Circular Photogalvanic Effect at Inter-Band Excitation in Semiconductor Quantum Wells

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We observed a circular photogalvanic effect (CPGE) in GaAs quantum wells at inter-band excitation. The spectral dependence of the CPGE is measured together with that of the polarization degree of the time resolved photoluminescence. A theoretical model takes into account spin splitting of conduction and valence bands.

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Spin photocurrents generated by excitation with circularly polarized radiation in quantum wells have attracted considerable attention in the recent decade. Several mechanisms of electric currents driven by optically generated spin polarization are observed in zinc-blende-structure based bulk semiconductors and quantum wells (QWs). Among these effect are inhomogeneous spin orientation induced currents in bulk GaAs, the circular photogalvanic effect (CPGE) and the spin-galvanic effect in QWs, the photovoltaic effect in $p-n$ junctions and currents due to quantum interference of one- and two-photon excitations. Except CPGE in QWs, all other spin photocurrents have been observed at optical excitation across the band gap of the semiconductor. CPGE is caused in zinc-blende structure based QWs by homogeneous optical spin orientation of carriers. This effect should also occur at inter-band excitation, but so far has been detected only at intra-band transitions by excitation with infrared radiation. In the present work we report on the first observation of the CPGE at inter-band excitation in GaAs QWs.

The experiments were carried out on (113)A-oriented molecular-beam-epitaxy (MBE) grown $p$-type GaAs/Al$_{0.32}$Ga$_{0.68}$As structure with 20 QW of 15 nm widths. The free hole density in the sample was $2 \cdot 10^{11}$ cm$^{-2}$ and the mobility was about $5 \cdot 10^5$ cm$^2$/Vs at 4.2 K. The sample edges were oriented along the [110]- and [332]-directions. Two pairs of ohmic contacts were centered along opposite sample edges pointing in the directions $x \parallel [110]$ and $y \parallel [332]$ (see Fig. 1). The sample belongs to the symmetry class $C_{\infty v}$ which allows the CPGE at normal incidence of the radiation. For optical inter-band excitation a cw-Ti:sapphire laser and pulsed Ti:sapphire laser were used providing radiation of wavelength in the range between 0.7 $\mu$m and 0.85 $\mu$m. The power of the cw-laser was about 80 mW. The pulsed laser provided 1 ps pulses with a repetition rate of 80 MHz and an average power of about 100 mW. One of the main features of the CPGE is that the photocurrent caused by spin polarization is proportional to the helicity of the incident light $P_{circ} = (I_{\sigma_+} - I_{\sigma_-})/(I_{\sigma_+} + I_{\sigma_-})$, where $I_{\sigma_+}$ and $I_{\sigma_-}$ are intensities of right- ($\sigma_+$) and left-handed ($\sigma_-$) polarized radiation. Therefore, the sign of the current changes upon switching from right to left circular polarization. This allows to distinguish the helicity dependent photocurrent from helicity independent currents like that of the Dember effect, photovoltaic effects at contacts and Schottky barriers. The linearly polarized laser beam was transmitted through a photoelastic modulator which yields a periodically oscillating polarization between $\sigma_+$ and $\sigma_-$. The photocurrent $I_x$ was measured in the unbiased structures at room temperature via the voltage drop across a 50 $\Omega$ load resistor in a closed circuit configuration. The signal was recorded by a lock-in amplifier in phase with the photoelastic modulator. In addition we carried out polarization and time resolved photoluminescence measurements using a synchroscan streak camera with a temporal and spectral resolution of 7 ps and 1 nm, respectively.
Illuminating our QW structure with polarization modulated radiation at normal incidence we observed a current signal in \( x \)-direction for both \( \text{cw} \) and pulsed excitations. In the perpendicular \( y \)-direction no helicity dependent current has been detected. Fig. 1 shows the spectral dependence of the photocurrent and the spectrally integrated polarization degree of photoluminescence. Both, photocurrent and polarization degree have peculiarities in their spectral dependences. In particular, there is a deep drop of both values at the photon energy \( \hbar \omega = 1.67 \text{ eV} \). While the photocurrent exhibits a distinct minimum, the polarization degree sharply drops (by a factor of two) with increasing energy. In the next step we studied the temporal dependence of the polarization degree on the excitation photon energy. Fig. 2 shows that the relaxation time of electron spins \( \tau_s \) is about 170 ps and is independent of \( \hbar \omega \).

