



Hole spin-relaxation in quantum wells from saturation of inter-subband absorption

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Abstract

Spin-sensitive saturation of absorption of infrared radiation has been investigated in p-type GaAs QWs. It is shown that the absorption saturation of circularly polarized radiation is mostly controlled by the spin relaxation time of the holes. The saturation behavior has been investigated for different QW widths and in dependence on the temperature with the result that the saturation intensity substantially decreases with smaller QW width. Spin relaxation times were experimentally obtained by making use of calculated (linear) absorption coefficients for inter-subband transitions. The question of selection rules for intersubband transitions between hole subbands is addressed.

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1. Introduction

The investigation of spin relaxation has recently attracted considerable attention because of its great importance for the development of active spintronic devices [1]. Investigations of the spin lifetime in semiconductors were based so far on measurements of the polarized time-resolved photoluminescence after optical spin-orientation with interband excitation. These studies gave important insights into the mechanisms of spin relaxation of photoexcited free carriers.

Recently, the spin-sensitive bleaching of infrared absorption, observed in p-type QWs, provided an alternative access to spin relaxation times under the condition of monopolar spin orientation [2]. The advantage of this method is that only one type of charge carriers is involved, which rules out spin relaxation connected with electron–hole interaction and exciton formation [3]. Here we focus on transitions between hole subbands excited with infrared radiation.

2. Experiment

The experiments have been carried out on p-type (113) MBE-grown GaAs QWs with various well widths L_W between 7 and 20 nm and on (001)-miscut QWs grown by MOCVD with $L_W = 20$ nm. Samples

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with free carrier densities p_s of about $2 \times 10^{11} \text{ cm}^{-2}$ and very high mobility μ around $5 \times 10^5 \text{ cm}^2/(\text{Vs})$ (at 4.2 K) were studied in the range of $T = 4.2 \text{ K}$ up to 120 K. In this contribution we restrict our discussion on the sample with $L_W = 15 \text{ nm}$. As radiation source a high power far-infrared (FIR) molecular laser, optically pumped by a TEA-CO₂ laser, has been used delivering 100 ns pulses with intensities up to 1 MW/cm^2 in a wavelength range between 35 and $148 \mu\text{m}$.

Investigating the intensity dependence of the absorption coefficient shows a saturation of the absorption at high intensities. It is observed that for circularly polarized radiation, as compared to linearly polarized radiation, this takes place at a lower level of intensity. The difference in absorption bleaching for circularly and linearly polarized radiation has been observed [2] employing the circular (CPGE) [4] and the linear (LPGE) photogalvanic effect [5]. The absorption coefficient is proportional to the photogalvanic current j_x normalized by the radiation intensity I . Fig. 1a shows that j_x measured on p-type GaAs QWs depends on I as $j_x \propto I/(1 + I/I_s)$, where I_s is the saturation intensity. This behavior indicates saturation due to homogeneous broadening [6] (slow relaxation [7]) in contrast to saturation due to inhomogeneous broadening [8] (Rabi oscillation [7]). For different temperatures and QW widths our experiments show that saturation intensities I_s for circularly polarized radiation are generally smaller than for linearly polarized radiation (Fig. 1a).

The basic physics of spin sensitive bleaching of absorption is sketched in Fig. 2. Excitation with FIR radiation results in direct transitions between heavy-hole hh1 and light-hole lh1 subbands. This process depopulates and populates selectively spin states in hh1 and lh1 subbands, respectively. The absorption is proportional to the difference of populations of the initial and final states. At high intensities the absorption decreases since the photoexcitation rate becomes comparable to the non-radiative relaxation rate to the initial state. For C_s -symmetry, applying to our (113)-grown QWs, the selection rules for the absorption at \mathbf{k} close to zero (but $\neq 0$) are such that almost only (for details see Section 3) one type of spins is involved in the absorption of circularly polarized light. Thus the absorption bleaching of circularly polarized radiation is governed by energy relaxation of photoex-

