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Spin relaxation of excitons in strained InGaAs/GaAs quantum wells

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Abstract: We have investigated the exciton spin dynamics in strained In$_x$Ga$_{1-x}$As/GaAs ($x \leq 0.2$) quantum wells. The variation of the initial polarization with the excitation energy is well correlated with the increasing mixing of the valence band states. The time behavior of the depolarization curves clearly demonstrates the prime importance of the hole and exciton spin relaxation processes.

Recently the spin dynamics of various excitations in GaAs/AlGaAs quantum wells (QW), initially oriented by optical pumping, has been investigated by time-resolved spectroscopy [1-3]. In nominally undoped samples, the luminescence polarization decays in a few tens of picoseconds. But this decay has been interpreted mainly in terms of an electronic spin relaxation time. In modulation n-doped QW’s, the luminescence depolarization is mainly governed at low excitation by the spin relaxation time of the holes ($\tau_h$), but the results published so far are quite controversial: Damen et al. [1] measured a $\tau_h$ value of about 4 ps, whereas Roussignol et al. [2] found a much longer one (~1 ns).

Recently, the $\tau_h$ value has been calculated theoretically in GaAs/AlGaAs QW’s. Whatever the hole spin relaxation process, the mixing of the hole states in the valence band plays a fundamental role. In GaAs/AlGaAs QW’s, this mixing may remain substantial, due to a relatively small HH-LH splitting. We believe that the study of a system with a larger HH-LH splitting, and thus a weaker Valence Band Mixing (VBM), should help the investigation of the hole spin relaxation.

In this communication we present the results of time resolved optical pumping experiments performed on nominally undoped In$_{0.2}$Ga$_{0.8}$As/GaAs heterostructures: the sample made of three 7 nm wells, separated by 10 nm barriers is typical. The straight line in figure 1 displays the stationary excitation photoluminescence spectrum. We clearly identify the El-HH1 (XH) and El-LH1 (LH) excitonic lines separated by 65 meV, a large splitting energy induced largely by the tetragonal part of the deformation.
The time-resolved experiments are performed at 1.7 K. The samples are excited by a picosecond tunable Ti-doped sapphire laser. The initially created exciton density is about $10^{10}$ cm$^{-2}$. The detection energy is set to the XH excitonic emission peak. The photoluminescence signal is detected by the up-conversion technique using a LiIO$_3$ non-linear crystal. The excitation light is right circularly polarized and we measure the luminescence components of same ($I_+$) and opposite ($I_-$) helicity as the excitation light. The time-dependence of the luminescence polarization is then calculated as $P(t) = (I_+ - I_-)/(I_+ + I_-)$.

Figure 2. Time evolution of the experimental luminescence polarization for two excitation energies: (a) $\hbar \nu = \text{XH} + 22$ meV; (b) $\hbar \nu = \text{LH} + 27$ meV. The solid line corresponds to the fit with the model presented in the text. Inset in (b): schematic diagram of the different exciton spin relaxation processes considered.

Figure 2.a shows the polarization rate versus time for an excitation energy of 22 meV above the XH absorption energy. The striking feature is that the initial polarization $P(0)$ is about 100 %, although the excitonic states are not created resonantly. Then the luminescence polarization disappears non-monoexponentially in about 400 ps.

In figure 2.b, the time evolution of the PL polarization for an excitation energy above the XL absorption line is presented. Two interesting features are noticeable:

- $P(0)$ is much lower, around 60 %, this comes from the fact that both the HH1 and LH1 bands are excited;
- the kinetic is non mono-exponential: an initial decay occurs within a time scale of a few ps, where the polarization drops to about 30 %. Then, the remaining polarization disappears in 400-500 ps.

The results presented in figures 2a and 2b prove that both the initial polarization $P(0)$ and the depolarization dynamics depend strongly on the excitation energy. We thus investigate systematically the depolarization dynamics for excitations below, around and greater than the XL energies.
Figure 1 displays the dependence of the initial polarization $P(0)$ upon the excitation energy. We plot also the calculated initial polarization for valence to conduction band transitions using the envelope function formalism and the equivalent Luttinger hamiltonian [4]. There is a reasonably good agreement between this theory and the experiment: this means that the variation of $P(0)$ versus the excitation energy is the result of VBM. The mismatch around the XL excitation is just due to the fact that the calculation does not take into account the excitonic effects.

