Spin relaxation in type-II GaAs/AlAs quantum wells

W. A. J. A. van der Poel, A. L. G. J. Severens, and H. W. van Kesteren
Philips Research Laboratories, Postbox 80 000, NL-5600 JA Eindhoven, The Netherlands

C. T. Foxon
Philips Research Laboratories, Cross Oak Lane, Redhill, Surrey RH1 5HA, England
(Received 21 November 1988)

We studied the spin relaxation for a type-II GaAs/AlAs quantum well, using time-resolved luminescence. The sample is resonantly excited at the exciton transition with circularly polarized light, and detection is on one of the two circularly polarized components in the direction perpendicular to the layers. The temperature is normally 1.7 K. No polarization of the luminescence is observed at zero magnetic field. In a small longitudinal magnetic field (100 mT) the luminescence becomes polarized. The polarization ratio stays the same during the decay of the luminescence. This means a spin-relaxation time longer than the lifetime of 7 μs. When the temperature is raised to 9 K, we observe a spin-relaxation time of 0.5 ± 0.1 μs. The magnetic field behavior is explained by the quenching of a fast dephasing process which occurs at zero field. The long spin-relaxation time for the type-II quantum well contrasts with the times for type-I quantum wells. This difference could be due to the difference in the exchange interaction.

In optical spectroscopy studies of GaAs/$Ga_{1-x}Al_xAs$ quantum wells (QW's), polarization is used to identify the various transitions.[1,2] The sample is excited with circularly polarized light and the polarization P of the luminescence is recorded. The polarization P is defined as $P=(I_+-I_-)/(I_++I_-)$ where $I_\pm$ is the $\sigma_\pm$ circular-polarization intensity. The magnitude and sign of P depend on the particular transition (see Fig. 1) and the magnitude of the spin-relaxation time compared to the lifetime. In excitation spectroscopy, the detection is fixed on the heavy-hole exciton emission and one scans the excitation energy. Transitions involving heavy or light holes are expected to be distinguishable by a change in the polarization.

Although spin orientation is thus being used as a tool to identify transitions in quantum wells, relatively little is known about the spin relaxation in QW's. We have started investigations by means of time-resolved methods to get more insight into the spin-relaxation times and mechanisms of excitons in QW's and present here the results obtained for a type-II QW. In type-II QW's (Refs. 3–5), the lowest conduction-band Γ-confined state in the GaAs has a higher energy than the lowest X-confined state in the AlAs barrier. The lowest-energy recombination is therefore between the holes in the well and the electrons in the barrier (see Fig. 2). This results in a small oscillator strength and in a long radiative lifetime, e.g., the decay constant for the sample which we have used amounts to 7 μs.[6] Recently, optically detected magnetic resonance (ODMR) has been observed in similar structures by van Kesteren *et al.*[7,8] To explain the sign and intensity of the observed signals they had to assume a nonthermal population distribution. This would imply a spin-relaxation time comparable to the lifetime, e.g., in the microsecond time domain. This is in contrast with results obtained for bulk GaAs (Ref. 9) and type-I QW's (Ref. 1) where spin-relaxation times of several hundred picoseconds are reported.

The sample used in this study was grown by molecular-beam epitaxy in a Varian Associates Gen II system. For more details, see Ref. 10. The layers were deposited on a (001)-oriented semi-insulating GaAs substrate in the following sequence: (a) 1.0 μm GaAs (buffer); (b) 60 periods of 22 Å GaAs and 41 Å AlAs; and (c) 1000 Å GaAs (capping layer). Luminescence and lifetime data are reported in Refs. 5 and 6. In our experiment, the sample, placed in a variable-temperature cryostat, was excited with circularly polarized laser light. The angle between the normal of the sample and the ex-

