Pergamon

Solid State Communications, Vol. 93, No. 1, pp. 57-60,1995

0038-1098/95 $9.50+.00

0038-1098(94)00721-7

HOLE SPIN RELAXATION IN n-MODULATION DOPED QUANTUM WELLS

B. Baylac, T. Amand, X. Marie, B. Dareys, M. Brousseau, G. Bacquet
Laboratoire de Physique des Solides, CNRS-URA 74
INSA, Avenue de Rangueil, 31077 Toulouse cedex, France

V. Thierry-Mieg
Laboratoire de Microstructures et Microélectroniques L2M-CNRS
BP107, 92225 Bagneux cedex, France

(Received 26 June 1994 by P.Burlet)

The hole spin relaxation in n-modulation doped quantum wells has been investigated as a function of the laser excitation energy and excitation intensity. We demonstrate for the first time that the heavy hole spin flip time is a very strong decreasing function of these two parameters. We present a simple interpretation which assigns these dependences to the carriers temperature variation.

Keywords : A. quantum wells A. Semiconductors D. spin dynamics

In spite of an intense research effort, spin relaxation in Quantum Wells (QW) is still poorly understood. The quality of samples is important since spin dynamics can be strongly influenced by localization or defects. On a more fundamental level, the differences in the behaviour of electrons, holes and excitons must be identified and carefully investigated [1]. The prediction of slower hole spin relaxation in QW compared to bulk semiconductors begins to be well-recognized [2,3]. Time-resolved optical pumping experiments performed on n-modulation doped QW provide in principle the direct measurement of the hole spin relaxation time τ_h. Such experiments have been performed by T.C. Damen et al. [1] and Ph. Roussignol et al. [4] but their results are very different : the first authors find $\tau_h \simeq 4ps$ and the second ones $\tau_h \simeq 1\ ns$.

In this work we present the results of time-resolved optical pumping measurements in n-modulation doped $GaAs/Al_{0.3}Ga_{0.7}As$ Multiple Quantum Wells (MQW). The results presented here concern a MQW sample which has 60 periods of 4.8 nm wells separated by 15 nm barriers with a central Si δ-doped layer. We have measured, by Hall experiments performed at 4.2 K, the electron density transferred from the doping layer ; we find : $n_e \simeq 6.10^{11}\ cm^{-2}$ (very similar results have been obtained on a $GaAs/Al_{0.3}Ga_{0.7}As$ MQW sample which differs by the doping level, $n_e \simeq 3.10^{11}\ cm^{-2}$). The sample, at 1.7 K, is excited by 1.8 ps pulses generated by a tunable Ti-doped sapphire laser at a repetition rate of 76 Mhz. The photocreated carriers density is varied from $n = 7.10^7\ cm^{-2}$ to $n = 2.10^{10}\ cm^{-2}$. The photoluminescence signal (PL) is detected by the up-conversion technique using a $LiIO_3$ non-linear crystal ; the overall time-resolution is thus limited by the laser temporal width. The excitation light is right circularly polarized using a Soleil-Babinet compensator. To measure the luminescence components of same (I_+) and opposite (I_-) helicity as the excitation light, a rotating $\lambda/4$ plate is placed before the non-linear crystal, which acts as an analyser.

Figure 1 shows the cw photoluminescence excitations I_+, I_- and the polarization

$$P = \frac{I_+ - I_-}{I_+ + I_-}$$

as a function of the excitation energy. The PL signal, also reported, corresponds to band-to-band recombination of photoexcited holes with majority electrons. We have checked that the shift between the PL line and the first peak in the PLE spectrum is in good agreement with the Fermi level determined from the measured electron density n_e. The polarization P measured at the peak of the PL spectrum (carriers near $k_{//} = 0$), is large for excitation close to the chemical potential E_F and drops to a constant low value ($\simeq 0.1$) at the electron-light hole excitation energy ($E1^* - LH1^*$) at 1.772 eV.

