Monopolar Optical Orientation of Electronic Spins in Semiconductors

S.A. Tarasenko[1], E.L. Ivchenko[1], V.V. Bel'kov[1], S.D. Ganichev[2], D. Schowalter[2], Petra Schneider[2], M. Sollinger[2], W. Prettl[2], V.M. Ustinov[1], A.E. Zhukov[1], and L. E. Vorobjev[3]

[1] A.F. Ioffe Physico-Technical Institute, 194021 St. Petersburg, Russia
[2] Fakultät für Physik, Universität Regensburg, D-93040 Regensburg, Germany
[3] St. Petersburg State Technical University, 195251 St. Petersburg, Russia

Abstract. It is shown that absorption of circularly polarized infrared radiation due to intraband (Drude-like) transitions in n-type bulk semiconductors and due to intra-subband or inter-subband transitions in quantum well (QW) structures results in a monopolar spin orientation of free electrons. Spin polarization in zinc-blende-structure based QWs is demonstrated by the observation of the spin-galvanic and the circular photogalvanic effects. The monopolar spin orientation in n-type materials is shown to be possible if an admixture of valence band states to the conduction band wave function and the spin-orbit splitting of the valence band are taken into account.

1. Introduction

Absorption of circularly polarized light in semiconductors may result in spin polarization of photoexcited carriers. This phenomenon of optical orientation is well known for interband transitions in semiconductors [1]. At interband excitation with circularly polarized light transitions from the valence to the conduction band are allowed only if the angular momentum is changed by ± 1. These selection rules lead to the spin orientation of carriers with the sign and degree of polarization depending on the light helicity. While optical orientation at interband excitation has been widely studied, it is not obvious that the absorption of infrared radiation due to intraband optical transitions can result in a spin polarization.

In this paper we show that free carrier absorption of circularly polarized radiation due to both, indirect optical transitions for bulk semiconductors or quantum well structures, and direct transitions between size-quantized subbands also leads to spin orientation of free electrons. This optical orientation has not been considered previously and may be referred to as 'monopolar spin orientation' because only one type of carriers is excited, electrons in n-type materials, holes in p-type materials. We present theoretical and experimental results on the monopolar optical orientation of intraband absorption by free carriers in n-type zinc-blende-structure bulk semiconductors and low dimensional structures. We show that the monopolar optical orientation of electrons can be obtained if an admixture of the Γ_7 and Γ_8 valence band states to the conduction band wave functions is taken into account.

2. Monopolar optical spin orientation in bulk semiconductors

Absorption of infrared light by free electrons in n-type semiconductors (Drude-like absorption) occurs by indirect intraband transitions where momentum conservation law is satisfied due to acoustic phonons, optical phonons, static defects etc. The intraband absorption

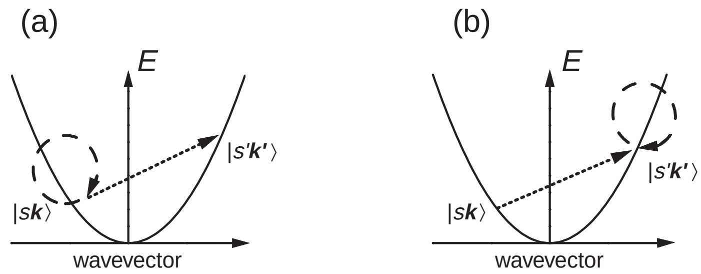

Figure 1. Schematic representation of indirect intraband optical transitions with intermediate states in the same band. Dashed and dotted curves indicate the electron-photon interaction and the electron momentum scattering. Figures (a) and (b) correspond to the first and the second term in Eq. (1).

of the light involving both electron-photon interaction and scattering is described by second-order processes with virtual intermediate states. The compound matrix elements for such kind of transitions with the initial and final states in the same band has the standard form

$$M_{cs'\boldsymbol{k}'\leftarrow cs\boldsymbol{k}} = \sum_{\nu}\left(\frac{V_{cs'\boldsymbol{k}',\nu\boldsymbol{k}}\,R_{\nu\boldsymbol{k},cs\boldsymbol{k}}}{E_{\nu\boldsymbol{k}}-E_{c\boldsymbol{k}}-\hbar\omega}+\frac{R_{cs'\boldsymbol{k}',\nu\boldsymbol{k}'}\,V_{\nu\boldsymbol{k}',cs\boldsymbol{k}}}{E_{\nu\boldsymbol{k}'}-E_{c\boldsymbol{k}}\pm\hbar\Omega_{\boldsymbol{k}-\boldsymbol{k}'}}\right)\,. \qquad (1)$$

