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Influence of spin conservation on the carrier dynamics in InAs/GaAs quantum dots

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We present a study of two types of low temperature Photoluminescence experiments probing the state blocking effects in quantum dots. Using a random population model, we discuss the theoretical predictions for these experiments depending on whether the spin is conserved or not. The comparison of experimental results with these predictions shows that spin indeed plays a major role in the carrier dynamics.

1 Introduction The study and possible manipulation of carriers' spin in semiconductors are the subject of intense research. Studies of the spin of the carriers confined into a quantum dot (QD) have so far mainly consisted of spin lifetime measurements, with time-resolved experiments using circularly polarized excitation, or with magnetic fields [1–4]. Long (≈1 ns) [1], or even very long (up to 20 ns) [2] spin lifetimes have been reported for carriers in the Ground State (GS) of InAs/GaAs QDs. However, the behaviour of the spin during carrier relaxation has not been addressed yet in detail. Moreover, no study focuses on the possible effects of long spin lifetimes and spin conservation on the carrier dynamics in QDs. However, due to the discrete density of states in QDs, spin conservation could have an important effect on the state blocking effects, which often governs the carrier dynamics. The aim of this paper is to assess the importance of spin in the carrier dynamics. To do so, we present theoretical modelling and experimental results for two simple experiments where state blocking effects play an important role. The first one is the excitation density dependence of the Photoluminescence (PL) signals from the GS and first excited state (X1) under cw excitation. The second is the time-resolved measurement of the PL decays from GS and X1 after a short pulse creates carriers around the dots. To take into account properly the state blocking effects, we use a random population type model [5], which we modify slightly to take spin into account. We show that the predictions of the model are very different whether spin is conserved or not. The experiments were carried out on InAs/GaAs QD samples having a small inhomogeneous broadening, where the small overlap between GS and X1 signals enables us to extract accurate data. These are shown to be in complete agreement with the hypothesis of spin conservation demonstrating that QDs could play an important role in spintronics. It also highlights the fact that spin needs to be taken into account when modelling the carrier dynamics in most cases, at least at low temperatures.

2 Presentation of the results We compare the prediction of the model for the two following hypotheses: H_S, where it is assumed that spin is always conserved. This implies that the spin lifetimes in the dots are infinite and that spin is conserved during relaxation. H_N, where spin is irrelevant to the dynamics. The spin can therefore rapidly flip when a carrier is in a given state of the dot or when it relaxes.

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We assume that electrons and holes are captured into the dots as excitons, which is likely to be the case at 10 K. Because capture and relaxation times are much faster (typically 1 ps) than the radiative lifetimes (≈1 ns), we can assume that once captured, a carrier will trickle down to the lowest available state where it will recombine and emit a photon. Under these conditions, the cw PL can be modelled statistically by considering the probability p_n of having a dot containing n carriers, which follows a Poissonian distribution [5]: $p_n = x^n\,\mathrm{e}^{-x}/n!$, where x is the average QD occupation.

Within H_N, and for a dot with degeneracies 2 and 4 in the GS and X1 respectively, the PL intensity from the GS is then $R_0 = p_1 + 2p_2 + 2p_3 + \dots$. We omit a coefficient η/τ which takes into account the collection efficiency η and the radiative lifetime τ, because this coefficient is the same in each state (we assume τ is similar in GS and X1). In order to see emission from X1 we need at least 3 carriers in a dot for state blocking to operate (2 in GS and one in X1). The PL intensity from X1 is therefore $R_1 = p_3 + \dots$. In principle, the calculations can be made up to any order of x, but here we restrict the analysis to the first few terms, since x will be small in our experiments. We then have

$$r = \frac{R_1}{R_0} = \frac{x^2}{6}\left(1 - \frac{x}{2}\right) \approx \frac{x^2}{6}. \tag{1}$$

If there are some non-linear effects in the creation of photo-carriers or during their diffusion towards the dots, the input laser power may not be exactly proportional to x. To avoid this problem, we can directly measure the Integrated PL intensity, I, to estimate accurately x. Within our approximations, we indeed have $I = x$. By measuring the variation of r as a function of I (which is changed by changing the laser power), we can then easily check whether the previous formula is correct.