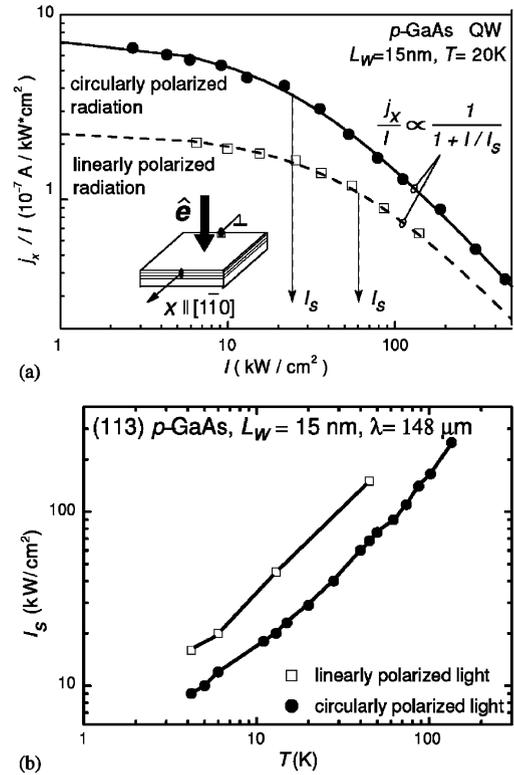


Fig. 1. (a) CPGE and LPGE currents j_x normalized by intensity I as a function of I for circularly and linearly polarized radiation of $\lambda = 148 \mu\text{m}$, respectively [2]. (b) Temperature dependence of the saturation intensities for $L_W = 15 \text{ nm}$ for linear (squares) and circular (circles) polarized light, respectively.

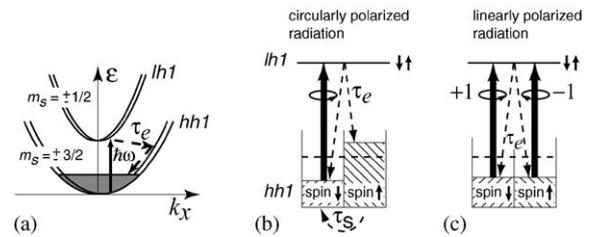


Fig. 2. Microscopic picture of spin-sensitive bleaching: (a) direct hh1–lh1 optical transitions. (b) and (c) process of bleaching for circularly and linearly polarized radiation. Dashed arrows indicate energy (τ_e) and spin (τ_s) relaxation.

cited carriers and spin relaxation in the initial subband (see Figs. 2a and b). These processes are characterized by energy and spin relaxation times τ_e and τ_s , respectively. We note, that during energy relaxation to the initial state in hh1 the holes lose their photoinduced orientation due to rapid relaxation [9]. Thus, spin

orientation occurs in the initial subband hh1, only. In contrast to circularly polarized light, absorption of linearly polarized light is not spin selective and the saturation is controlled by the energy relaxation only (see Fig. 2c). If τ_s is longer than τ_e , bleaching of absorption becomes spin sensitive and the saturation intensity I_s of circularly polarized radiation drops below the value of linear polarization (see Fig. 1a). The non-linear behavior of the photogalvanic current has been analyzed in terms of excitation-relaxation kinetics taking into account both optical excitation and non-radiative relaxation processes. It can be shown [2] that the photocurrent j_{LPGE} induced by linearly polarized radiation is described by $j_{\text{LPGE}}/I \propto (1 + I/I_{\text{se}})^{-1}$, where I_{se} is the saturation intensity controlled by energy relaxation of the hole gas. The photocurrent j_{CPGE} induced by circularly polarized radiation is proportional to $I/(1 + I(I_{\text{se}}^{-1} + I_{\text{ss}}^{-1}))$ where $I_{\text{ss}} = \hbar\omega p_s/(\alpha_0 L_W \tau_s)$ is the saturation intensity controlled by hole spin relaxation. Here α_0 is the absorption coefficient at low intensities and the spin relaxation time τ_s can be evaluated as

$$\tau_s = \frac{\hbar\omega p_s}{\alpha_0 L_W I_{\text{ss}}}. \quad (1)$$

In order to obtain τ_s the absolute value of α_0 is needed, which is determined theoretically.

