Spin sensitive bleaching and monopolar spin orientation in quantum wells

S.D. Ganichev$^{1,2}$, S.N. Danilov$^1$, V.V. Bel’kov$^2$, E.L. Ivchenko$^2$, M. Bichler$^3$, W. Wegscheider$^{1,3}$, D. Weiss$^1$, and W. Prettl$^1$

$^1$ Institut für Experimentelle und Angewandte Physik, Universität Regensburg, D-93040 Regensburg, Germany

$^2$ Ioffe Physico-Technical Institute, RAS, 194021, St. Petersburg, Russia

$^3$ Walter Schottky Institut, TU München, 85748, Garching, Germany

(March 22, 2022)

Abstract

Spin sensitive bleaching of the absorption of far-infrared radiation has been observed in $p$-type GaAs/AlGaAs quantum well structures. The absorption of circularly polarized radiation saturates at lower intensities than that of linearly polarized light due to monopolar spin orientation in the first heavy hole subband. Spin relaxation times of holes in $p$-type material in the range of tens of ps were derived from the intensity dependence of the absorption.
A substantial portion of current research in condensed-matter physics is directed towards understanding various manifestations of spin-dependent phenomena like giant magnetoresistance, heavy fermions, Kondo scattering, superconductivity and others. In particular, the spin of electrons and holes in solid state systems is the decisive ingredient for active spintronic devices [1,2] and several schemes of quantum computation [3–5]. Especially the combination of ferromagnetic materials with semiconductors seems to be a promising combination for novel functional concepts. Open problems which have to be addressed in this respect involve spin-injection into semiconductors, spin relaxation in low dimensional semiconductor structures as well as spin detection. Significant progress was made recently: it was shown that spin polarized electrons (or holes) can be injected from semimagnetic (or ferromagnetic) semiconductor materials into semiconductors [6,7]. The presence of spin polarized electrons can be probed by analyzing the Kerr effect [8] or by analyzing the degree of circular polarization of light which gets emitted when polarized electrons (holes) recombine with holes (electrons). The inverse process, exciting free carriers by circularly polarized light due to optical orientation [9] is frequently used to prepare an ensemble of spin polarized carriers. In low-dimensional systems with band splitting in k-space due to k-linear terms in the Hamiltonian optical excitation not only leads to a spin polarized ensemble of electrons but also to a current whose sign and magnitude depends on the degree of circular polarization of the incident light (circular photogalvanic effect, [10]).

For the realization of spintronic devices sufficiently long spin dephasing times in semiconductor quantum well (QW) structures are crucially needed. Spin transport must occur without destroying the relevant spin information. Current investigations of the spin lifetime in semiconductor devices [11–16] are based on optical spin orientation by interband excitation and further tracing the kinetics of polarized photoluminescence and investigation of the dynamics of subsequent relaxation of spin polarized electron-hole pairs. Studies of bi-polar spin orientation, where both electrons and holes got excited, gave important insights into the mechanisms of spin relaxation of photoexcited free carriers. We show below that by combining the circular photogalvanic effect (CPGE) [10] with saturation (bleaching of absorption)
spectroscopy \[17\text{–}21\] we are able to probe spin relaxation for monopolar spin orientation. In contrast to the conventional methods of optical spin orientation, in our measurements only one type of charge carriers (electrons or holes) gets spin oriented and is involved in relaxation processes. This is achieved by using radiation in the terahertz range which excites intraband or intersubband, but no interband (from valence to conduction band), transitions. Monopolar spin orientation allows to study spin relaxation without electron-hole interaction and exciton formation. The most important advantage of monopolar spin orientation is that relaxation processes can be investigated for electrons in n-type material and for holes in p-type material; both can be measured independently. These conditions have not been met previously in quantum well structures where, due to interband excitation, only the spin relaxation times of optically generated minority carriers were accessible (for reviews see \[14\text{–}16\]).