In figure 2, the cw polarization versus the excitation energy has been measured for two different photocreated carriers densities, namely $n = 8.10^7\ cm^{-2}$ (bold line) and $n = 6.10^8\ cm^{-2}$ (thin line). We notice a lowering of the polarization degree when the photocreated carriers density increases. We shall now

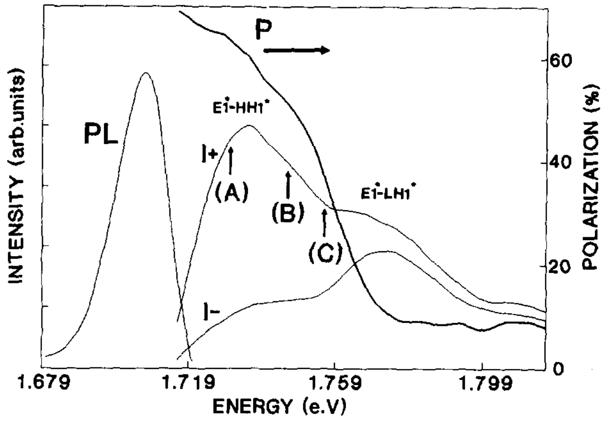

Figure 1.
cw PL , PLE, and the deduced polarization (P) spectra.

analyze this effect more thoroughly in time-resolved experiments.

Figure 3 shows the time evolutions of I_+, I_- and P at the peak of the PL spectrum, for excitation at B (defined in figure 1), and for an intermediate photogenerated carriers density of $n \simeq 2.10^9\ cm^{-2}$. The polarization decays non mono-exponentially with an initial decay time of about 40 ps. Figure 4.a shows the time evolution of the polarization P, recorded at the peak of the PL spectrum, for excitation at B also, but for three very different photogenerated carriers densities of $n \simeq 7.10^7\ cm^{-2}$, $2.10^9\ cm^{-2}$ and $2.10^{10}\ cm^{-2}$ respectively. The important result is a dramatic decrease of the decay time from 1.2 ns to 6 ps as the excitation power density increases. We observe immediately that this result could conciliate the opposed conclusions of Ph. Roussignol *et al.* ($\tau_h \simeq 1\ ns$) and T.C. Damen *et al.* ($\tau_h \simeq 4\ ps$) if their excitation densities were very different. However the first authors, who detect with a

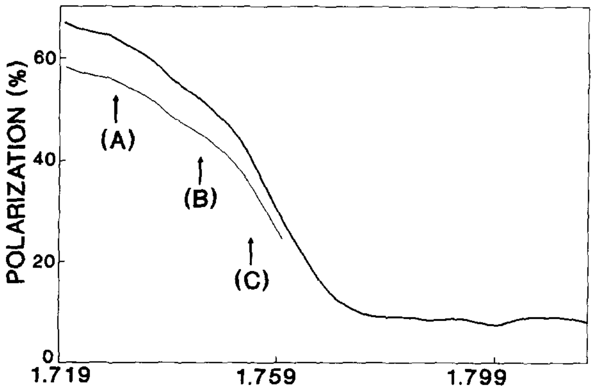

Figure 2.
cw polarization versus the excitation energy at the excitation density $n = 8.10^7\ cm^{-2}$ (bold line) and $n = 6.10^8\ cm^{-2}$ (thin line).

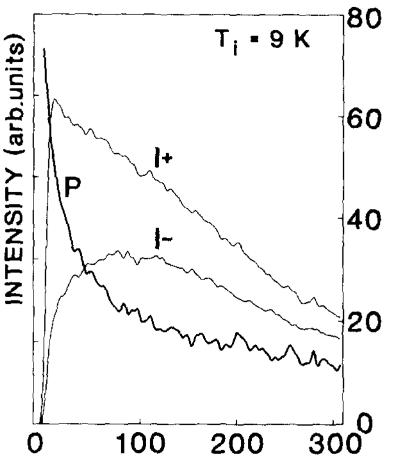

Figure 3.
Time evolutions of I_+, I_- and P for excitation at B (the excitation density is $n = 2.10^9\ cm^{-2}$). T_i denotes the *calculated* initial temperature of the electron-hole gas (see text).