On the macroscopic level the CPGE can be described by the following phenomenological expression [15]:

\[
j_x = \sum_\mu \gamma_{\lambda\mu} i (E \times E^\ast)_\mu = \sum_\mu \gamma_{\lambda\mu} \hat{e}_\mu \bar{E}_0^2 P_{\text{circ}},
\]

where \( j \) is the photocurrent density, \( \gamma \) is a second rank pseudo-tensor, \( E \) is the complex amplitude of the electric field of the electromagnetic wave, and \( \bar{E}_0, \hat{e} \) are the electric field amplitude and the unit vector pointing in the direction of light propagation, respectively. For QWs of \( C_s \) symmetry, as investigated here, a photocurrent at normal propagation along \( z \parallel [113] \), is given by

\[
j_x = (\gamma_{xx} \hat{e}_x) \bar{E}_0^2 P_{\text{circ}}, \quad j_y = 0.
\]

The current flows in \( x \)-direction which is perpendicular to the mirror reflection plane of \( C_s \) symmetry. Eq. 2 describes the experimental observation, in particular, the absence of a helicity dependent current in \( y \)-direction.

Fig. 3 shows the microscopic picture of the spin orientation induced circular photogalvanic effect at inter-band excitation close to the band edge. We assume direct inter-band transitions in a QW of \( C_s \) symmetry. For the sake of simplicity we discuss a band structure consisting only of the lowest conduction subband \( e1 \) and the highest heavy-hole subband \( hh1 \). The energy dispersion in the conduction band is given by

\[
\epsilon_{e1,\pm 1/2}(k) = \frac{\hbar^2 k^2}{2m_e} \pm \beta_{e1} k_x + \varepsilon_g
\]

and in the valence band by

\[
\epsilon_{hh1,\pm 3/2}(k) = \frac{-\hbar^2 k^2}{2m_{hh1}} \pm \beta_{hh1} k_x,
\]

where \( \varepsilon_g \) is the QW energy gap, \( \beta_{e1} \) and \( \beta_{hh1} \) are components of second rank pseudo-tensor responsible for the removal of spin degeneracy in the first electron and the first heavy-hole subbands, respectively.

For absorption of circularly polarized radiation of photon energy \( \hbar \omega \), momentum and energy conservation allow transitions only for two values of \( k_x \). Due to the selection rules the optical transitions occur from \( s = -3/2 \) to \( s = -1/2 \) for right handed circular polarization (\( \sigma_+ \)) and from \( s = 3/2 \) to \( s = 1/2 \) for left handed circular polarization (\( \sigma_- \)). Here \( s \) are the spin quantum numbers of the electron states. The corresponding transitions for e.g. \( \sigma_+ \) photons occur at

\[
k_x^\pm = \frac{\mu}{\hbar^2} (\beta_{e1} + \beta_{hh1}) \pm \sqrt{\left[ \frac{\mu^2}{\hbar^4} (\beta_{e1} + \beta_{hh1})^2 + \frac{2\mu}{\hbar^2} (\hbar \omega - \varepsilon_g) \right]}.
\]

and are shown in Fig. 3 by the solid vertical arrows. Here \( \mu = (m_e \cdot m_{hh1})/(m_e + m_{hh1}) \) is the reduced mass. The...
the subbands, and \( \dot{\tau} \). Thus the sum of the electron velocities in the excited states in the conduction band,

\[
v_{e,t} = \frac{\hbar}{m_e} (k_x^+ + k_x^- - 2k_x^{\text{min}}) = \frac{2}{\hbar} \frac{\beta_{hl} m_{hl} - \beta_{el} m_e}{m_e + m_{hl}},
\]