In order to identify the mechanism of depolarization, we compared our experimental data to the phenomenological model proposed recently by Maialle et al. [5]. The rate equations of the different exciton spin states $|N\rangle$, where $N = \pm 1, \pm 2$, are written, in the following equation, as a function of the electron, hole and exciton spin transition rates, $W_e = 1/2\tau_e$, $W_h = 1/2\tau_h$ and $W_{ex} = 1/2\tau_{ex}$ respectively, the latter being driven by the exchange interaction:

<table>
<thead>
<tr>
<th>$N_1$</th>
<th>$N_2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$-\left( W_e + W_h \right)$</td>
<td>$W_e$</td>
</tr>
<tr>
<td>$-\left( 1/\tau_e + W_{ex} + W_e + W_h \right)$</td>
<td>$W_{ex}$</td>
</tr>
<tr>
<td>$W_h$</td>
<td>$W_h$</td>
</tr>
<tr>
<td>$0$</td>
<td>$W_e$</td>
</tr>
<tr>
<td>$W_{ex}$</td>
<td>$-\left( 1/\tau_e + W_{ex} + W_e + W_h \right)$</td>
</tr>
</tbody>
</table>

The calculated polarization is thus given by:

$$P_{cal.}(t) = \frac{N_1 - N_{-1}}{N_1 + N_{-1}}$$

The initial conditions of these rate equations are fundamental. We have checked that the hypothesis of exciton formation from geminate pairs always leads to an unsatisfactory fit. Consequently we performed a full description of the bimolecular formation process. The model is consistent as long as the exciton formation time is shorter than the exciton spin relaxation time [6]. We introduce the initial polarization ratio of electron and heavy hole populations:

$$\frac{n(\downarrow)}{n(\uparrow) + n(\uparrow)} = \frac{p(-3/2)}{p(-3/2) + p(3/2)} = 1 - \epsilon$$

This expression is obvious for an excitation energy below the XL energy. It is also justified above, provided that one assumes a quasi-instantaneous transfer of the photo-generated $m=-1/2$ light-holes towards the $m=+3/2$ heavy-hole states: it appears that the present experimental data, as well as others obtained previously in GaAs/GaAlAs, cannot be understood without this hypothesis [4, 7]. The $\epsilon$ value is adjusted in order to recover the experimental polarization $P(0)$.

The straight lines in figures 2a and 2b correspond to least squares fits of the experimental curves assuming the previous rate equations. It turns out that the depolarization dynamics is well described by an exciton spin relaxation time $\tau_{ex}$ (resp. 58 and 79 ps) and a shorter time (resp. 17 and 7 ps) which is
identified to $\tau_h$. The fit gives the third time ($\tau_e$) much longer, greater than 1 ns: as a matter of fact, the fit is not sensitive to this third time.

Two points have been carefully checked:
- it is impossible to fit the data with only \( \tau_e \) and \( \tau_h \) : an excitonic spin relaxation \( \tau_{ex} \) is compulsory to get a good agreement;
- the excitonic spin relaxation time alone can not explain the polarization decay as it leads to a calculated curve which is mono-exponential whereas the experimental ones are not.

As can easily be seen in equation (1), \( \tau_e \) and \( \tau_h \) play strictly equivalent roles in the fit. The assertion that the short time is \( \tau_h \) and not \( \tau_e \) is suggested by the strong variation with the excitation energy, seen in figure 3: indeed one expect no such singularity to occur for \( \tau_e \) at the energy position of the light hole subband excitation.

Figure 3 shows the dependence of the two relaxation times \( \tau_h \) and \( \tau_{ex} \) on the excitation energy \( \hbar \nu \). We point out they both decrease up to the \( \hbar \nu \) value corresponding to the photo-generation of the XL excitons, due to the increase of the VBM.

**Fig. 3.** Dependence of the heavy hole (*) and exciton (o) spin relaxation times on the excitation energy. The lines are guides to the eyes.

In conclusion, we have shown that the variation of the initial photoluminescence polarization with the excitation energy is consistent with the change of the valence band mixing. Moreover, the time behaviour of the depolarization curves clearly demonstrates the predominance of the hole and exciton spin relaxation processes. We emphasize that the hole spin relaxation is directly measured by the fast initial decay of the luminescence polarization when the first light-hole subband is excited.