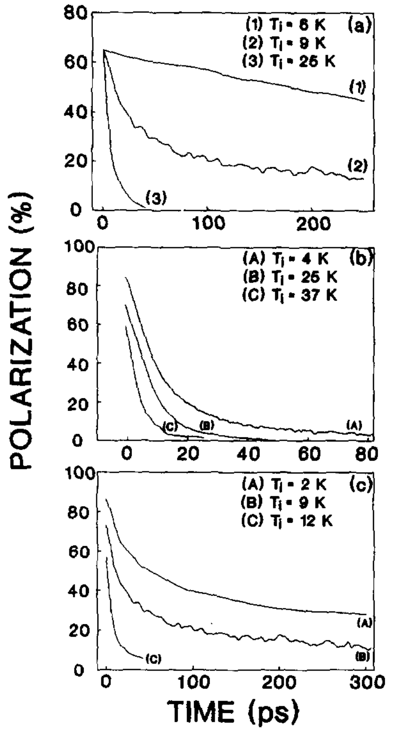

Figure 4.
(a) Time evolution of the polarization P at the excitation energy B for three excitation densities corresponding to (1) $7.10^7\ cm^{-2}$, (2) $2.10^9 cm^{-2}$, (3) $2.10^{10}\ cm^{-2}$
(b) time evolution of the polarization P for a photogenerated density $n = 2.10^{10}\ cm^{-2}$; the three excitation energies A, B and C of figure 1 are considered.
(c) idem (b) for $n = 2.10^9\ cm^{-2}$.
The *calculated* initial temperature T_i is also mentioned.

synchroscan streak camera, announce $n \simeq 10^{10}$ cm^{-2} with a sample at $T = 4$ K, while the second ones, who detect with an up-conversion experiment, claim $n \simeq 2.10^{10}$ cm^{-2} with a sample mounted in a cold finger cryostat at $T = 10$ K. We mention that Roussignol *et al.* explain such dispersion of the measured τ_h values by a D'Yakonov-Perel like mechanism [5], strongly dependent on the sample quality.

Figure 4.b and 4.c show the time evolution of the polarization P, at the peak of the PL spectrum, at high and low excitation intensities respectively, for the three excitation energies A, B, C quoted in figure 1. For these energies, the electrons are photo-created approximatively at the Fermi level (A), 10 meV above (B) and 19 meV above (C), evaluations which consider an in-plane heavy hole effective mass $m_{hh} = 0.17$. Two observations can be made :

(i) the initial polarization P(0) decreases slightly as the excitation energy increases ; this decrease is strictly independent of the excitation intensity, as the comparison between the two figures shows (this is also in agreement with the results displayed in figure 4.a) ;

(ii) the main depolarization occurs during the subsequent transient decrease ;

(iii) the polarization decay time decreases as soon as the excitation energy increases, for all excitation intensities.

The initial polarization P(0) and its variation with the excitation energy cannot be entirely attributed to the polarization rate of the photo-generated holes deduced from the valence band mixing, which we expect to remain above at least 80 % in this excitation energy range [6]. A first depolarization, relatively small in comparison with the subsequent depolarization, has probably occured in a time shorter or of the order of our time resolution.

Since the density of photo-excited holes considered above is one order of magnitude or more lower than the doping level, the spin relaxation of electrons is irrelevant and the measured decay of the luminescence polarization gives the spin relaxation time τ_h of thermalized holes. As a consequence, the cw polarization P_{cw} depends on the initial polarization P(0) and on the recombination and spin lifetimes, τ_r and τ_h respectively, by :