Here we report the first observation of spin sensitive bleaching of the heavy hole \(hh1\)-light hole \(lh1\) absorption in \(p\)-type QW structures which allows to investigate spin relaxation for a monopolar spin orientation. The basic physics is sketched in Fig. 1. Exciting with circularly polarized light results in direct intersubband transitions (solid arrow) which depopulate and populate selectively spin states in the valence subbands (\(hh1\) and \(lh1\)) and cause a monopolar spin polarization. Spin relaxation inside the \(hh1\)-subband is characterized by the relaxation time \(\tau_s\). Relaxation from the \(lh1\)-subband back to \(hh1\) is characterized by a spin independent energy relaxation time \(\tau_e\) as the dominating transitions in this energy range are indirect (broken arrows in Fig. 1), mediated by phonons. The absorption coefficient \(\alpha\) is proportional to the difference of the populations of the initial and final states. At high intensities the absorption coefficient decreases since the photoexcitation rate becomes comparable non-radiative relaxation rate into the initial state. Thus absorption bleaching of circularly polarized radiation governed by both the hole spin relaxation in the initial state and the energy relaxation of photoexcited carriers characterized by the spin relaxation time \(\tau_s\) and the energy relaxation time \(\tau_e\), respectively. In contrast to circularly polarized light, optical transitions induced by linearly polarized light are not spin selective and the satu-
ration is controlled by the energy relaxation of photoexcited carriers only. The difference in absorption bleaching for circularly and linearly polarized radiation can be observed experimentally as is pointed out below. The method introduced here can also be applied for n-type QWs using direct inter-subband transitions in the conduction band.

The experiments have been carried out on modulated doped p-GaAs/AlGaAs (311)-MBE-grown samples with a single QW or 20 QWs of the width $L_W=15$ nm and with the period $d=1300$ nm for multiple QW-structures. Samples with free carrier densities $p_s$ of about $2 \cdot 10^{11}$ cm$^{-2}$ and mobilities around $5 \times 10^5$ cm$^2$/Vs were studied in the range from liquid helium to room temperature. A pair of ohmic contacts have been centered on opposite sample edges along the direction $x \parallel [1\bar{1}0]$ (see inset in Fig. 2) [10]. As terahertz radiation source a high power far-infrared molecular laser, optically pumped by a TEA-CO$_2$ laser, has been used delivering 100 ns pulses with intensities up to 1 MW/cm$^2$ at wavelength $\lambda =148$ µm. The radiation induces direct optical transitions between the first heavy-hole and the first light-hole valence subband (see Fig. 1). A crystalline quartz $\lambda/4$ plate has been used to obtain from the initially linearly polarized laser light circularly polarized radiation with the degree of circular polarization $P_{circ}$ equal to ±1 for right and left handed circularly polarized light, respectively.

The weak absorption of terahertz radiation by free carriers in QWs is difficult to determine by direct transmission measurements particularly in the case of bleaching at high power levels. Therefore the nonlinear behavior of the absorption has been investigated employing the recently observed circular and linear photogalvanic effects [10,22]. Both, the circular photogalvanic effect (CPGE) and the linear photogalvanic effect (LPGE) yield an electric current in $x$ direction [23]. The absorption coefficient is proportional to photogalvanic current $j_x$ normalized by the radiation intensity $I$ [24]. By choosing circularly or linearly polarized radiation we thus obtain a photoresponse corresponding to the absorption coefficient of circularly or linearly polarized radiation, respectively.

Our measurements, displayed in Fig. 2, indicate that the photocurrent $j_x$ depends at low power levels linearly on the light intensity and gradually saturates with increasing intensity.
\[ I \propto \frac{I}{1 + I/I_s} \]

where \( I_s \) is the saturation intensity. This behavior of the current corresponds to a constant absorption coefficient at low power levels and decreasing absorption with rising intensity. Saturation intensities \( I_s \) have been measured for a wide range of temperatures between 4.2 K and 200 K. The key experimental result is plotted in Fig. 3 and show that the magnitude \( I_s \) for the photogalvanic response on circularly polarized radiation is generally smaller than that for linearly polarized radiation. The experimentally obtained values increase from about 10 kW/cm\(^2\) at liquid helium temperature to 300 kW/cm\(^2\) at 200 K. At room temperature the saturation intensities get non-measurably large.

Over the whole temperature range the holes occupy in equilibrium the lowest heavy-hole subband \( hh_1 \). Absorption of far-infrared radiation occurs by direct optical transitions from \( hh_1 \) to the first light hole subband \( lh_1 \) and for the wavelength 148 \( \mu m \) (photon energy \( \hbar \omega = 8.3 \) meV) used here takes place close to \( k = 0 \), as is sketched in Fig. 1. Thus, the optical dipole selection rules for the absorption are \( \Delta m = \pm 1 \) with angular momentum quantum number \( m = \pm 3/2 \) for the initial state and \( m = \pm 1/2 \) for the final state [25]. The insets in Fig. 3 show the corresponding transitions for linear (top left) and circular (bottom right) polarization by full arrows. Linearly polarized radiation has been decomposed in right and left handed circularly polarized light of identical amplitudes. Broken lines in these insets indicate non-radiative energy and spin relaxation transitions.