Here $E_{c\boldsymbol{k}}$, $E_{c\boldsymbol{k}'}$ and E_ν are the electron energies in the initial $|c,s,\boldsymbol{k}\rangle$, final $|c,s',\boldsymbol{k}'\rangle$ and intermediate $|\nu\rangle$ states, respectively, s is the spin index, $\boldsymbol{k}$ is the electron wavevector, R is the matrix element of electron interaction with the electromagnetic wave, V is the matrix element of electron-phonon or electron-defect interaction, and $\hbar\Omega_{\boldsymbol{k}-\boldsymbol{k}'}$ is the energy of the involved phonon. At static defect assisted scattering $\hbar\Omega = 0$. The sign $\pm$ in Eq. (1) correspond to emission and absorption of phonons. A dominant contribution to the light absorption due to indirect transitions in the conduction band is caused by processes with intermediate states in the same band (see Fig. 1). The corresponding matrix element has the form

$$M_{cs'\boldsymbol{k}'\leftarrow cs\boldsymbol{k}} \propto \frac{V_{cs'\boldsymbol{k}',cs\boldsymbol{k}}}{\hbar\omega}\,\frac{m_0}{\hbar}\,\boldsymbol{e}\cdot\left(\frac{\partial E_{c\boldsymbol{k}'}}{\partial\boldsymbol{k}'}-\frac{\partial E_{c\boldsymbol{k}}}{\partial\boldsymbol{k}}\right)\,, \qquad (2)$$

where m_0 is the free electron mass, and $\boldsymbol{e}$ is the unit vector of the electric field polarization. The absolute value of the matrix element in Eq. (2) is independent of the degree of circular polarization of radiation, P_{circ}. Hence, the intraband transitions with intermediate states in the same band *do not* contribute to the optical orientation.

Spin orientation caused by intraband absorption of circularly polarized light can be obtained considering processes with intermediate states in the valence band and taking into account its spin-orbit splitting. Fig. 2 demonstrates schematically the spin orientation at intraband absorption of right handed circularly polarized light (σ^+). Because of the selection rules for interband optical matrix elements, the electron transitions with spin reversal $-1/2 \to +1/2$ are possible via intermediate states in the light-hole and spin-orbit split subbands, while the opposite processes, $+1/2 \to -1/2$ are forbidden. We assume the photon energy $\hbar\omega$ to exceed the typical energy of equilibrium electrons, $\hbar\omega \gg k_BT$ for a nondegenerate electron gas at the temperature T, or $\hbar\omega \gg \varepsilon_F$ for the case of degenerate statistics with the Fermi energy ε_F. Considering acoustic-phonon-assisted light absorption as an example, the following equation for the spin generation rate is obtained

$$\dot{\boldsymbol{S}} = \frac{1}{6}\frac{\Xi_{cv}^2}{\Xi_c^2}\frac{\Delta_{so}^2}{E_g(E_g+\Delta_{so})(3E_g+2\Delta_{so})}\,K\,\hat{\boldsymbol{e}}\,I\,P_{circ}\,. \qquad (3)$$

Here Ξ_{cv} and Ξ_c denote the interband and intraband deformation potential constants, respectively, E_g stands for the energies of the band gap and Δ_{so} for the valence band spin-orbit

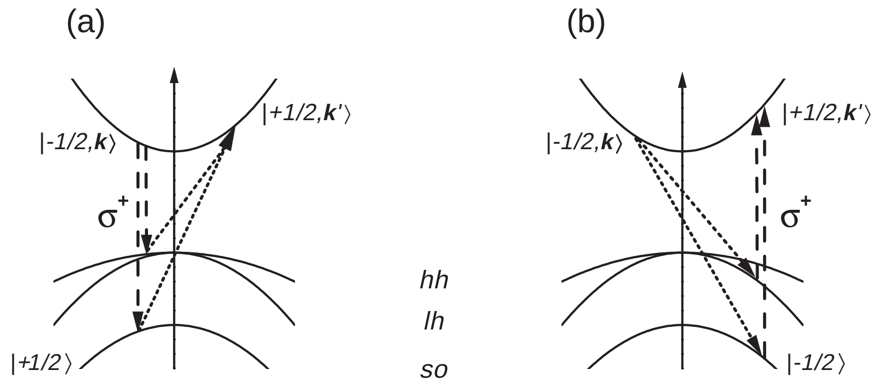

Figure 2. Sketch of indirect intraband optical transitions with intermediate states in the valence band. Dashed and dotted curves indicate the electron-photon interaction and the electron momentum scattering.

