Optical spin-filtering effect in charged InAs/GaAs quantum dots

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(Received 10 September 2010; accepted 6 October 2010; published online 27 October 2010)

We present time resolved photoluminescence results using nonresonant polarized light which show that the electron spin-flip time is much longer than the recombination time for an ensemble of *p*-doped InAs/GaAs quantum dots. Under continuous wave excitation the degree of optical polarization of the ground state is found to be around 10%. However, the excited state polarization is twice this value. We attribute this effect to Pauli blocking of the injected spin population captured into the dots and show that the effect persists up to room temperature. For resonant excitation, values are nearly doubled in accordance with increased spin injection efficiency. © *2010 American Institute of Physics*. [doi:10.1063/1.3506507]

There is considerable interest in spin systems as possible qubits for quantum information processing,[1] and for development of spin-based devices including spin light-emitting diodes[2–5] and spin lasers.[6] Quantum dots (QDs) are appealing because they shield the trapped carriers from any external influences that might randomize the carrier spin. Photogeneration of carriers results in the generation of excitons whose spin lifetimes in QDs are limited to a few hundreds of picoseconds due primarily to the electron-hole exchange interaction.[7–9] This is much shorter than the typical exciton recombination time of 1 ns and it seems unlikely that excitons will lead to useful qubits. Interest has now focused on localized electron or hole spins introduced by doping or charging in Schottky devices. Results have shown that isolated electron[10] or hole[11] spin lifetimes can extend to milliseconds. *p*-doped samples exhibit reduced electron capture and relaxation times,[12,13] reduced radiative lifetimes,[14] and also enhanced spin lifetimes,[4,15,16] this enhancement being attributed to suppression of the anisotropic electron-hole exchange interaction by formation of a spin singlet consisting of one electron and two holes.[8,9] In this work we investigate the circularly polarized, time-resolved emission for a range of *p*-doped QD samples excited with nonresonant circularly polarized light and show that the luminescence lifetime is reduced by almost a factor of 2 due to the presence of holes while the spin-flip lifetime is increased by a factor of 5 in a sample containing 10 holes per dot. This allows spin effects to be seen in steady state measurements and these reveal that the polarization of the QD first excited state transition is roughly twice that of the ground state (GS) indicating a spin-filtering effect due to Pauli blocking[17] and the state degeneracy. The polarization decreases with increasing excitation power but interestingly persists to room temperature albeit slightly reduced. Under resonant excitation the degree of polarization is roughly doubled due to the increased spin injection efficiency arising from the lifting of the degeneracy of the heavy and light hole states in a quantum-confined structure.[18]

Samples were grown by molecular beam epitaxy on semi-insulating GaAs (001) substrates. The QD layer was grown between AlGaAs barriers and has a dot density of $\sim 2\times 10^{10}$ cm^{-2}, with average uncapped height 8 nm and diameter 40 nm as deduced from atomic force micrographs. These samples were previously used in a study of the effect of *p*-doping on exciton radiative lifetimes and further details can be found in Ref. 14. Steady state and time resolved photoluminescence (TRPL) measurements were performed using a Ti–sapphire laser operating in cw or pulsed mode (2.4 ps pulses, 82 MHz repetition rate) at a wavelength of 790 nm. Circularly polarized excitation was obtained using a Glan–Thompson polarizing prism and quarter wave plate. The PL was resolved into copolarized and cross-polarized components using a broadband (1000–1300 nm) quarter wave plate and near infrared linear analyzer, dispersed with a Spex 0.85 m double monochromator and detected either with a microchannel plate with an extended S1 response or an InGaAsP photomultiplier with a response from 950–1400 nm, using time-correlated single-photon counting techniques for the time-resolved measurements. Resonant excitation was achieved using a 980 nm diode laser.