The calculations of the linear absorption coefficient α_0 for inter-subband transitions are based on the self-consistent multi-band envelope function approximation (EFA) [10], that takes into account the crystallographic orientation of the QW (here the (1 1 3) direction) and the doping profile. Calculations are performed here within the Luttinger model of the heavy and light hole states to obtain the hole subband dispersion $\varepsilon_i(\mathbf{k})$ and eigenstates $|i, \mathbf{k}\rangle$ of the hole subband i and in-plane wave-vector \mathbf{k} . For direct (electric dipole) transitions between subbands i and j the contribution to the absorption coefficient $\alpha_{i \rightarrow j}(\omega)$ as a function of the excitation energy $\hbar\omega$ is then given by [11]

$$\alpha_{i \rightarrow j}(\omega) = \frac{e^2}{4\pi\epsilon_0\omega cn L_W} \int d^2k | \langle j, \mathbf{k} | \mathbf{e} \cdot \hat{\mathbf{v}}(\mathbf{k}) | i, \mathbf{k} \rangle |^2 \times [f_j(\mathbf{k}) - f_i(\mathbf{k})] \frac{e^{-(\varepsilon_j(\mathbf{k}) - \varepsilon_i(\mathbf{k}) - \hbar\omega)^2 / \Gamma^2}}{\sqrt{\pi}\Gamma}, \quad (2)$$

where \mathbf{e} is the light polarization vector, n is the refractive index, ϵ_0 is the free-space permittivity, $f_i(\mathbf{k})$ is the Fermi distribution function in the subband i and Γ is a broadening parameter to account for the level

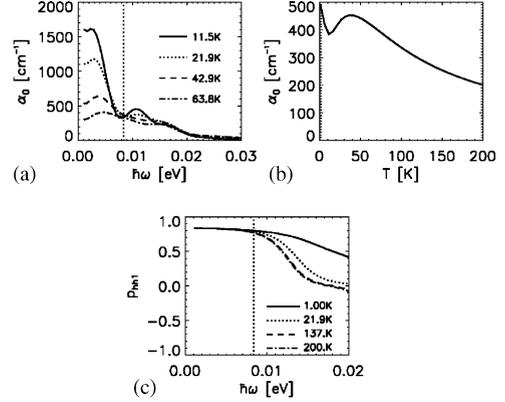


Fig. 3. (a) Absorption coefficient as a function of photon energy $\hbar\omega$ for various temperatures and (b) as a function of T for $\hbar\omega = 8.4$ meV (vertical dotted line in (a)), calculated for a (1 1 3)-grown 15 nm GaAs–AlGaAs QW with carrier density $2 \times 10^{11} \text{ cm}^{-2}$ and the broadening Γ was set to 2.47 meV. (c) Hole spin orientation efficiency p_{hh1} (for definition see Section 3) for the hh1 subband as a function of excitation energy for different temperatures.

broadening due to scattering. Within EFA, the velocity $\hat{\mathbf{v}}(\mathbf{k})$ is a matrix operator expressed as the gradient in \mathbf{k} -space of the Luttinger Hamiltonian. Its matrix elements are calculated from the EFA wave functions.

Following this scheme we have calculated the absorption coefficient $\alpha_0(\omega) = \sum_{ij} \alpha_{i \rightarrow j}(\omega)$. The absorption spectrum for the system with $L_W = 15$ nm is shown in Fig. 3a. At low temperatures two pronounced peaks evolve, which correspond to the transitions from the lowest (spin split) hole subband to the second and third subband, respectively. Fig. 3b shows the temperature dependence (due to the Fermi distribution function) of α_0 at the excitation energy $\hbar\omega = 8.4$ meV for the sample with $L_W = 15$ nm.

Using the calculated absorption coefficients α_0 , the experimental saturation intensities I_{ss} are converted with Eq. (1) into spin relaxation times. The results for QWs of $L_W = 15$ nm are shown in Fig. 4. Compared to the values given in Ref. [2], where α_0 was derived from Ref. [11], here we obtain smaller τ_s at high temperatures due to a more realistic theoretical model for the calculation of α_0 .