splitting, I is the light intensity, and $\hat{\boldsymbol{e}}$ is the unit vector in the light propagation direction. The factor K in Eq. (3) is the coefficient of the light absorption under phonon-assisted intraband optical transitions. It is determined by the dominant processes with intermediate states in the same band, and has the form

$$K = \frac{4\alpha}{3n_\omega}\left(\frac{\Xi_c}{\hbar\omega}\right)^2 \frac{k_B T}{\rho v_s^2}\left(\frac{2m^*\omega}{\hbar}\right)^{1/2} N_e \,, \tag{4}$$

where α and n_ω are the fine structure constant and the refraction index of the medium, respectively, ρ is the mass density of the crystal, v_s is the sound velocity, m^* is the effective mass of the conduction electron, and N_e is the carrier concentration.

It should be emphasized that the spin generation depends strongly on the energy of spin-orbit splitting of the valence band, Δ_{so}. It is due to the fact that the contributions to the matrix element giving rise to the optical orientation from the valence band Γ_8 and spin-orbit split band Γ_7 have the opposite signs. An estimation for GaAs material shows that the typical value of spin generated per one absorbed photon is of order 10^{-6} at a photon energy of $\hbar\omega = 10$ meV.

3. Monopolar optical spin orientation in QWs

3.1. Experimental technique

Experimentally, the monopolar spin orientation has been investigated on MBE (001)-grown n-GaAs/AlGaAs QW of width $d_W = 7$ nm and n-$In_{0.2}Ga_{0.8}As$ QWs of 7.6 nm width. Samples with free carrier densities of about $2 \cdot 10^{11}$ cm^{-2} were studied in the temperature range from liquid helium to room temperature. A pair of ohmic contacts was centered on opposite sample edges along the direction $x \parallel [1\bar{1}0]$ (see in Fig. 3).

A high power pulsed mid-infrared (MIR) TEA-CO_2 laser and a far-infrared (FIR) NH_3 laser were used as radiation sources delivering 100 ns pulses with radiation power P up to 100 kW. Several lines of the CO_2 laser between 9.2 μm and 10.6 μm and of the NH_3-laser [2] between λ = 76 μm and 280 μm were chosen for excitation in the MIR and FIR range, respectively. Excitation of samples by FIR radiation with photon energy less than the separation of size-quantized subbands leads to absorption caused by indirect optical

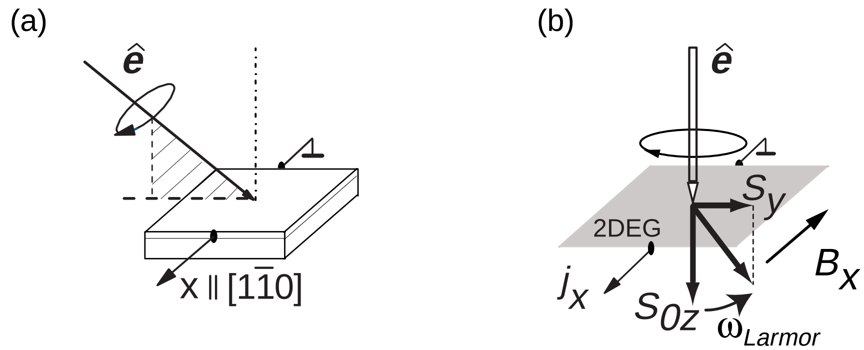

Figure 3. Experimental set-up used for measuring of (a) the circular photogalvanic effect and (b) the spin-galvanic effect in (001)-grown QWs. The photocurrent occurs in the first case at oblique incidence only and in the second case at normal incidence in combination with an in-plane magnetic field B_x. The current flow in both, the circular photogalvanic effect and the spin-galvanic effect, is driven by an asymmetric distribution of carriers in $\boldsymbol{k}$-space in systems with lifted spin degeneracy due to $\boldsymbol{k}$-linear terms in the Hamiltonian. The spin-galvanic effect is caused by asymmetric spin-flip scattering of spin polarized carriers and it is determined by the process of spin relaxation. If spin relaxation is absent, the effect vanishes. In contrast, the circular photogalvanic effect is the result of selective photoexcitation of carriers in $\boldsymbol{k}$-space with circularly polarized light due to optical selection rules.

transitions in the lowest subband (Drude absorption). The MIR radiation induces direct optical transitions between the first and the second subband of QWs.