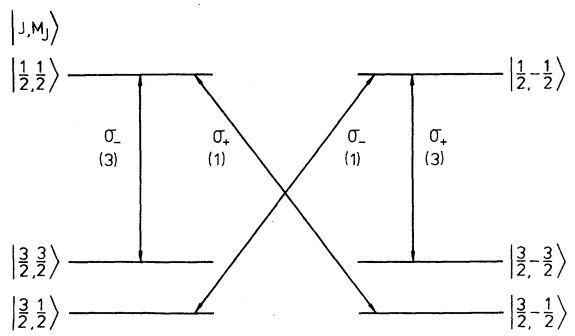

FIG. 1. Transition moments for circularly polarized light in a GaAs QW. The relative oscillator strengths are indicated in parentheses. The $|\frac{3}{2},\pm\frac{3}{2}\rangle$ heavy holes have a different energy than the $|\frac{3}{2},\pm\frac{1}{2}\rangle$ light holes. The lowest-energy transition involves creation of an exciton build-up from an electron and a heavy hole. There are four heavy-hole exciton states, two radiative, $|\frac{1}{2},+\frac{1}{2}\rangle|\frac{3}{2},+\frac{3}{2}\rangle$ and $|\frac{1}{2},-\frac{1}{2}\rangle|\frac{3}{2},-\frac{3}{2}\rangle$, and two nonradiative, $|\frac{1}{2},+\frac{1}{2}\rangle|\frac{3}{2},-\frac{3}{2}\rangle$ and $|\frac{1}{2},-\frac{1}{2}\rangle|\frac{3}{2},+\frac{3}{2}\rangle$.

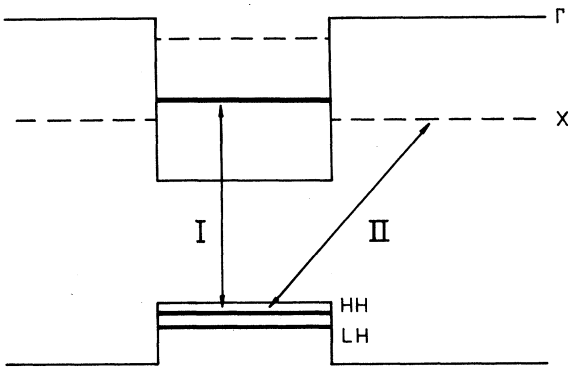

FIG. 2. Valence- and conduction-band structure of a GaAs/$Al_xGa_{1-x}As$ QW. The type-I and -II heavy-hole–electron transitions are indicated by arrows.

TABLE I. Polarization P at the saturation field of 100 mT for various excitation energies together with the position of the type-I transitions.

E_{exc} (eV)	P_{sat}
1.799 (Resonant type-II)	0.68
1.833	0.24
1.916	0.25
1.959	0.15
2.281	0.01
E (eV)	**Assignment**
1.95	E1Γ-HH1
2.01	E1Γ-LH1

citing beam was approximately 10°. Several excitation laser sources were used: a synchronously pumped cavity-dumped dye laser for resonant excitation in the type-II heavy-hole exciton transition, a mode-locked Kr^+ laser at 1.833 or 1.916 eV, and a cw He-Ne laser at 1.959 or 2.281 eV. The luminescence was collected almost along the normal to the layers. A quarter-wave plate followed by a linear polarizer was used to select one of the two circularly polarized components of the luminescence, which was further analyzed by means of a 30-cm monochromator equipped with a cooled photomultiplier with a GaAs cathode. Photoluminescence decays were measured via single-photon counting techniques. A magnetic field of 0–200 mT could be applied parallel to the luminescence direction.

In a QW, the existence of well-width fluctuations gives a broadening of the absorption line, and excitons excited with a specific energy corresponding to a certain well width can diffuse to a part of the sample where the well is wider. This means an energy relaxation of the exciton and results in an emission line shifted down in energy with respect to the lowest-energy absorption line. In our sample, due to this localization of the exciton, the difference between resonant heavy-hole exciton excitation and detection energy has to be 7–8 meV. In zero magnetic field and a temperature of 1.7 K under the above conditions, no polarization, either time integrated or time resolved (details will follow), is observed. When we apply a magnetic field, the luminescence becomes polarized and it saturates at a field of about 50 mT. A plot of the polarization P of the time-integrated luminescence as a function of the magnetic field is given in Fig. 3. If we use the other circular polarization in excitation, the intensities of the two circular polarizations of the luminescence are interchanged. The polarization ratio was constant over the luminescence line. Table I shows the time-integrated polarization ratios in the saturation magnetic field for the various excitation energies. For reference, the energy positions of the type-I transitions as reported in Ref. 5 are also shown. From this table it follows that, as the energy increases, the emission becomes less polarized, but there is still an appreciable polarization left if the excitation is via the type-I transition. The polarization disappears when the exciting laser has an energy which is far in the continuum of the type-I transition. With 1.959-eV excitation, the polarization as a function of the magnetic field has a much better signal to noise, and is similar, but scaled down, to that with resonant excitation. From this experiment we determine the magnetic field where the polarization is half the saturation value to be 18±3 mT.