The inset to Fig. 1 shows low temperature (12 K) TRPL for the sample doped at a level of 10 holes per dot excited using σ^+ light and resolved into σ^+ or σ^- components by rotating the analyzing wave plate through 90°. At the excitation wavelength absorption occurs into the bulk GaAs creat-

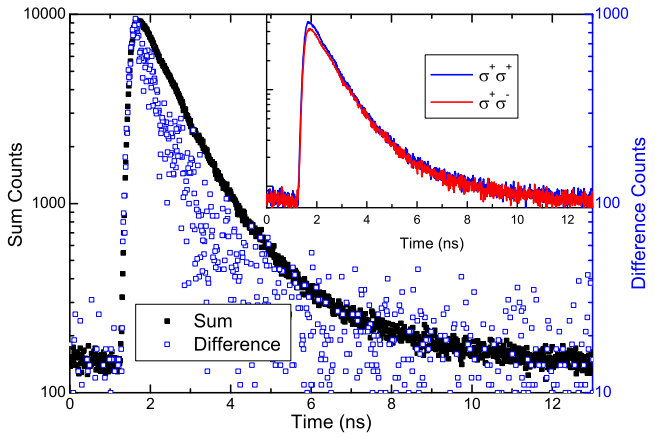

FIG. 1. (Color online) TRPL measured at the GS emission peak for the most heavily doped sample (10 holes per dot) excited with σ^+ light at 790 nm. The inset shows the copolarized and counter polarized emission. The main part of the figure shows the sum and difference spectra.

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TABLE I. Low temperature (12 K) luminescence and spin-flip lifetimes obtained from TRPL measurements from samples *p*-doped at a level of 0, 1, 3, and 10 holes per QD.

(holes/QD) *p*-doping	(ns)	0	1	3	10
τ_{lum}	(±0.01)	1.19 ns	1.27 ns	1.10 ns	0.70 ns
τ_{sf}	(±0.1)	⋯	0.6 ns	0.9 ns	3.6 ns

ing a 50% spin polarized population of excitons.[18] Rapid spin relaxation of the holes occurs leaving a relatively small majority of spin polarized electrons captured into the QDs. This is not evident in the TRPL traces where the maximum signal is dependent on the integration time. Recombination of either spin up or spin down electrons with a randomized population of holes should not result in any difference in the PL decay time, τ_{lum}, and this is evident in the data. Nevertheless we anticipate some electron spin flipping prior to recombination with a lifetime, τ_{sf}, and we model the two spin populations by the following rate equations:

$$\frac{dN^+}{dt} = -\frac{N^+}{\tau_{lum}} - \frac{N^+}{\tau_{sf}} + \frac{N^-}{\tau_{sf}}, \tag{1}$$

$$\frac{dN^-}{dt} = -\frac{N^-}{\tau_{lum}} - \frac{N^-}{\tau_{sf}} + \frac{N^+}{\tau_{sf}}, \tag{2}$$

where $N^{\pm}$ are the initial spin up/down populations. By adding $(S=N^+ + N^-)$ and subtracting $(D=N^+ - N^-)$ these rate equations we can separate the spin-flip and luminescence lifetimes according to:

$$S = S_0 e^{-t/\tau_{lum}}, \tag{3}$$

$$D = D_0 e^{-t(1/\tau_{lum} + 2/\tau_{sf})}. \tag{4}$$

The time evolution of S and D are shown in Fig. 1 for the most heavily *p*-doped sample. Table I shows the values of τ_{lum} and τ_{sf} extracted for all four samples: undoped and doped with 1, 3, and 10 holes per dot. The results show a significant reduction in the luminescence lifetime as expected due to the resident hole population.[14] The surprising result is the dramatic increase in the spin-flip lifetime to 3.6 ns for the 10 holes per dot sample; a factor of 5 greater than the luminescence lifetime for this sample. For the samples doped with 3 holes per dot the luminescence and spin lifetimes are similar. We attribute the increased spin-flip lifetime to suppression of the electron-hole exchange interaction due to the filling of the lowest hole states, with the sample containing 3 holes per dot representing the break point where this effect becomes important.