It is straightforward to adapt this analysis to a case where carriers can have two different spin states (up and down). The average number of carriers per dot in each state is now x_u and x_d (with $x = x_u + x_d$). If spin is conserved, then the two populations are independent and the probability q_{ij} of having i carriers with spin up and j with spin down is $q_{ij} = (x_u^i x_d^j)\,\mathrm{e}^{-x}/(i!j!)$. The main difference is that within H_S, it is now possible to have emission from X1 with only two carriers in a dot, provided they have the same spin. We therefore have $R_0 = q_{10} + q_{01} + q_{20} + q_{02} + 2q_{11} + \dots$ and $R_1 = q_{20} + q_{02} + \dots$. For the simple case where $x_u = x_d = x/2$ (unpolarized excitation), we then obtain

$$r = \frac{R_1}{R_0} = \frac{x}{4}\left(1 + \frac{x}{12} - \frac{x^2}{48}\right) \approx \frac{x}{4}. \tag{2}$$

For polarized excitation and spin conservation, we would have $x_u = x$ and $x_d = 0$, which gives

$$r = \frac{R_1}{R_0} = \frac{x}{2}\left(1 + \frac{x}{6} - \frac{x^2}{12}\right) \approx \frac{x}{2}. \tag{3}$$

It is interesting to note that the same equation would also be obtained within hypothesis H_N (no spin effects) if the degeneracies of the states were half what is usually assumed (e.g. 1 for GS, 2 for X1, . . .)

Figure 1 shows a plot of the ratio $r = R_1/R_0$ as a function of I. In the inset is shown the PL spectrum obtained for the point corresponding to the highest excitation density. The overlap between the GS and X1 peaks is small, which enables a precise deconvolution of the two peaks and an accurate determination of the ratio r. The plot is clearly linear and not quadratic as would have been expected if spin was not conserved. This strongly supports the fact that H_N is incorrect and H_S is correct.

We have shown for a simple cw experiment that the state blocking effects can be very different whether spin is conserved or not. However, these effects are even more important in a time-resolved experiment, where the PL decay from each state is measured. Many such experiments have been carried out to study carrier dynamics [6, 7]. By studying the rise of the transients, the capture and relaxation times of carriers can be estimated [7] and are measured to be very fast (a few picoseconds). Moreover, radiative lifetimes can be extracted from the measure of the decay rates at long times. It has been pointed out, that providing that the relaxation times are much faster than the radiative lifetimes (which is usually

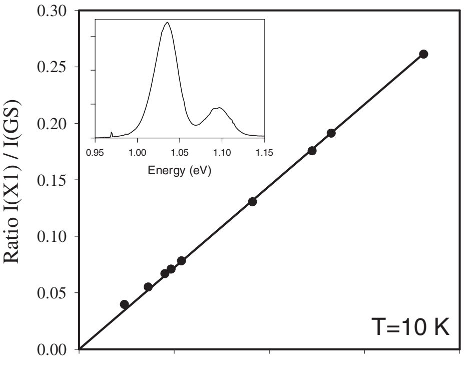

Fig. 1 Plot of the ratio r between PL intensities of X1 and GS as a function of the integrated PL intensity. The inset shows the PL spectrum corresponding to the highest excitation.

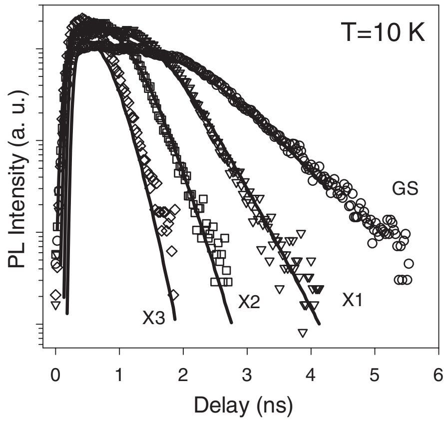

Fig. 2 PL decays measured from ground state (circles), first (triangles), second (squares), and third (diamonds) excited states of the dots after non-resonant high power pulsed excitation at $t = 0$.

the case), the actual values of the relaxation and capture times do not affect the behaviour at long times [5, 6]. This is very important, since it enables major simplifications to be made by assuming these rates are infinitely fast. It is then relatively easy to relate the measured decay times τ_0^m, τ_1^m, and τ_2^m, in the ground, first and second excited states to the real values τ_0, τ_1, and τ_2 of the radiative lifetimes in these states [6],

$$(\tau_0^m)^{-1} \approx \tau_0^{-1}\,, \qquad (\tau_1^m)^{-1} \approx \tau_1^{-1} + g_0\tau_0^{-1}\,, \qquad (\tau_2^m)^{-1} \approx \tau_2^{-1} + g_1\tau_1^{-1} + g_0\tau_0^{-1}\,, \tag{4}$$