is non-zero. The contributions of \( k_x^\pm \) photoelectrons to the current do not cancel each other except in the case of \( \beta_{el} m_e = \beta_{hl} m_{hl} \) which corresponds to an equal splitting of the conduction and the valence band. We note that the group velocity is obtained taken into account that \( k_x^\pm \) are to be counted from the conduction subband minima \( k_x^{\text{min}} \) because the current is caused by the difference of the group velocities within the subband. The same consideration applies for holes in the initial states in \( hh1 \). Consequently, a spin polarized net current results in the \( x \) direction. Changing the circular polarization of the radiation from \( \sigma+ \) to \( \sigma- \) reverses the current because the ‘center of mass’ of these transitions is now shifted to \(- (\beta_{el} + \beta_{hl}) (\mu / h^2)\).

The microscopic theory of spin orientation induced CPGE in QWs at inter-band excitation was developed in \[17\]. We consider the asymmetry of the momentum distribution of electrons excited under direct inter-band optical transitions in \( p \)-doped \((113)\)-grown QWs of \( C_s \) symmetry. The photocurrent density is given by

\[
j = e \sum_{\mathbf{k}} \text{Tr} \left[ \mathbf{v}^{(e)}(\mathbf{k}) \tau^{(e)} \mathbf{\rho}^{(e)}(\mathbf{k}) + \mathbf{v}^{(h)}(\mathbf{k}) \tau^{(h)} \mathbf{\rho}^{(h)}(\mathbf{k}) \right],
\]

where \( e \) is the elementary charge, \( \mathbf{v}^{(e,h)}(\mathbf{k}) = (1/\hbar) \partial E^{(e,h)}(\mathbf{k}) / \partial \mathbf{k} \) are the group velocities of electrons and holes, \( \tau^{(e,h)} \) are the momentum relaxation times in the subbands, and \( \mathbf{\rho}^{(e,h)} \) are their non-equilibrium generation rates. The expressions for them have the form

\[
\mathbf{\rho}^{(e,h)} = \frac{\pi}{\hbar} \sum_{n,n'} M_{nl} M_{n'l'} [\delta(E_n + E_l - \hbar \omega) + \delta(E_{n'} + E_{l'} - \hbar \omega)].
\]

Here \( M_{nl}(\mathbf{k}) \) is the inter-band optical matrix element, and the indices \( n, n', l \) enumerate the electron and hole subband spin states involved into the transitions. The CPGE current excitation spectrum is given by

\[
j_{x}(\omega) = \sum_{\nu,\nu'} [\beta_{e\nu} F_{\nu\nu'}(\omega) + \beta_{h\nu} F_{\nu\nu'}(\omega)],
\]

where \( \beta_{e\nu}, \beta_{h\nu} \) are the spin-splitting parameters for \( \nu^{\text{th}} \) electron and \( \nu^{\text{th}} \) hole subbands of size-quantization. The spectral functions \( F_{\nu\nu'}(\omega) \) and \( \tilde{F}_{\nu\nu'}(\omega) \) are calculated in \[17\].

The most important result of the microscopic theory is that both the initial and final states of the carriers involved in the optical transition contribute to the circular photogalvanic current with different strengths and directions. The partial currents are proportional to the group velocity being dependent on \( \mathbf{k} \), the momentum relaxation times \( \tau_e \) and \( \tau_h \), and the occupation of the initial states. Therefore the sign of the total current depends on the details of experimental conditions and may even change by varying the radiation frequency, temperature etc.

The photocurrent is due to carrier spin polarization, thus the coincidence of spectral peculiarities in both the CPGE and the circular polarization degree is natural. The reason for the peculiarities in both spectra is a sharp dependence of spin polarization on the wavevector for the direct inter-band transitions. Close to the band edge, the transitions obey the simple selection rules and occur from pure \( \pm 3/2 \) heavy-hole states as assumed in the model above. Increase of the excitation energy shifts the transitions to higher wavevectors at which the selection rules are lifted. This is caused by the heavy-light hole mixing in QWs leading to a strong dependence of carrier spin orientation on the excitation energy theoretically considered in \[18\]. On the other hand, transitions from hole subbands to electron continuum are possible at the excitation energy \( \hbar \omega \gtrsim 1.7 \text{ eV} \). Such process is known as ‘photoionization’ of QWs \[19\].

