$ is fulfilled.

Then, the contribution to γ_2 caused by the scattering anisotropy has the form

$$\gamma_2' = e\tau_p \frac{u_3}{u_0} \frac{\omega z_{21}}{\varepsilon_{21}} \frac{m^* E}{\hbar^2} Q\eta, \qquad (15)$$

where u_0 and u_3 are related to the symmetric and antisymmetric parts of the impurity potential by

$$u_0 = \int U_0(|\mathbf{r}|)\, d\mathbf{r}\ , \ \ u_3 = \int U_3(|\mathbf{r}|)\, x^2 y^2 z^2 d\mathbf{r}; \qquad (16)$$

E is an energy given by $E = 6\bar{\varepsilon}$ if $\hbar\omega \ll \bar{\varepsilon}$ and $E = \hbar\omega$ if $\hbar\omega \gg \bar{\varepsilon}$; Q is a parameter, which is determined by the QW confinement potential and the doping profile,

$$Q = \int\limits_{-\infty}^{+\infty} \frac{d\, \varphi_1^3(z)\varphi_2(z)}{dz}\, w(z)dz \Bigg/ \int\limits_{-\infty}^{+\infty} \varphi_1^4(z) w(z) dz. \qquad (17)$$

The parameter Q equals to $2\pi/a$ for a rectangular quantum well with infinitely high barriers where the layer of impurities is placed in the QW center.

3. Discussion. We have shown that the free-carrier absorption of circularly polarized radiation in quantum well structures leads to an electric current, which reverses its direction upon switching the light helicity. The proposed pure orbital mechanism of the circular photogalvanic effect is based on interference of different pathways contributing to the light absorption and does not involve spin of free carriers.

In QW structures grown along [110] or [113] crystallographic direction the circular photocurrent can be excited at normal incidence of the radiation. Following Eqs. (2) and (13) we can estimate the magnitude of the circular photocurrent density. It gives $j \sim \sim 2 \cdot 10^{-10}$ A/cm for the photon energy $\hbar\omega = 10$ meV, the light intensity $I = 1\,\mathrm{W/cm^2}$, the carrier density $N_e = 10^{12}\,\mathrm{cm^{-2}}$, and the band parameters $P/\hbar \approx \approx \sqrt{E_g/2m^*} \approx 1.3 \cdot 10^8$ cm/s, $|\Xi_{cv}/\Xi_c| \approx 0.36$ [13], which are appropriate to GaAs-based structures.

In (001)-oriented QWs the helicity dependent photocurrent can be induced at oblique incidence of the radiation only, and the current magnitude varies in a wide range depending on the QW width and asymmetry. Equations (1) and (7) give $j \sim 10^{-8}$ A/cm for a QW of the width $a = 150$ Å, the asymmetry degree $\xi = 0.2$, $l_x = 0.2$, and the photon energy, the light intensity and the carrier density presented above. The estimated magnitude of the photocurrent corresponds to that measured in experiments on (001)-oriented structures [3].

In addition to the orbital mechanism, a contribution to CPGE in quantum well structures may come from **k**-linear spin-orbit splitting of the quantum subbands together with the spin-sensitive selection rules for the optical transitions. However, estimations show that the spin contribution to the circular photocurrent induced by the free-carrier absorption in n-doped structures vanishes to the first order in spin-orbit interaction and, therefore, is considerably smaller than the orbital term.

Additional experiments, such as measurements of the frequency and temperature dependencies of the photocurrent, can be useful in establishing the roles of the orbital and spin-related mechanisms of CPGE more reliable. In particular, the ratio γ_1/γ_2 is expected to be independent of the photon energy $\hbar\omega$ for the spin-related mechanism, while it does depend on $\hbar\omega$ for the orbital mechanism provided $\hbar\omega > \bar{\varepsilon}$. Besides, increase of the temperature from liquid helium to the room temperature leads to changing the dominant scattering mechanism. It can strongly affect the orbital contribution to CPGE, which is inherently related to details of scattering.

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16. Impurity potential of the form (14) can give rise to non-diagonal components of the conductivity tensor, σ_{xy} and σ_{yx}, in asymmetrical (001)-grown QWs.