Linearly polarized radiation (top left inset in Fig. 3) equally depopulates both spin-up and spin-down states of the heavy hole subband and populates the first light hole subband. With rising intensity these non-equilibrium populations approach each other causing the bleaching of absorption which is controlled by the energy relaxation time \( \tau_e \). In contrast to linear polarization the absorption of circularly polarized light is spin selective because only one type of spin component is involved in the absorption process (illustrated in the right bottom inset of Fig. 3). During energy relaxation to the initial state in subband \( hh_1 \) the holes loose the photoinduced orientation due to rapid relaxation [25]. Thus, spin orientation occurs in the initial subband \( hh_1 \), only. Bleaching of absorption is hence controlled by two time constants, the energy relaxation time \( \tau_e \) and the spin relaxation time \( \tau_s \). Note that \( \tau_e \)

\[ \tau_e = 5\]
is the same for circular and linear polarization. If $\tau_s$ is of the order of $\tau_e$ or larger, bleaching of absorption becomes spin sensitive and the saturation intensity of circularly polarized radiation drops below the value of linear polarization.

Spin sensitive bleaching can be analyzed in terms of excitation-relaxation kinetics taking into account both optical excitation and non-radiative relaxation processes. The probability rates for direct optical transitions from the $hh1$ states with $m = \pm 3/2$ to higher subbands are denoted as $W_{\pm}$. For linearly polarized light, $W_+$ and $W_-$ are equal. For the circular polarization, right handed, $\sigma_+$, or left handed, $\sigma_-$, the rates $W_{\pm}$ are different but, due to time inversion symmetry, satisfy the condition $W_+(\sigma_\pm) = W_- (\sigma_\mp)$. If $p_+$ and $p_-$ are the 2D densities of heavy holes with spin $+3/2$ and $-3/2$, respectively, then the rate equation for $p_+$ can be written as

$$\frac{\partial p_+}{\partial t} + \frac{p_+ - p_-}{2\tau_s} = -W_+ + \frac{1}{2}(W_+ + W_-).$$  \hspace{1cm} (1)

The corresponding equation for $p_-$ is obtained by exchange of indices $\pm \to \mp$. Since the laser pulse duration was longer than any expected relaxation time we consider the steady-state solution of the rate equations and omit the time derivative in Eq. (1). The second term on the left-hand side of Eq. (1) describes the spin relaxation trying to equalize the polarization of the $\pm 3/2$ states. The first term on the right-hand side describes the removal of holes from the $hh1$ subband due to photoexcitation while the second term characterizes the relaxation of holes which come down to the $+3/2$ and $-3/2$ states with equal rates (see insets in Fig. 3). The right side of the Eq. (1) is proportional to $W_+ - W_- = \frac{\alpha dI}{\hbar \omega}(\rho_0 P_{\text{circ}} - \eta \rho)$, where $\rho = (p_+ - p_-)/p_s$ is the hole spin polarization degree, $\rho_0$ is the excitation induced spin polarization, and $\eta \approx f_i/(f_i - f_f) \approx 1$ describes the difference between the population of the initial state, $f_i$, and the final state, $f_f$, respectively.

Bleaching of absorption with increasing intensity is described by the function $\alpha = \alpha_0[1 + (I/I_{se})]^{-1}$ where $\alpha_0$ is the absorption coefficient at low intensities and $I_{se}$ is the characteristic saturation intensity controlled by energy relaxation of the hole gas. Since the photocurrent $j_{LPGE}$ induced by the linearly polarized radiation is proportional to $\alpha I$, one has
\[
\frac{j_{LPGE}}{I} \propto \frac{1}{1 + \frac{1}{I_{se}}}.
\]  

(2)

The photocurrent \( j_{CPGE} \) induced by the circularly polarized radiation is proportional to \( W_+ - W_- \). Solving Eqs. (1,2) in the steady-state regime we obtain

\[
\frac{j_{CPGE}}{I} \propto \frac{1}{1 + I \left( \frac{1}{\tau_{se}} + \frac{1}{I_{ss}} \right)},
\]

(3)

where \( I_{ss} = \hbar \omega p_s / (\alpha_0 d \tau_s) \) is the saturation intensity controlled by the hole spin relaxation. The saturation intensities \( I_{ss} \) and \( I_{se} \) were extracted from the measured saturation intensities \( I_s \) of linear and circular photogalvanic current (Fig. 3). Using \( I_{ss} \) together with the absorption coefficient \( \alpha_0 \), calculated after [26] for the wavelength used here, spin relaxation times \( \tau_s \) have been derived. The results are plotted in Fig. 4 as a function of temperature. At low temperatures the relaxation times vary like \( T^{-\frac{1}{2}} \).

