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Citation: Applied Physics Letters **84**, 2793 (2004); doi: 10.1063/1.1695637
View online: http://dx.doi.org/10.1063/1.1695637
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/84/15?ver=pdfcov
Published by the AIP Publishing

Efficient electron spin detection with positively charged quantum dots

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(Received 5 November 2003; accepted 11 February 2004)

We report the application of time- and polarization-resolved photoluminescence up-conversion spectroscopy to the study of spin capture and energy relaxation in positively and negatively charged, as well as neutral InAs self-assembled quantum dots. When compared to the neutral dots, we find that carrier capture and relaxation to the ground state is much faster in the highly charged dots, suggesting that electron–hole scattering dominates this process. The long spin lifetime, short capture time, and high radiative efficiency of the positively charged dots, indicates that these structures are superior to both quantum well and neutral quantum dot light-emitting diode spin detectors for spintronics applications. © *2004 American Institute of Physics.*
[DOI: 10.1063/1.1695637]

The efficient detection of spin-polarized carriers is a crucial issue for the design of semiconductor-based spintronic devices.[1] A light-emitting diode (LED) configuration employing a quantum well as an optical marker has proven to be an effective means of detecting spin-polarized carriers.[2,3] In a spin LED, the degree of circular polarization of the quantum well luminescence provides a direct measure of the spin polarization of the carriers arriving at the spatial location of the quantum well.

The application of semiconductor quantum dots (QDs) in such a spin detection scheme is expected to provide a substantial improvement in spin sensitivity over the use of a quantum well.[4–7] Recent studies of electron spin dynamics in neutral QDs[4,5,8] have revealed that the discrete energy level structure in quantum dots blocks the dominant spin relaxation channels present in higher dimensional systems, resulting in considerably longer spin lifetimes. Combined with the high optical luminescence efficiency observed in QDs,[9–11] these long spin lifetimes should lead to larger spin-dependent luminescence signatures in spin detection applications incorporating QDs as an optical marker. The first spin LED using neutral QDs was recently demonstrated.[6,12]

Few experiments have examined electron spin dynamics in charged QDs.[13,14] Through a comparison of spin capture and relaxation dynamics in neutral, positively (+QDs) and negatively (−QDs) charged QDs, we demonstrate that +QDs act as a highly efficient detector for spin-polarized electrons. Our room-temperature time-resolved measurements reveal that, following capture of spin-polarized electrons, the initial degree of circular polarization of the QD ground-state luminescence is more than four times larger in +QDs compared to neutral QDs. The larger spin signature in +QDs originates from an increased rate of capture of spin-polarized electrons through interaction with the built-in hole population,[15,16] as indicated by the early time dynamics of the QD photoluminescence (PL). Rapid electron capture into +QDs reduces spin relaxation in the GaAs barriers prior to capture, resulting in a six-fold enhancement in the time-integrated spin detection efficiency with the incorporation of positive charge on the QDs. Our experiments, which represent the first measurement of electron spin dynamics in positively charged QD nanostructures, also indicate that the presence of a large population of excess holes has little if any influence on the electron spin relaxation time in the QDs, in contrast with the findings in *p*-type bulk semiconductors[17] and in modulation *p*-doped quantum wells.[18,19]

The self-assembled InAs QDs were grown by molecular-beam epitaxy under identical conditions for structures with and without modulation doping. Each sample contains a single layer of QDs embedded in the center of a 30 nm layer of GaAs, cladded on both sides with 340 nm AlGaAs barriers. For all samples, 15 nm of undoped GaAs was deposited, followed by 2.7 monolayers of InAs to form the QD layer. Atomic force microscopy and cross-sectional transmission electron microscopy on similar structures indicate that the QDs are (25±5) nm in diameter and 3 nm in height, with an areal density of 3×10^{10} cm^{-2}. For the neutral QD structure, 15 nm of undoped GaAs was deposited over the QDs, while for the modulation-doped structures, 12 nm of undoped GaAs was followed by 3 nm of GaAs doped with Si (−QDs) or Be (+QDs) at a density of 2×10^{18} cm^{-3}. Free charge carriers from the doped layer accumulate in the lower-energy states within the InAs QDs. Since this doping density corresponds to ~20 free carriers per QD, the QDs are highly charged, in contrast to previous measurements of spin dynamics in −QDs with a single extra electron.[13,14] Evidence of substantial charge accumulation in the QD states is provided by results of continuous-wave photoluminescence (CWPL) experiments, as shown in the inset of Fig. 1(c). In previous CWPL studies on charged QDs, the ground-state optical transition was observed to shift to lower (higher) energies with the accumulation of excess electrons (holes) by 2 to 4 meV per excess carrier.[20,21] The large energy shifts in Fig. 1, corresponding to a few tens of meV, indicate the high degree of charge accumulation in the QDs studied here.

