Optical Spin Orientation under Inter- and Intra-Subband Transitions in QWs

S.A. Tarasenko¹, E.L. Ivchenko¹, V.V. Bel'kov¹, S.D. Ganichev², D. Schowalter², Petra Schneider², M. Sollinger², W. Prettl², V.M. Ustinov¹, A.E. Zhukov¹, and L. E. Vorobjev³

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 achieved by inter-subband and intra-subband (Drude-like) transitions results in a monopolar spin orientation of free carriers. The monopolar spin polarization in zinc-blende-based quantum wells (QWs) is demonstrated by the observation of the spin-galvanic and circular photogalvanic effects. It is shown that monopolar spin orientation in n-type QWs 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.

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]. In this paper we show that the absorption of terahertz radiation with photon energies much less than the energy gap also leads to spin orientation of free carriers. This optical orientation may be referred to as 'monopolar' because only one type of carriers, electrons or holes, is excited.

We present theoretical and experimental results on monopolar optical orientation of a two-dimensional electron gas or hole gas. Both direct inter-subband and indirect (Drude-like) intra-subband transitions induced by circularly polarized radiation are considered for *n*- and *p*type QWs based on zinc-blende-structure semiconductors. A transfer of the photon angular momentum to the electron spin is linked to the spin-orbit interaction. Therefore, 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. Generally the expression for the generation rate of electron spin polarization due to optical excitation can be written as

$$\dot{S} = s(\eta I/\hbar\omega) P_{circ} , \qquad (1)$$

where s is the average electron spin generated per one absorbed photon of circularly polarized radiation, η is the fraction of the energy flux absorbed in the QW, I is the light intensity, $\hbar\omega$ is the photon energy, and P_{circ} is the degree of circular polarization. While optical orientation at interband excitation has been widely studied, it is not obvious that inter-subband and intra-subband transitions can result in a spin polarization. Below we show that for both kinds of transitions within one band - valence or conduction band - absorption of circularly polarized light leads to spin polarization which has not been considered previously.

Experimental technique

The experiments have been carried out on MBE (001)-grown *n*-GaAs/AlGaAs QW of 7 nm width, *n*-GaAs/AlGaAs single heterojunction, $In_{0.2}Ga_{0.8}As$ QWs of 7.6 nm width, and on *p*-type GaAs/AlGaAs QWs of 20 nm width MOCVD grown on (001)-miscut substrates as well as (113)-MBE-grown samples with QWs of various widths between 7 and 15 nm. 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 $x \parallel [1\overline{10}]$ (see inset in Fig. 1).

A high power pulsed mid-infrared (MIR) TEA- CO_2 laser and a far-infrared (FIR) NH₃ laser were used as radiation sources delivering 100 ns pulses with radiation power P up to 100 kW. Several lines of the CO₂ laser between $9.2\,\mu\mathrm{m}$ and $10.6\,\mu\mathrm{m}$ and of the NH₃-laser [2] between λ $= 76 \,\mu\text{m}$ and $280 \,\mu\text{m}$ were chosen for excitation in the MIR and FIR range, respectively. In *n*-type samples the MIR radiation induces direct optical transitions between the first and the second subband of QWs. Direct transitions between heavy-hole and light-hole subbands have been achieved in *p*-type samples applying FIR radiation in the spectral range between $76 \,\mu\text{m}$ and $148 \,\mu\text{m}$. In both n- and p-type samples application of FIR radiation with photon energies less than the separation of subbands leads to absorption caused by indirect optical transitions in the lowest subband (Drude absorption).

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 [4]. For investigation of spin-galvanic effect an in-plane magnetic field B up to 3 T has been applied as shown in the inset of Fig. 1. The samples were placed in a magnetooptical cryostat with a split-coil superconducting magnet. 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.

Direct inter-subband transitions

With illumination of (001)-oriented *n*-type GaAs and InAs QWs at oblique incidence of MIR radiation of the CO_2 laser a current signal proportional to the helicity P_{circ} has been observed indicating the circular photogalvanic effect [3]. At normal incidence of radiation, at which the CPGE effect vanishes, the spin-galvanic current [4] is also observed applying an in-plane magnetic field. Both effects are due to spin orientation, therefore the observation of the CPGE and the spin-galvanic effect clearly gives evidence that the absorption of infrared circularly polarized radiation results in spin orientation. The wavelength dependence of the spin-galvanic effect obtained between 9.2 μ m and 10.6 μ m repeats the spectral behaviour of direct intersubband absorption between the first and the second subbands measured in transmission by Fourier spectroscopy in multipath geometry. 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 intersubband 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 QW. 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, s = 1. In this geometry the fraction of the energy flux absorbed in the QW by transitions from the first subband e1 to the second subband e2 has the form



Figure 1: Spectral dependence of the spin-galvanic current (dots) caused by spin orientation due to direct optical transitions between e1 and e2 conduction subbands. The inset shows the geometry of the experiment. Optical excitation is at normal incidence of right-handed cicularly polarized radiation. For comparison the absorption spectrum obtained from transmission in a multiple-reflection waveguide geometry shown by the full line. Results are plotted for (001)-grown GaAs QWs of 7 nm width at room temperature.