In the time-resolved experiments, the time resolution is about 25 ns. This time is set by the fact that, just after the laser flash, the intensity of scattered laser light, from

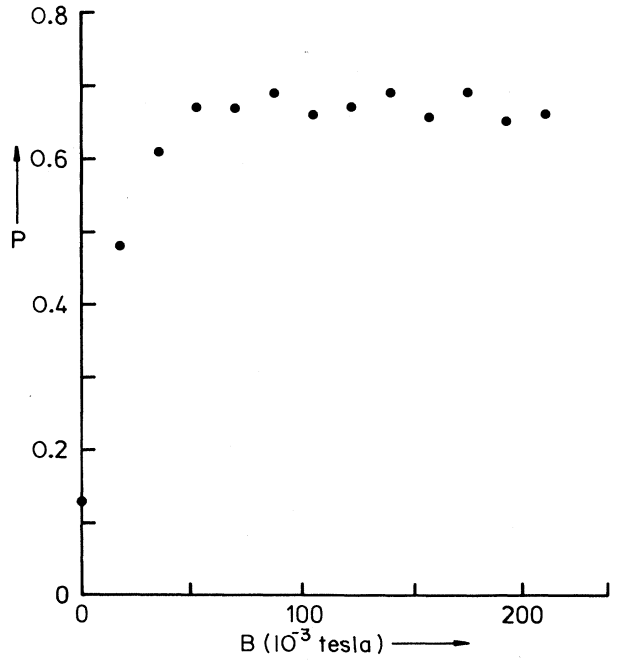

FIG. 3. Polarization P of the time-integrated luminescence at 1.7 K while resonantly exciting, at 1.799 eV, on the type-II heavy-hole exciton transition, as a function of the magnetic field. The detection was at 1.792 eV, which is the top of the emission line.

the main pulse and the cavity dumper leakage pulses, exceeds the luminescence emission. In zero magnetic field for times >25 ns after the laser flash, the luminescence under the above conditions has a polarization of less than 10%. When a magnetic field of 100 mT is applied, the time-resolved experiments show that at this field the ratio between the σ_+ and the σ_- luminescence intensity stays constant during the decay of the luminescence (see Fig. 4). From the data in Fig. 4, we determine the spin-relaxation time to be $>20\ \mu$s.

We also investigated the temperature dependence of the spin relaxation. This was done with resonant excitation on the type-II transition. The time-integrated luminescence again still shows no polarization in zero magnetic field, but at the saturation field it decreases when the temperature is raised. Figure 5 shows the decays obtained, for the two circular polarizations, at a temperature of 9 K and in a field of 100 mT. Decays like those of Fig. 5 show that there are two contributory factors: (1) the initial polarization just after the laser flash is reduced, and (2) the spin-relaxation time becomes shorter than the lifetime. From the data of Fig. 5 we inferred that at 9 K, the initial polarization disappears with a time constant of $0.5\pm0.1\ \mu$s.

Of the above observations, the most striking result is an apparent fast spin relaxation at zero magnetic field which disappears, provided the temperature is low enough, once a sufficient magnetic field is applied. Under our resonant excitation conditions, only heavy-hole excitons are created. If there were no spin relaxation present, a polarization of 100% would result, when using circularly polarized light. In contrast, no polarization is observed in zero magnetic field. The reason for this behavior lies in the fact that, at zero magnetic field, a small interaction exists which mixes the σ_+ and σ_- emitting states.[11,12] The nature of this interaction will be discussed further elsewhere. At this moment, the exact nature is not relevant; the only fact of importance is the mixing. Due to the mixing, the circular polarization will

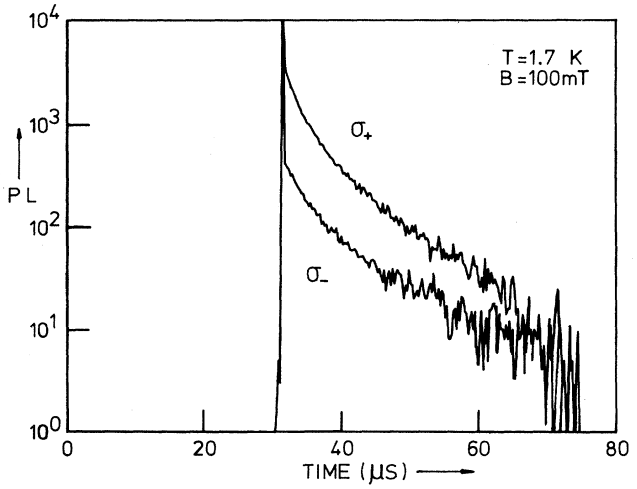

FIG. 4. Photoluminescence decays for left-handed and right-handed circularly polarized light obtained in a magnetic field of 100 mT. The other experimental conditions are the same as in Fig. 3.