$$P_{cw} = \frac{P(0)}{1 + \tau_r/\tau_h} \qquad (1)$$

The measured values of P_{cw} at A, B and C (figure 1) are 62, 51 and 39 % respectively ; the measured values of P(0) at the same excitation energies (figure 4.b) are 85, 70 and 58 % respectively. We deduce a ratio $P_{cw}/P(0) \simeq 0.7$ independent of the excitation position A, B or C. This means that the decrease of P_{cw} versus the excitation energy observed in figure 1 is completely related to the decrease of the initial polarization P(0), a result in agreement with the conclusions of Ph. Roussignol *et al.* [4]. One checks that the longer spin relaxation time, measured at the lower excitation density ($n \simeq 7.10^7$ cm^{-2}) is in agreement with the measured cw polarization performed at $n \simeq 8.10^7$ cm^{-2} when one considers the same excitation energy. For instance, for the excitation energy B, we measure $\tau_h = 1200$ ps (figure 4.a) and $\tau_r = 220$ ps (independant of the excitation energy), which gives $P_{cw}(calculated) = 59\%$ compared to $P_{cw}(measured) = 51\%$.

As we demonstrate that τ_h decreases when the photocreated carrier density n increases, the decrease of the cw polarization at a given excitation energy, observed in figure 2 when n increases, is now qualitatively explained by equation (1).

The initial temperature of electrons and holes is one of the important parameters of each excitation condition. This initial temperature is an increasing function of the excitation energy and excitation intensity. Assuming that the photogenerated electron-hole pairs redistribute quasi-instantaneously their excess energy to the whole electron-hole plasma, one can calculate the initial temperature of the thermalized carriers (note that even at our higher excitation level, holes remain non-degenerate). This temperature (T_i) is reported in figures 3 and 4. Considering the inhomogeneity of the sample (revealed by the large PL line), and thus the uncertainty on the exact position of the Fermi level, it is clear that this calculated temperature is only an estimation. The important observation here is that the hole spin relaxation time τ_h is a decreasing function of T_i in each of these experiments.

The spin-flip inside the HH1 subband involves transitions between ± 3/2 states. These transitions are forbidden at $k_{//} = 0$ in quantum wells : as a consequence the hole spin lifetime is infinite. At $k_{//} > 0$ however, there is a mixing between the pure heavy and light hole states which allows spin flip within the HH1 sub-band. Then any energy or momentum relaxation process can lead to spin relaxation. For instance, spin-flip due to carrier-phonon interaction has been discussed in [3,7]. This is one of the hole spin relaxation process which has been proposed so far.

Two other spin relaxation mechanisms may be considered :

(i) first , a D'Yakonov-Perel like mechanism due to the presence of an inversion asymetry term in the Hamiltonian, associated with the GaAs zinc-blende lattice. This proposed mechanism is supported by detailed calculations of the hole spin lifetime taking into account several kinds of elastic scatterers : coulombic impurities, interface defects, alloy scattering [8]. Its efficiency increases also as $k_{//} = 0$ increases. Acoustic phonon-assisted spin-flip transitions show the same trend [4] ;

(ii) one can expect that hole scattering on electrons in heavily n-doped quantum wells may also lead to spin flip transitions due to exchange interaction (the transposition of the Bir-Aronov-Pikus mechanism [9] to holes). In presence of degenerate electrons, because of the limitation imposed by the Pauli principle, only electrons with energy near the Fermi level may cause this hole spin relaxation. The energy width of this interacting electrons layer below the Fermi-level increases with the hole kinetic energy, so the corresponding hole spin flip rate is a function which increases with $k_{//}$.

One can conclude this rapid review that all these hole spin flip processes are increasing functions of $k_{//}$, i.e. of the hole kinetic energy. As a consequence, the corresponding hole spin relaxation time is expected to decrease when the electron-hole gas temperature increases (the lattice being maintained at low temperature) or when the sample temperature T_L increases.

The measured variation of the *initial* polarization decay time as a function of the lattice temperature T_L, at low excitation density $n \simeq 4.10^8\ cm^{-2}$ is reported in figure 5. The expected decrease of τ_h is indeed observed. Moreover, we clearly verify in figure 4.a, 4.b and 4.c that the decrease of the *initial* polarization decay time, observed both when the excitation density or when the excitation energy increase, is correlated as expected to the variation of the initial electron-hole gas temperature T_i.