$$\eta = \frac{128\alpha}{9n_{\omega}} \frac{\Delta_{so}^2 (2E_g + \Delta_{so})^2 (E_2 - E_1) E_1}{E_g^2 (E_g + \Delta_{so})^2 (3E_g + 2\Delta_{so})^2} \frac{\hbar^2 n_e}{m^*} \,\delta(\hbar\omega - E_1 + E_2),\tag{2}$$

where α is the fine structure constant, n_{ω} is the refraction index, n_e is the 2D electron concentration, m^* is the effective mass, $\hbar \omega$ is the photon energy, E_g is the energy gap, Δ_{so} is the valence band spin-orbit splitting, E_1 and E_2 are the energies of size-quantization of the e1 and e2 subbands, respectively. The δ -function describes the resonant behaviour of the inter-subband transitions.

The helicity dependent current of the CPGE has also been observed in *p*-type GaAs QWs due to transitions between heavy-hole (hh1) and light-hole (lh1) subbands demonstrating spin orientation of holes. The measurements were carried out on (001)-miscut QWs and (113)-oriented QWs which, in contrast to (001)-oriented samples and in accordance to the phenomenological theory of the CPGE. yield a maximum of the CPGE at normal incidence of radiation. QWs with various widths in the range from 7 to 20 nm were investigated. For direct inter-subband transitions photon energies between 35 meV and 8 meV of FIR radiation corresponding to these QW widths were applied. Due to the different effective masses of light and heavy holes the absorption does not show narrow resonances. A spin-galvanic current in p-type QWs could not be detected because of the very small in-plane g-factor for

heavy holes [6] which makes the effect of the magnetic field negligible [4]. Optical orientation caused by heavy-hole to light-hole absorption of circularly polarized radiation occurs for transitions at in-plane wavevector $\mathbf{k} \neq 0$ due to the mixing of heavy-hole and light-hole subbands

Intra-subband transitions

In the far-infrared range, where the photon energy is not enough for direct inter-subband transition in n- or even in p-type samples, the absorption of light by free carriers occurs by indirect intra-subband transitions where momentum conservation law is satisfied due to acoustic or optical phonons, static defects etc. In the case of such free carrier absorption spin orientation is obtained as well. This is proved by the observation of the CPGE in both n- and p-type samples in response to FIR radiation. For n-type samples also the spin-galvanic effect caused by spin orientation was detected (Fig. 2).



Figure 2: Magnetic field dependence of the spin-galvanic current achieved by intra-subband transitions within e1 conduction subband by the excitation with the radiation of 280 μ m wavelength. Results are plotted for an (001)-grown GaAs single heterojunction at room temperature.

The experiments were carried out on *n*-type (001)-oriented GaAs and InAs QWs applying FIR radiation in the range from 76 μ m to 280 μ m (16 meV - 4.4 meV which is much less than $E_2 - E_1 = 120$ meV). For *p*-type materials long wavelength laser lines from 280 μ m to 496 μ m (4.4 meV-2.5 meV) were used to ensure free carrier intra-subband absorption for all QW widths. In this case the photon energies are less than hh1-lh1 energy separation.

The intra-subband optical transitions in QWs involving both the electron-photon interaction and momentum scattering are described by second-order processes with virtual intermediate states. A dominant contribution to the optical absorption is caused by processes with intermediate states in the same subband. This is the channel that determines the coefficient of intra-subband absorption, η . However such transitions conserve the electronic spin and, hence, do not lead to an optical orientation.

In order to obtain optical orientation due to intra-subband transitions we considered virtual interband transitions with intermediate states in the valence band. Taking into account the spin-orbit splitting of the valence band and the selection rules for interband transitions, the absorption of circularly polarized light gives rise to the spin orientation of electrons.

For this particular mechanism of monopolar optical orientation one can derive the following expression for the spin generated per one absorbed photon of right-handed circularly polarized radiation

$$s \propto \frac{V_{cv}^2}{V_c^2} \frac{\hbar\omega \,\Delta_{so}^2}{E_g(E_g + \Delta_{so})(3E_g + 2\Delta_{so})} \,. \tag{3}$$

Here V_c and V_{cv} are the intraband and interband matrix elements of scattering, respectively. The prefactor in Eq.(3) depends on the mechanism of momentum scattering. An estimation for typical GaAs/AlGaAs QWs shows that $s \simeq 10^{-6}$ for acoustic phonon assisted indirect optical transitions at the photon energy $\hbar \omega = 10$ meV.

In conclusion, our results demonstrate that in both *n*- and *p*-type QWs monopolar spin orientation can be achieved applying radiation with photon energies less than the fundamental energy gap. Spin orientation has been observed by direct inter-subband transitions as well as by Drude-like intra-subband absorption. It is shown that monopolar spin orientation in n-type QWs 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 Γ_8 and spin-orbit split band Γ_7 contribute to the matrix element of spin-flip transitions with opposite signs.

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