The laser light polarization was modified from linear to circular using a Fresnel rhombus and quartz $\lambda/4$ plates for MIR and FIR radiation, respectively. The helicity of the incident light was varied according to $P_{circ} = \sin 2\varphi$ where φ is the angle between the initial plane of linear polarization and the optical axis of the polarizer. Spin polarization has been investigated making use of the circular photogalvanic effect (CPGE) [3] and the spin-galvanic effect (SGE) [4]. The experimental procedure is sketched in Fig 3. For investigation of the spin-galvanic effect an in-plane magnetic field B up to 1 T has been applied. The current j generated by polarized light in the unbiased structures was measured via the voltage drop across a 50 Ω load resistor in a closed circuit configuration. The voltage was recorded with a storage oscilloscope. The measured current pulses of 100 ns duration reproduce the temporal structure of the laser pulses.

3.2. Intra-subband transitions in QWs

In quantum well structures absorption of infrared radiation may be achieved by indirect intra-subband optical transitions and, for photon energies being in resonance with the energy distance between size quantized subbands, by direct transitions between these subbands. To obtain absorption caused by indirect transitions we used FIR radiation in the range from 76 μm to 280 μm (corresponding photon energies are from 16 meV to 4.4 meV). The experiments were carried out on GaAs and InAs QWs with the energy separation $\Delta E = E_2 - E_1$ between $e1$ and $e2$ size-quantized subbands equal to 120 meV and 112 meV, respectively. Therefore the photon energies used are much smaller than ΔE and absorption is caused by indirect intra-subband optical transitions. With illumination of (001)-grown GaAs and InAs QWs at oblique incidence of FIR radiation a current signal proportional to the helicity P_{circ} has been observed (see Fig. 4a) indicating the circular photogalvanic effect [3]. At normal incidence of radiation,

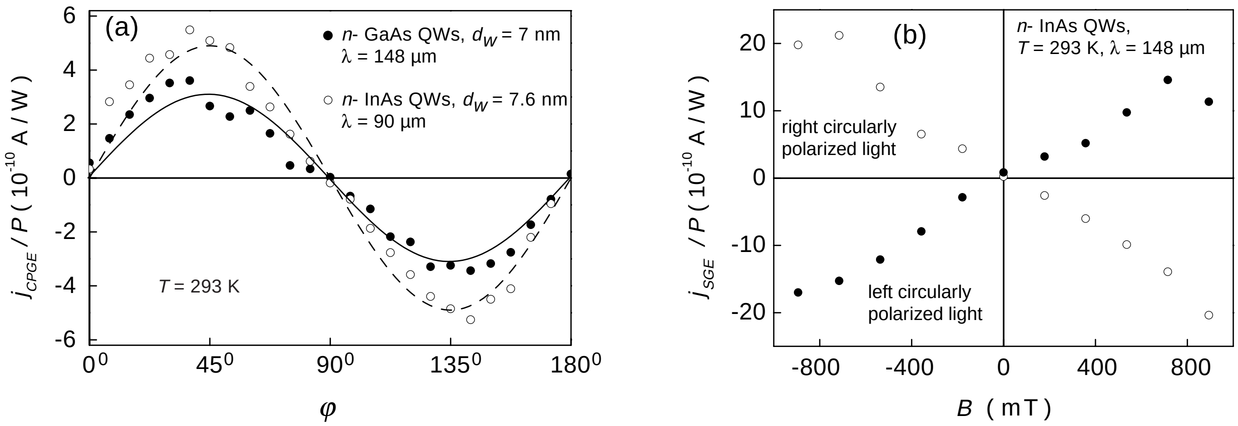

Figure 4. Monopolar spin orientation due to indirect intra-subband transitions within the $e1$ conduction subband in QW structures demonstrated by (a) the circular photogalvanic effect: the photocurrent in QWs normalized by light power P is plotted as a function of the phase angle φ defining helicity. Full lines are fitted using one parameter according to $j \propto \sin 2\varphi$, (b) the spin-galvanic effect: the figure shows magnetic field dependence of the photocurrent normalized by light power P. Measurements are presented for $T = 293$ K.

where the CPGE vanishes, the spin-galvanic current [4] is also observed applying an in-plane magnetic field (see Fig. 4b). Both effects are due to spin orientation. Therefore the observation of the CPGE and the spin-galvanic effect gives clear evidence that the absorption of far infrared circularly polarized radiation results in spin orientation. We note, that monopolar orientation has also been observed for p-type QW structures, but this is out of the scope of the present paper.