It is interesting to notice that the hole spin relaxation time $\tau_h$we measured recently in intrinsic MQW shows the same trend : τ_h is found to be long ($\tau_h > 50\ ps$) in resonant excitation (the electron hole temperature is in this case very low) [10, 11], whereas it is very short ($\tau_h \simeq 3\ ps$) in non-resonant excitation when the initial temperature is about $T_i \simeq 60\ K$ [12, 13]. All these results emphasize the role played by the temperature in the hole spin relaxation process. The question of the determination of the dominant process is however out of the scope of the present paper.

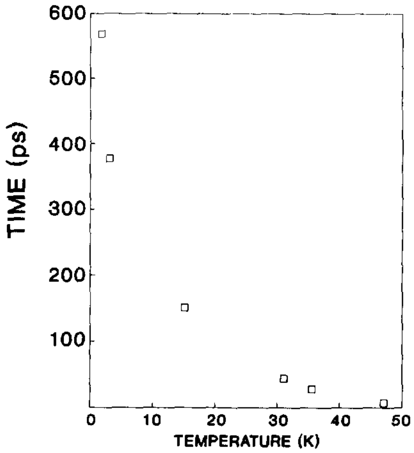

Figure 5.
Variation of the initial decay time of the polarization as a function of the lattice temperature at a low photogenerated carrier density of $n = 4.10^8\ cm^{-2}$; the dotted line is a guide for the eyes.

In conclusion, we have performed systematic measurements of the hole spin relaxation time τ_h in n-modulation doped quantum wells. Our results show for the first time the drastic dependence of τ_h both on the excitation density and on the excitation energy. We propose an interpretation which assigns these two dependences to a single cause, the initial temperature variation. However this approach may be too simple and discards other factors which could also influence the hole spin relaxation time. It is clear that further investigations are required to identify the relevant spin relaxation mechanisms.

ACKNOWLEDGEMENTS : We are grateful to M.I. D'Yakonov, V.I. Perel, Ph. Lavallard and C. Benoît à la Guillaume for useful discussions.

REFERENCES

[1] T.C. Damen, L. Vina, J.E. Cunningham, J. Shah, L.J. Sham, Phys. Rev. Lett., **67** (24), 3432 (1991)

[2] G. Bastard, R. Ferreira, Surface Science, **267**, 335 (1992)

[3] T. Uenoyama, L.J. Sham, Phys.Rev., **B 42**, 11 (1990)

[4] Ph. Roussignol, R. Ferreira, C. Delalande, G. Bastard, A. Vinattieri, J. Martinez-Pastor, L. Carraresi, M. Colocci, J.F. Palmier, B. Etienne, Surface Science, **305**, 263 (1994)

[5] M.I. D'Yakonov, V.I. Perel, Sov.Phys. JETP, **33**, 1053 (1971)

[6] B. Dareys, X. Marie, T. Amand, J. Barrau, I. Razdobreev, R. Planel, Superlattices and Microstructures, **13**, 3 (1993)

[7] J. Barrau, G. Baquet, F. Hassen, N. Lauret, T. Amand, M. Brousseau, Superlattices and Microstructures, in Press

[8] R. Ferreira, G. Bastard, Phys.Rev.B, **43**, 12 (1991)

[9] G.L. Bir, A.G. Aronov, G.E. Pikus, Sov. Phys. JETP **42**, 705 (1976)

[10] B. Baylac, T. Amand, X. Marie, B. Dareys, J. Barrau, M. Brousseau, R. Planel, to be published

[11] A. Vinattieri, J. Shah, T.C. Damen, D.S. Kim, L.N. Pfeiffer, L.J. Sham, Solid State Com., **88** (3), 189 (1993)

[12] T. Amand, B. Dareys, X. Marie, B. Baylac, J. Barrau, M. Brousseau, D. Dunstan, R. Planel, to be published in the Proceedings of the 22 nd International Conference on the Physics of Semiconductors, Canada (August 1994), ed. World Scientific Publishing Co. Ltd.

[13] however, the comparison of the hole spin relaxation time in n-doped and intrinsic QW is not straightforward since, in the latter, the hole is bound into the exciton.