3. Selection rules and spin orientation

We note that in the definition of I_{ss} it was assumed that the spin selection rules are fully satisfied at the

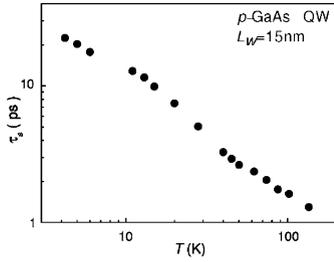


Fig. 4. Spin relaxation times obtained for a p-type GaAs sample with a QW width of $L_W = 15$ nm, $p_s = 1.66 \times 10^{11}$ cm $^{-2}$ and mobility μ of about 5×10^5 cm 2 /(Vs).

transition energy. However, in (1 1 3)-grown systems, hh1 and lh1 subbands are mixed due to the low symmetry even at $\mathbf{k} = 0$. This may reduce the strength of the selection rules [12] and, therefore, the efficiency of spin orientation. The lowest subband, which for (0 0 1) grown systems is purely heavy hole ($m_s = \pm \frac{3}{2}$) at $\mathbf{k} = 0$, has for growth direction (1 1 3) an admixture of about 10% light hole spinor components ($m_s = \pm \frac{1}{2}$) [13]. This admixture is sufficiently small to justify labeling the subbands according to the dominant spinor component at $\mathbf{k} = 0$.

Strict selection rules for intersubband transitions between hole subbands only exist in some idealized limits (e.g. spherical approximation for the Luttinger Hamiltonian or growth directions of high symmetry and $\mathbf{k} = 0$). However, assuming a symmetrically doped (1 1 3) grown QW, the lowest hh and lh subband states (hh1 and lh1, respectively) have even parity at $\mathbf{k} = 0$ and no transition between hh1 and lh1 is possible, as the velocity operator projected on the light polarization direction $\hat{\mathbf{v}} \cdot \mathbf{e}$ couples only states of different parity. Therefore a strictly valid selection rule cannot be obtained and a more quantitative discussion of the relative weight of the possible transitions is necessary. For \mathbf{k} small enough to ensure that the admixture of odd parity spinor components is negligible, only contributions in $\hat{\mathbf{v}} \cdot \mathbf{e}$ linear in \mathbf{k} are to be considered.

A more detailed analysis gives the following results: The spin-conserving transitions $\text{hh1}\uparrow \rightarrow \text{lh1}\uparrow$ and $\text{hh1}\downarrow \rightarrow \text{lh1}\downarrow$ are much weaker than the corresponding spin-flip transitions $\text{hh1}\uparrow \rightarrow \text{lh1}\downarrow$ and $\text{hh1}\downarrow \rightarrow \text{lh1}\uparrow$. Depending on the left/right circular polarization of the exciting light, one of the spin-flip transitions is dominant. To investigate the hole spin ori-

entation, we also performed a numerical calculation of $\alpha_{i \rightarrow j}$ for excitation with right-hand circularly polarized light. We obtained that the transition $\text{hh1}\downarrow \rightarrow \text{lh1}\uparrow$ is far more probable than all other transitions. This is quantitatively described by the heavy hole spin polarization efficiency

$$p_{\text{hh1}} = \frac{\sum_i \alpha_{\text{hh1}\downarrow \rightarrow i} - \alpha_{\text{hh1}\uparrow \rightarrow i}}{\sum_i \alpha_{\text{hh1}\downarrow \rightarrow i} + \alpha_{\text{hh1}\uparrow \rightarrow i}}, \quad (3)$$

where the summation is performed over all subbands. If p_{hh1} is +1 (−1) the excitation leaves only heavy holes belonging to the up (down) branch of the dispersion in the hh1 subband. In our case, p_{hh1} is around 80% at the laser excitation energy and almost independent of the temperature (Fig. 3c). Therefore we may neglect effects due to incomplete spin orientation, as assumed in this contribution.

The future extraction of spin relaxation times for all investigated QW widths will enable us to discuss the well width dependence of spin relaxation times.

Acknowledgements

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