The magnitude of the observed hole spin relaxation time \( \tau_s \) in the first heavy hole subband \( hh1 \) is in very good agreement to previously published data obtained from the interband recombination kinetics of circularly polarized photoluminescence [14–16, 27–31]. These works, the relation between spin relaxation time and free carrier density has been discussed in terms of the D’yakonov-Perel and the Bir-Aronov-Pikus mechanisms. In our case of monopolar spin orientation photocreated carriers and electron-hole pairs do not exist. Thus, in contrast to all previous experiments were hole spin relaxation times were probed by interband excitation, the Bir-Aronov-Pikus mechanism is absent and the spin relaxation is not affected by high density photocreated carriers, exciton formation and interband recombination. The D’yakonov-Perel mechanism was investigated theoretically by Ferreira and Bastard [23, 32] for spin relaxation of \( hh1 \)-holes in GaAs based QWs. The values of \( \tau_s \) on the order of 10 ps as well as the observed temperature dependence \( \tau_s \propto T^{-1/2} \) are in accordance with these calculations for samples with parameters as in our experiment.

In conclusion, our experimental results demonstrate that absorption of terahertz radiation by inter valence band transitions in p-type QWs becomes spin sensitive at high power levels. The saturation of circularly polarized radiation, which is measured in QWs by the
circular photogalvanic effect, yields the spin relaxation times of majority carriers, in our case holes. Finally we would like to emphasize that spin sensitive bleaching is also expected for inter-subband transitions in n-type QWs and may be used to extract spin relaxation times of electrons in n-type materials.

We thank L. Golub for helpful discussions. Financial support by the DFG, the RFBR, the INTAS and the NATO linkage program is gratefully acknowledge.
REFERENCES

[1] S. Datta, B. Das, Appl. Phys. Lett. 56, 665 (1990).

[2] G. A. Prinz, Phys. Today 48, 58 (1995).

[3] B. E. Kane, Nature 393, 133 (1998).

[4] D. Loss, D. P. DiVincenzo, Phys. Rev. A 57, 120 (1998).

[5] D. P. DiVincenzo, D. Bacon, J. Kempe, G. Burkard, K. B. Whaley, Nature 408, 339 (2000).

[6] R. Fiederling et. al., Nature 402, 787 (1999).

[7] Y. Ohno et. al., Nature 402, 790 (1999).

[8] I. Malajovich, J.J. Berry, N. Samarth, and D.D. Awschalom, Nature 411, 770 (2001).

[9] Optical orientation, F. Meier, B. P. Zakharchenya, Eds. (Elsevier Science Publ., Amsterdam, 1984).

[10] S. D. Ganichev, S. N. Danilov, J. Eroms, W. Wegscheider, D. Weiss, W. Prettl, E. L. Ivchenko, Phys. Rev. Lett. 86, 4358 (2001).

[11] J. M. Kikkawa, I. P. Smorchkova, N. Samarth, D. D. Awschalom, Science 227, 1284 (1997).

[12] D. Hägele, M. Oestreicht, W. W. Rühle, N. Nestle, K. Eberl, Appl. Phys. Lett. 73, 1580 (1998).

[13] J. M. Kikkawa, D. D. Awschal, Nature 397, 139 (1999).

[14] J. Shah, “Ultrafast spectroscopy of semiconductor nanostructures”, Springer (1999), pp. 243-261.

[15] L.J. Sham, J. Phys.: Condens. Matter 5, A51 (1993).
[16] J. Fabian, and S. Das Sarma, J. Vac. Sci. Technol. B 17, 1708 (1999).

[17] E. V. Beregulin, S. D. Ganichev, I. D. Yaroshetskii, and I. N. Yassievich, Sov. Phys. Semicond. 16, 179 (1982).

[18] C. R. Pidgeon, A. Vass, G. R. Allan, W. Prettl, and L. Eaves, Phys. Rev. Lett. 50, 1309 (1983)

[19] E. V. Beregulin, S. D. Ganichev, K. Yu. Glukh, and I. D. Yaroshetskii, Sov. Phys. Semicond. 21, 615 (1987)

[20] M. Helm, T. Fromherz, B. N. Murdin, C. R. Pidgeon, K. K. Geerinck, N. J. Hovenyer, W. Th. Wenckebach, A. F. G. van der Meer, and P. W. van Amersfoort, Appl. Phys. Lett. 63, 3315 (1993)

[21] W. J. Li, B. D. McCombe, J. P. Kaminiski, S. J. Allen, M. I. Stockman, L. S. Muratov, L. N. Pandey, T. F. George, and W. J. Schaff, Semicond. Sci. Technol. 9, 630 (1994)

[22] S. D. Ganichev, H. Ketterl, W. Prettl, E. L. Ivchenko, L. E. Vorobjev, Appl. Phys. Lett. 77, 3146 (2000).