Monopolar optical orientation caused by indirect intra-subband transitions in QWs can be obtained in the similar way as that for bulk semiconductors (see previous section). For this particular mechanism of the monopolar optical orientation, the spin generation rate has been derived for two scattering processes: acoustic phonons and elastic scattering on static defects.

Considering acoustic-phonon-assisted processes with virtual intermediate states in the heavy-hole hh, light-hole lh, and spin-orbit split so subband, we obtain for the spin generation rate

$$\dot{\boldsymbol{S}}_{ph} = \frac{1}{6}\frac{\Xi_{cv}^2}{\Xi_c^2}\frac{\Delta_{so}^2}{E_g(E_g+\Delta_{so})(3E_g+2\Delta_{so})}\left[\hat{\boldsymbol{e}}_{\parallel} + \frac{d_W}{3}\sqrt{\frac{2m^*\omega}{\hbar}}\,\hat{\boldsymbol{e}}_z\right]\eta_{ph}\, I\, P_{circ}, \quad (5)$$

where d_W is the width of a QW, η_{ph} is the fraction of the energy flux absorbed in a QW under normal incidence. It is given by

$$\eta_{ph} = \frac{3\pi\alpha}{n_\omega}\left(\frac{\Xi_c}{\hbar\omega}\right)^2\frac{k_B T}{\rho a v_s^2}N_e\,, \quad (6)$$

where N_e is the concentration of the two-dimensional electron gas.

In the case of elastic scattering on static impurities, the expression for spin generation rate has the form

$$\dot{\boldsymbol{S}}_{imp} = \frac{2}{3}\frac{\Delta_{so}^2}{E_g(E_g+\Delta_{so})(3E_g+2\Delta_{so})}\left[\frac{V_{\parallel}^2}{V_0^2}\hat{\boldsymbol{e}}_{\parallel} + \frac{V_z^2}{V_0^2}\hat{\boldsymbol{e}}_z\right]\eta_{imp}\, I\, P_{circ}\,. \quad (7)$$

Here V_0 is the intraband matrix element of scattering, the factors V_z and $V_{\parallel}$ describe impurity-induced mixing of the conduction band Bloch function S with the valence band Bloch