In conclusion we have observed the CPGE in QW structures at inter-band excitation for the first time. The strength of the CPGE signal correlates with the degree of spin polarization and is well described by phenomenological and microscopical theories.

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[1] S. D. Ganichev, and W. Prettl, J. Phys.: Condens. Matter, 15, R935 (2003).
[2] N.S. Averkiev, and M.I. D’yakonov, Fiz. Tekh. Poluprov. 17, 629 (1983) [Sov. Phys. Semicond. 17, 393 (1983)].
[3] A.A. Bakun, B.P. Zakharchenya, A.A. Rogachev, M.N. Tkachuk, and V.G. Fleisher, Pis’ma ZhETF 40, 464 (1984) [Sov. JETP Lett. 40, 1293 (1984)].
[4] S.D. Ganichev, E. L. Ivchenko, S.N. Danilov, J. Eroms, W. Wegscheider, D. Weiss, and W. Prettl, Phys. Rev. Lett. 86, 4358 (2001).
[5] S.D. Ganichev, E.L. Ivchenko, V.V. Bel’kov, S.A. Tarasenko, M. Sollinger, D. Weiss, W. Wegscheider, and W. Prettl, Nature (London) 417, 153 (2002).
[6] I. Zutić, J. Fabian, and S. Das Sarma, Appl. Phys. Lett. 79, 1558 (2001).
[7] I. Zutić, J. Fabian, and S. Das Sarma, Phys. Rev. Lett. 88, 066603 (2002).
[8] R.D.R. Bhat, and J.E. Sipe, Phys. Rev. Lett. 85, 5432 (2000).
[9] M.J. Stevens, A.L. Smirl, R.D.R. Bhat, J.E. Sipe, and H.M. van Driel, J. Appl. Phys. 91, 4382 (2002).
[10] J. Hübner, W.W. Rühle, M. Klude, D. Hommel R.D.R. Bhat, J.E. Sipe, and H.M. van Driel, Phys. Rev. Lett. 90, 216601 (2003).
[11] E.L. Ivchenko, and G.E. Pikus, Pis’ma ZhETF 27, 640 (1978) [Sov. JETP Lett. 27, 604 (1978)].
[12] V.I. Belinicher, Phys. Lett. A 66, 213 (1978).
[13] V.M. Asnin, A.A. Bakun, A.M. Danishevs’kii, E.L. Ivchenko, G.E. Pikus, and A.A. Rogachev, Pis’ma ZhETF 28, 80 (1978) [Sov. JETP Lett. 28, 74 (1978)].
[14] B.I. Sturman, and V.M. Fridkin, The Photovoltaic and Photorefractive Effects in Non-Centrosymmetric Materials, Gordon and Breach Science Publishers, New York, 1992.
[15] E.L. Ivchenko, and G.E. Pikus, Superlattices and Other Heterostructures. Symmetry and Optical Phenomena, (Springer, Berlin 1997).
[16] M. Oestreich, M. Bender, J. Hübner, D. Hägele, W. Rühle, Th. Hartmann, P.J. Klar, W. Heimbrot, M. Lampalzer, K. Volz, and W. Stolz, Semicond. Sci. Technol. 17, 285 (2002).
[17] L.E. Golub, Phys. Rev. B 67, 235320 (2003).
[18] I.A. Merkulov, V.I. Perel, and M.E. Portnoi, ZhETF 99, 1202 (1991) [Sov. Phys. JETP 72, 669 (1991)].
[19] Z.C. Feng (Ed.) Semiconductor interfaces, microstructures and devices, properties and applications, Bristol and Philadelphia, IoP, 1993.