[23] The photogalvanic currents $j_x$ generated by the polarized radiation at normal incident has been measured as a function of intensity in unbiased samples via the voltage drop across a 50 $\Omega$ load resistor in a closed circuit configuration. For LPGE linearly polarized radiation with the electric field vector $E$ oriented at 45 degrees to the direction $x$ was used.

[24] E. L. Ivchenko, G. E. Pikus, Superlattices and Other Heterostructures. Symmetry and Optical Phenomena, (Springer, Berlin 1997).

[25] R. Ferreira, and G. Bastard, Phys. Rev. B. 43, 9687 (1991).

[26] L.E. Vorobjev, D.V. Donetskii, L.E. Golub, JETP Lett.63, 981 (1996)

[27] T.C. Damen, L. Vina, J.E. Cunningham, J. Shah, and L.J. Sham, Phys. Rev. Lett. 67,
3432 (1991).

[28] S. Bar-Ad and I. Bar-Joseph, Phys. Rev. Lett. 68, 349 (1992).

[29] B. Baylac, X. Marie, T. Amand, M. Brousseau, J. Barrau, and Y. Shekun, Surface Science 326, 161 (1995).

[30] S. Adachi, T. Mityasita, S. Takeyama, Y. Takagi, and A. Tackeuchi, J. Luminescence 72-74, 307 (1997).

[31] M.D. Martin, E. Perez, L. Vina, L. Gravier, M. Potemski, K. Ploog, and A. Fisher, Physica E 2, 186 (1998).

[32] G. Bastard, and R. Ferreira, Europh. Lett. 23, 439 (1993).
FIGURES

FIG. 1. Sketch of direct optical transitions (full line) between the first heavy hole and the first light hole subband in p-GaAs/AlGaAs QWs. While the splitting of the bands in $k$-space is necessary for an understanding of the circular photogalvanic effect [10] it is unimportant for the saturation process and ignored in the sketch. The absorption of far-infrared radiation with wavelength of 148 $\mu$m (photon energy $\hbar\omega = 8.3$ meV) occurs very close to $k = 0$. Therefore the initial and final states are characterized by angular momentum quantum numbers $m = \pm 3/2$ and $\pm 1/2$, respectively. Dashed lines shows the energy relaxation of photoexcited carriers. $\varepsilon_F$ is the Fermi energy.

FIG. 2. Photogalvanic current $j_x$ normalized by the intensity $I$ as a function of $I$ for circularly and linearly polarized radiation. Measurements are presented for $T = 20$ K. The inset shows the geometry of the experiment where $\hat{e}$ indicates the direction of the incoming light. The current $j_x$ flows along the [110]- direction at normal incidence of radiation on p-type (113)A- grown GaAs/AlGaAs QWs. In order to obtain the circular photogalvanic effect (CPGE) right or left circularly polarized light has been applied. To obtain the linear photogalvanic effect (LPGE) linearly polarized radiation with the electric field vector $E$ oriented at 45 degrees to the direction $x$ was used. The measurements are fitted to $j_x/I \propto 1/(I + I/I_s)$ with one parameter $I_s$ for each state of polarization (full line: circular, broken line: linear).

FIG. 3. Temperature dependence of the saturation intensity $I_s$ for linearly and circularly polarized radiation. The dependence is shown for one p-GaAs/AlGaAs (311)-MBE-grown sample with a single QW of $L_W=15$ nm width. The free carrier density is $1.66 \cdot 10^{11}$ cm$^{-2}$ and the mobility is $6.5 \times 10^5$ cm$^2$/(Vs). Insets show a microscopic picture explaining the origin of the difference in saturation intensities.
FIG. 4. Experimentally determined spin relaxation times $\tau_s$ of holes in $p$-type GaAs/AlGaAs QWs as a function of temperature. Open triangles and full dots corresponds to (113) MBE-grown 15 nm single and multiple (20) QWs, respectively. Free carrier densities of all samples were about $2 \cdot 10^{11}$ cm$^{-2}$ for each QW.
$j_x / l \propto \frac{1}{1 + l / l_s}$

$p$-GaAs/AlGaAs QW

circularly polarized radiation

linearly polarized radiation

$x \parallel [110]$