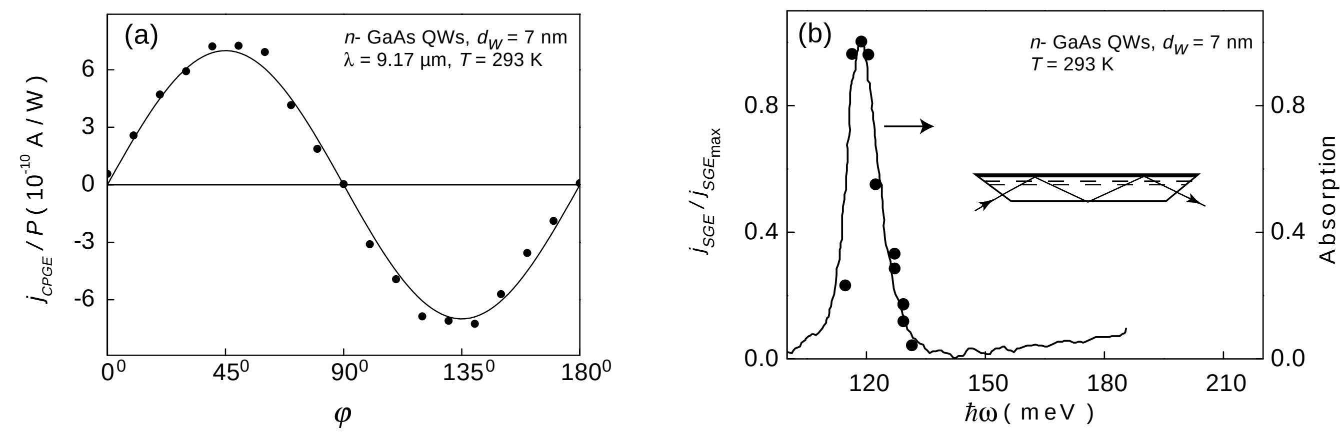

Figure 5. Monopolar spin orientation due to direct inter-subband transitions between $e1$ and $e2$ conduction subbands in QW structures demonstrated by (a) the photogalvanic effect: the photocurrent in QWs normalized by light power P is plotted as a function of the phase angle φ defining helicity. Full lines are fitted using one parameter according to $j \propto \sin 2\varphi$, (b) the spin-galvanic effect: the figure shows the spectral dependence of the magnetic field induced photocurrent (dots). Data are presented for optical excitation at normal incidence of right-handed circularly polarized radiation. A magnetic field of $B = 1$ T was used. For comparison the absorption spectrum is shown by the full line. The absorption has been determined by transmission measurements making use of a multiple-reflexion waveguide geometry shown in the inset. Results are plotted for (001)-grown GaAs QWs of 7 nm width at room temperature.

functions Z and X, Y respectively. The fraction of the energy flux absorbed in the QW at impurity-assisted intrasubband optical transitions under normal incidence is given by

$$\eta_{imp} = \frac{2\pi\alpha}{n_\omega} \left(\frac{V_0}{\hbar\omega} \right)^2 N_e \, N_d \,, \tag{8}$$

where N_d is the concentration of scatterers.

3.3. Direct inter-subband transitions in QWs

Absorption of radiation in the range from 9 μm to 11 μm in our GaAs and InAs samples is dominated by resonant direct inter-subband optical transitions between the first and the second subband. Fig. 5b shows the resonance behaviour of absorption measured in GaAs QWs by making use of transmission Fourier spectroscopy in a multiple-reflexion waveguide geometry (see inset Fig. 5b). Applying MIR radiation of the CO_2 laser, which causes direct transitions in GaAs and InAs QWs, the circular photogalvanic current at oblique incidence (Fig. 5a) and the spin-galvanic current at normal incidence of radiation (Fig. 5b) have also been observed. The wavelength dependence of the spin-galvanic effect obtained between 9.2 μm and 10.6 μm repeats the spectral behaviour of direct inter-subband absorption. This unambiguously demonstrates that in this case the spin orientation of n-type QWs is obtained by inter-subband transitions.

We would like to emphasize that spin sensitive inter-subband transitions in n-type QWs have been observed at normal incidence when there is no component of the electric field of the radiation normal to the plane of the QWs. Generally it is believed that inter-subband transitions in n-type QWs can only be excited by infrared light polarized in the growth direction z of the QWs [5]. Furthermore such transitions are spin insensitive and, hence, do not lead to optical orientation. Since the argument, leading to these selection rules, is based on the effective mass approximation in a single band model, the selection rules are not rigorous.

In order to explain the observed spin orientation as well as the absorption of light polarized in the plane of the QW we show that a $\mathbf{k}\cdot\mathbf{p}$ admixture of valence band states to the conduction band wave functions has to be taken into account. Calculations yield that inter-subband absorption of circularly polarized light propagating along z induces only spin-flip transitions resulting in 100% optical orientation of photoexcited carriers. In this geometry the spin generation rate for resonant inter-subband transitions has the form

$$\dot{S}_z = \frac{128\alpha}{9n_\omega}\,\frac{\Delta_{so}^2(2E_g+\Delta_{so})^2E_1}{E_g^2(E_g+\Delta_{so})^2(3E_g+2\Delta_{so})^2}\,\frac{\hbar^2N_e}{m^*}\,\delta(\hbar\omega - E_1 + E_2)\,I\,P_{circ} \quad (9)$$

where the function δ describes the resonant behaviour of the inter-subband transitions.

4. Summary

In conclusion, our results demonstrate that in n-type bulk semiconductors as well as QW structures monopolar spin orientation can be achieved applying circularly polarized radiation with photon energies less than the fundamental energy gap. Spin orientation has been observed by indirect intra-subband absorption as well as by inter-subband transitions. It is shown that monopolar spin orientation in n-type materials becomes possible if an admixture of valence band states to the conduction band wave function and the spin-orbit splitting of the valence band are taken into account. We emphasize that the spin generation rate under monopolar optical orientation depends strongly on the energy of spin-orbit splitting of the valence band, Δ_{so}. It is due to the fact that the valence band Γ_8 and the spin-orbit split band Γ_7 contribute to the matrix element of spin-flip transitions with opposite signs.

5. Acknowledgments

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