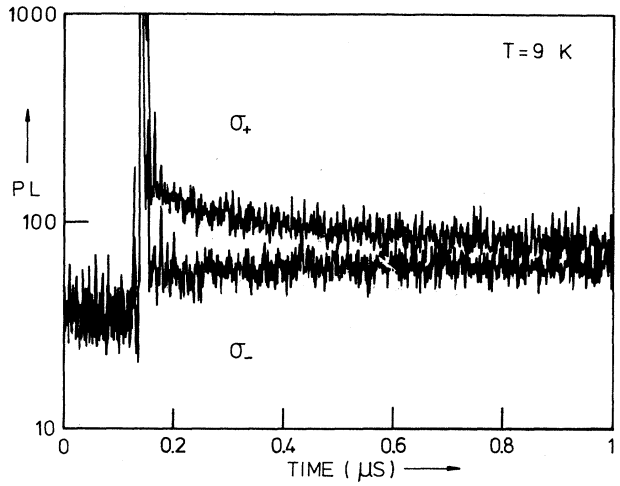

FIG. 5. Decays at 9 K and a field of 100 mT for the two circularly polarized components of the luminescence. For the excitation and detection energies see Fig. 3.

disappear by a dephasing process, which occurs on a fast time scale. This time is given by the smaller of T_2^* or $h/\Delta E$, where ΔE is the zero-field energy splitting between the radiative states and T_2^* is the inhomogeneous lifetime. The longitudinal magnetic field will reduce the mixing, and at sufficient high fields the states are pure σ_+ and σ_- emitting states. Provided the dephasing process is fast compared to the lifetime and/or the spin-relaxation time, the steady-state polarization is given by

$$P(B)=\frac{P_0}{1+\left[\dfrac{\Delta E}{g\mu_B B}\right]^2}\frac{\tau_s}{\tau_s+\tau}\ , \tag{1}$$

where P_0 is the polarization just after the localization, $g\mu_B B$ is the Zeeman splitting energy, τ_s is the spin-relaxation time, and τ is the lifetime. The first factor of the equation gives the polarization left after the dephasing, while the second part reflects the competition between the normal decay and the spin relaxation, and only holds for exponential decay and relaxation processes. At sufficiently high magnetic fields, the dephasing is no longer of importance and the first part becomes constant. The decays taken at 100 mT, therefore, allow us to determine τ_s. At 1.7 K, we observe that part of the polarization is lost during the preparation of the exciton in its lowest-energy state, and after this energy relaxation there is no observable spin relaxation.

Assuming P_0, τ, and τ_s to be independent of the magnetic field, we expect from (1) that the field $B_{1/2}$ where the polarization is half the polarization at the saturation field to be given by $B_{1/2}=\Delta E/|g\mu_B|$. For ΔE we have found a value of $0.34\pm0.04\ \mu$eV.[11] Using this value we get $g=\pm(0.33\pm0.06)$. The effective g factor is built up from contributions from the electron and the hole, and it is given, for a longitudinal magnetic field, by $g=g_{ez}+6(K+\frac{9}{4}q)$.[7] In this expression, g_{ez} is the g factor for the electrons and K and q are the Luttinger parameters for the Zeeman energy splitting of the holes.

The value of g_{ez} has been determined by ODMR (Ref. 8) and amounts to 1.89. As usual we will take $q=0$. The fact that the sign of g is undetermined gives us two solutions: $K=-0.26\pm0.01$ or $K=-0.37\pm0.01$. Since both values are in agreement with the value reported from ODMR,[7] we cannot decide which value to use. What is clear is that the electron g value and the hole K value must have opposite signs.

If we want to compare the result obtained for the type-II QW with those for the type-I QW's, we are left with the question of whether or not the mixing of the σ_+ and σ_- states also plays a role in the type-I QW's. In the type-II QW's investigated up to now, this interaction scales with the (normal) exchange interaction which splits the dark and radiative states of the exciton.[11] Compared to a type-I QW, the exchange interaction is reduced in a type-II QW due to the smaller overlap between the electron and the hole wave functions. The exchange splitting between the dark and radiative states for the structure we have used amounts to 0.7 μeV.[8] In the bulk it is estimated at 50 μeV (Ref. 13) and we expect the same order of magnitude or bigger for a type-I QW. If the splitting between the radiative states indeed scales with this exchange, the dephasing occurs on a time scale of 20–80 ps. Secondly, a large magnetic field is then needed to unmix the states. This is in qualitative agreement with our experiments on a 50-Å type-I QW (Ref. 14) where a fast, 50–60-ps spin relaxation, which was not influenced by a magnetic field up to 200 mT, was found. Maan *et al.*[15] concluded from steady-state experiments in fields >10 T, on a type-I QW, that at these high fields the spin relaxation is slow compared to the lifetime. Whether this is due to the quantization into Landau levels or the above-discussed unmixing is not clear. Further experiments are needed to settle this.