where g_0, g_1 and g_2 are the degeneracies of the ground, first and second excited states. The importance of the degeneracies clearly appears in these formula, but is often overlooked. Moreover, the importance of spin was rarely mentioned or taken into account in existing reports [6]. To understand its importance, one can look at the origin of the term g_0 in the expression of τ_1^m. Because the relaxation times are very fast, we only see emission from X1 when a carrier is present in X1 *and* is inhibited from relaxing to the GS because of Pauli blocking. Such a carrier has two ways of disappearing from X1. First, it can be emitted radiatively, with a rate τ_1^{-1}. Alternatively, it can relax to the GS, providing a space becomes available. This second path will occur with a rate equal to the rate of appearance of a vacancy in the GS. This rate is τ_0^{-1} for one carrier in the GS but is multiplied accordingly if more carriers are present, hence the total rate for this path: $g_0\tau_0^{-1}$. However, if spin is conserved during relaxation, a carrier in X1 with a given spin will be able to relax to the GS only if a carrier with the same spin disappears from the GS. The rate of appearance of an appropriate vacancy in the GS is therefore half of what it was without spin effects. The measured asymptotic decay times are then given by Eq. (4), but replacing g_i with $g_i/2$. It is important to realize that the predictions when spin is conserved or not are remarkably different. For example, if we take $\tau_0 = \tau_1$ to simplify, we would have $\tau_1^m = \tau_0^m/3$ when spin is not conserved, and $\tau_1^m = \tau_0^m/2$ when it is. Such a large difference is easily observable in an experiment.

In Fig. 2 are shown typical PL decays obtained under high excitation from the first four confined states of a QD sample. We are here interested in the decay rate of each state at long times, which can easily be extracted from these data:

$$\begin{aligned} (\tau_0^m)^{-1} &\approx 1.45\ \text{ns}^{-1}, & \tau_0^m &\approx 690\ \text{ps}, \\ (\tau_1^m)^{-1} &\approx 2.6\ \text{ns}^{-1}, & \tau_1^m &\approx 385\ \text{ps}, \\ (\tau_2^m)^{-1} &\approx 4.2\ \text{ns}^{-1}, & \tau_2^m &\approx 238\ \text{ps}\,. \end{aligned} \tag{7}$$

We infer from these measurements that the radiative lifetime in the GS is $\tau_0 = \tau_0^m = 690$ ps. Inserting the values of τ_0 and τ_1^m in Eq. (6), we see that $g_0 = 2$ is impossible since it leads to a negative value for τ_1. However, using $g_0 = 1$ leads to a reasonable value for τ_1 of 870 ps. Similarly, the Eq. in (4) for τ_2 leads to aberrant results for $g_0 = 2$ and $g_1 = 4$, while reasonable agreement is obtained for $g_0 = 1$ and $g_1 = 2$. We note that the value of τ_2 obtained with this method is quite sensitive to any errors in the measurements of the rates τ_i^m. Nevertheless, these arguments again show that the QD state degeneracies have to be taken equal to $g_0 = 1$, $g_1 = 2$, etc... Moreover, only using these values is it possible to obtain good fits to the experimental data as shown by the solid lines in Fig. 2.

3 Conclusions The two sets of experiments presented above are clearly incompatible with hypothesis H_N, and strongly support hypothesis H_S. More precisely, they unambiguously show that carrier dynamics in QDs occur as if the degeneracies of the states were $g_0 = 1$, $g_1 = 2$, etc... However, because of the spin degeneracy, the reality of having such degeneracies is difficult to justify physically. One could argue that the energy states we observe are not confined energy levels of charged carriers as usually assumed, but are the eigenstate of a more complicated Hamiltonian resulting from a strong coupling to another particle (such as polarons as a result of a strong coupling with phonons). The degeneracies of such a strongly coupled system could then be different to what is usually expected. Another way to explain this result would be to assume that carrier–carrier interactions (such as Coulombic effects) could prevent two carriers to be present in the GS at the same time. However, it is unlikely that the same effect would allow two (and not one or four) carriers to be present at the same time in the first excited state. The most probable explanation is therefore that spin plays a major role in the carrier dynamics in QDs. For this to be true, the spin lifetime in the confined levels of a dot needs to be much longer than the radiative lifetimes. This has already been shown to be the case, at least for the GS [2]. The second prerequisite is that carrier relaxation in QDs can only occur between states with a given spin configuration. The more natural mechanism would be that spin is conserved during relaxation, but it would also be compatible with the present experiments if spin was systematically flipped during relaxation.

To conclude, this simple analysis demonstrates how important spin can be to carrier dynamics in QDs. It is interesting to see that its importance can readily be observed in experiments with unpolarized excitation, because it has a direct effect on state blocking of carriers.

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