If τ_{sf} is long compared to τ_{lum}, evidence for spin polarization should also be present in frequency domain spectra. Figure 2 shows the cw spectra obtained for the most heavily doped sample excited using σ^+ light with the detection system measuring copolarized ($I_{\sigma^+\sigma^+}$ black line) or counterpolarized ($I_{\sigma^+\sigma^-}$ red line) emission. Using linearly polarized excitation there is no measurable polarization in the emitted light. In order to account for the effect of emission from the GS of small QDs that is coincident with the peak of X1 emission,[19] we have measured the emission under conditions of extremely low excitation to deduce the emission spectrum due to GS alone; this is indicated by the shaded region which

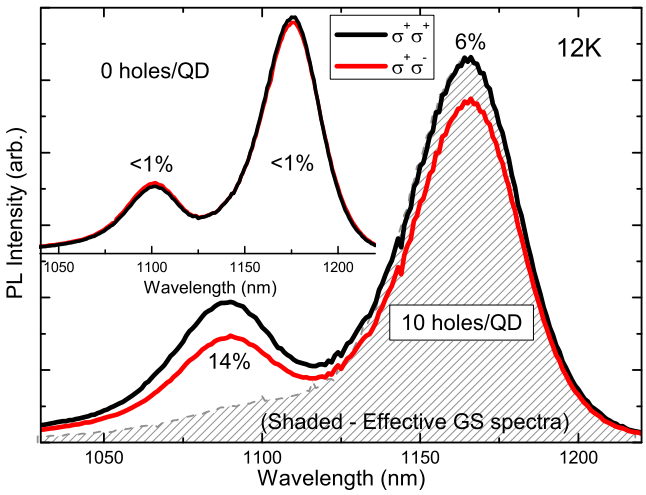

FIG. 2. (Color online) Low temperature (12 K), high power cw excitation, polarization resolved PL spectra obtained from the most heavily doped sample. The measured polarization is indicated for the GS and X1 peaks. The shaded region shows the spectral shape of the unpolarized PL obtained at very low excitation where the X1 signal is negligible normalized to the $\sigma^+\sigma^-$ spectrum. The inset shows the high excitation power polarized data obtained from the undoped sample.

in this case has been normalized to the peak of $I_{\sigma^+\sigma^+}$. Subtracting this from the actual spectrum reveals the true X1 emission. The measured degree of optical polarization, given by $\Pi=(I_{\sigma^+\sigma^+}-I_{\sigma^+\sigma^-})/(I_{\sigma^+\sigma^+}+I_{\sigma^+\sigma^-})$, is then deduced to be 6% for the GS emission and 16% for the first excited state emission, X1 at a relatively high excitation of 600 W cm^{-2}. The inset shows the spectra obtained from the undoped sample excited with σ^+ light. The difference in intensity of the σ^+ and σ^- emitted light is less than 1% and represents our experimental accuracy.

The relative populations of GS and X1 are dependent on the laser excitation level and the degeneracy of the states. We have investigated the relative polarization properties of the GS and X1 emission over a range of excitation power, Φ, and these data are shown in Fig. 3. For the GS, Π is 10.5% at the lowest value of Φ; we take this to be a measure of the