In bulk GaAs no hole polarization is observed.[9,16] This means that the holes have a high spin-flip rate and, because both of the constituent particles of the exciton have to change their spin to see the other polarized component, the spin relaxation is governed by the electron spin-flip rate. The experiments on the type-I transition can be used to determine if the same also holds in a type-II QW. If it is assumed that the holes have a high spin-flip rate, one expects no polarization upon excitation in the type-I transition because, in this case, the electrons are created in the Γ point in the well and have to relax to the X point in the barrier and it is very likely that they change their spin in that process. Experimentally, however, an appreciable polarization is observed. This means that, unlike the situation in the bulk, hole polarization might exist in type-II QW's. This difference might be due to the fact that, in contrast to the QW, the light and heavy holes in the bulk have the same energy at $k=0$. Lifting this degeneracy by stress indeed resulted in a longer spin-relaxation time in bulk GaAs.[9]

In conclusion, we observed at 1.7 K, in comparison to the radiative lifetime of 7 μs, a long spin-relaxation time for the excitons in a type-II QW. Raising the temperature reduces the spin-relaxation time, and at 9 K it becomes shorter than the lifetime.

The authors are indebted to G. W. 't Hooft for many useful discussions on the subject of this paper.

[1]R. C. Miller, D. A. Kleinman, W. A. Nordland, Jr., and A. C. Gossard, Phys. Rev. B **22**, 863 (1980).

[2]R. C. Miller and D. A. Kleinman, J. Lumin. **30**, 520 (1985).

[3]P. Dawson, B. A. Wilson, C. W. Tu, and R. C. Miller, Appl. Phys. Lett. **48**, 541 (1986).

[4]E. Finkman, M. D. Sturge, and M. Tamargo, Appl. Phys. Lett. **49**, 1299 (1986).

[5]K. J. Moore, P. Dawson, and C. T. Foxon, Phys. Rev. B **38**, 3368 (1988).

[6]P. Dawson, K. J. Moore, C. T. Foxon, G. W. 't Hooft, B. A. Wilson, and C. E. Bonner, in *Quantum Well and Superlattice Physics II, Newport Beach, 1988,* Vol. 943 of SPIE Proceedings, edited by F. Capasso, G. H. Döhler, and J. N. Schulman (SPIE, Bellingham, WA, 1988), p. 145.

[7]H. W. van Kesteren, E. C. Cosman, F. J. A. M. Greidanus, P. Dawson, K. J. Moore, and C. T. Foxon, Phys. Rev. Lett. **61**, 129 (1988).

[8]H. W. van Kesteren, E. C. Cosman, P. Dawson, K. J. Moore, and C. T. Foxon, in Proceedings of the 19th International Conference on the Physics of Semiconductors, Warsaw, 1988 (unpublished).

[9]A. N. Titkov, V. I. Safarov, and G. Lampel, in *Proceedings of the 14th International Conference on the Physics of Semiconductors, Edinburgh, 1979,* edited by B. L. H. Wilson (The Institute of Physics, Bristol, 1979), p. 1031.

[10]C. T. Foxon and J. J. Harris, Philips J. Res. **41**, 313 (1986).

[11]W. A. J. A. van der Poel, A. L. G. J. Severens, and C. T. Foxon (unpublished).

[12]H. W. van Kesteren and W. A. J. A. van der Poel (unpublished).

[13]D. D. Sell, S. E. Stokowski, R. Dingle, and J. Dilorenzo, Phys. Rev. B **7**, 4568 (1973).

[14]W. A. J. A. van der Poel, A. L. G. J. Severens, H. W. van Kesteren, and C. T. Foxon, Superlatt. Microstruc. (to be published).

[15]J. C. Maan, M. Potemski, and Y. Y. Wang, in *Proceedings of the Application of High Magnetic Fields in Semiconductor Physics, Würzburg, 1988,* edited by G. Landwehr (Springer, Berlin, 1989).

[16]*Optical Orientation,* edited by F. Meier and B. P. Zakharchenya (North-Holland, Amsterdam, 1984).