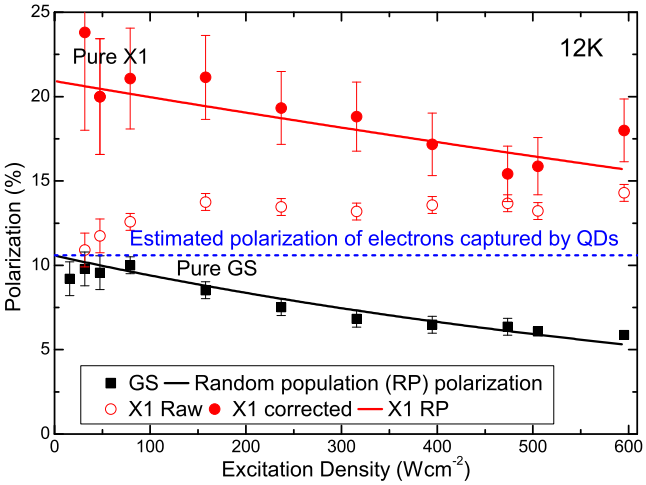

FIG. 3. (Color online) Variation in Π measured for the GS and X1 features with cw excitation power for the most heavily doped sample. The lines are fits to the data applying a random population model. Note the factor of 2 difference in Π for the two optical transitions. The open circles show the measured data ignoring the scaling of the X1 signal due to overlap with the GS signal.

residual spin polarization of electrons captured by the dots[13] and this is indicated by the dashed line. This value of the spin polarization of the captured QDs is larger than has been reported for spin injection into undoped QDs[2,3] and is comparable to previous reports for *p*-doped QDs[4,9] (although here we do not use an applied magnetic field). The reduction from the expected initial spin polarization of the photogenerated carriers to the measured spin polarization from the QDs, over a timescale of τ_{lum}, is attributed to the hyperfine interaction between the electron and nuclei in the QD.[9] With increasing Φ, Π drops almost linearly to 6% at the highest value used here. It is well known that carriers are captured randomly during relaxation from the bulk and we anticipate that this may have some influence on the results. Thus, we have applied a simple statistical model, first proposed in Ref. 20, to include random population of the dots in the ensemble. The model has also been slightly modified to take into account the two spin populations weighted according to the value of Π and in the limit that $\tau_{lum} \ll \tau_{sf}$. The full line is a fit to the data and is seen to give excellent agreement with the results. To understand the deviation from the dashed line we need to consider the X1 emission. No X1 emission is observed for the lowest Φ, as expected. As Φ increases the raw data produces a value of Π that is comparable with the GS value indicated by the open circles. With increasing Φ, Π increases and saturates at a value around 14%. As previously explained these raw data must be scaled to take into account the overlap with emission arising from GS emission of smaller dots. The scaled data are shown by the full circles and indicate an increase of a factor of 2 compared to the GS polarization across the whole range. Applying the same random population model generates an excellent fit with the data indicated by the full line. The difference between GS and X1 polarization is attributed to the effects of Pauli blocking;[17] under circularly polarized excitation a majority spin polarization is generated indicated by the dashed line in Fig. 3. Since the GS is twofold degenerate the exclusion principle forbids spin-conserving relaxation from X1. Consequently X1 retains more of the majority spin carriers than GS. The *p*-doped dot therefore acts as a spin-filter offering polarizations around 20% (16%) at the X1 emission energy under low (high) excitation. It is arguable that these measurements have been obtained under the favorable condition of low temperature where thermal escape is likely to enhance the degree of polarization. Repeating these experiments at 300 K halves the value of Π but the measured value of 7% is nevertheless substantial. An improvement in this value might be expected for resonant excitation into a dot level or the wetting layer (WL) (raising the value of the dashed line in Fig. 3). In practice it becomes difficult to exclude the effects of the exciting laser and the absorption is lower by an order of magnitude compared with the results presented above. Nevertheless we have obtained a value of $\Pi = 16\%$ for X1 for excitation at 980 nm almost resonant with the WL at 300 K. This is roughly double the value measured for nonresonant excitation as expected (to be published).

In conclusion, we have shown that nonresonant optical excitation into *p*-doped QDs results in a spin injected population close to 10%. The measured polarization of the X1 state is double this value reflecting the degeneracy of the states. The effect persists, albeit at a lower level, up to room temperature even in the absence of an applied magnetic field. Using resonant optical excitation the spin population can be roughly doubled but the same effect is also seen. These results may offer an avenue for enhanced spin detection by using the increased polarization of emission from X1 in *p*-doped QDs.

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