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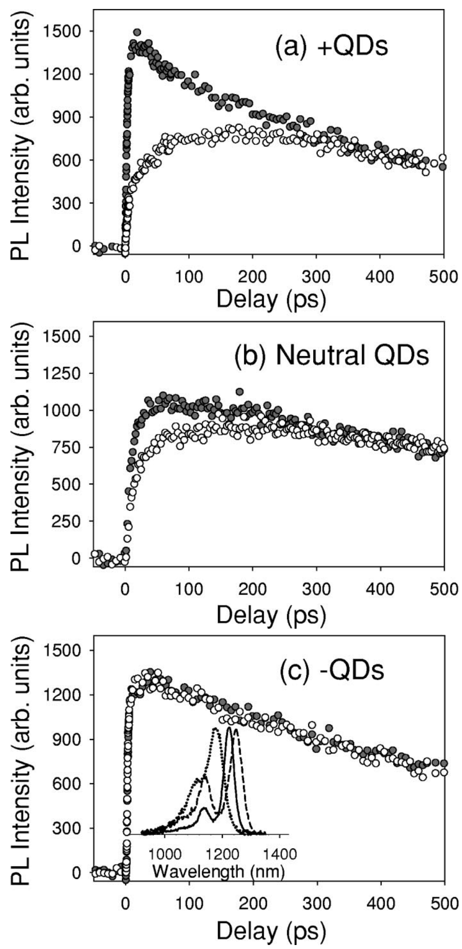

FIG. 1. Results of time-resolved PL experiments at 300 K on (a) +QDs, (b) neutral QDs, and (c) −QDs, showing emission from the ground-state optical transition. Filled (open) circles indicated dynamics of the optically injected (minority) carrier spin population following capture and relaxation into the QDs. Inset: Results of CWPL experiments on neutral QDs (solid curve), +QDs (dotted curve), −QDs (dashed curve), normalized to the peak of the ground-state emission.

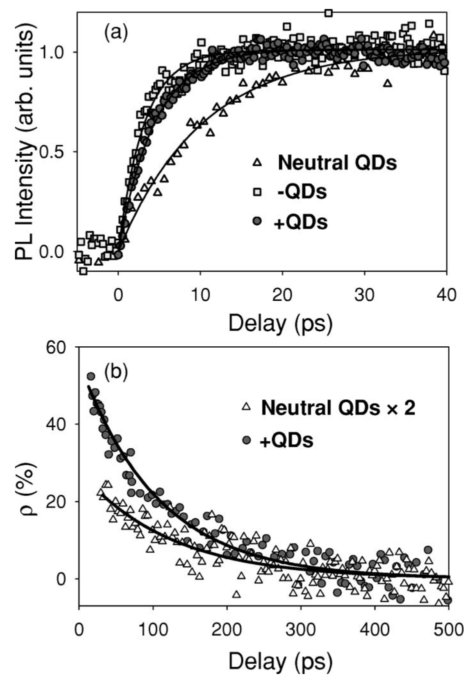

FIG. 2. (a) Early PL dynamics under excitation with linearly polarized, 1.42 eV pulses in order to probe the spin-independent carrier capture rates in charged and neutral QDs. Circles: +QDs, triangles: Neutral QDs, and squares: −QDs. In neutral QDs, the PL reaches a maximum at 45 ps, while in charged QDs, this maximum occurs earlier (13 ps, −QDs and 18 ps, +QDs). (b) Decay of degree of circular polarization in +QDs and neutral QDs. The data for neutral QDs is multiplied by a factor of two for clarity. Single exponential fits (solid curves) indicate decay times of (110±5) ps and (120±8) ps (neutral QDs).

Time-resolved PL experiments were performed with 100 fs, 1.42 eV pulses from a Ti:Sapphire laser tuned to excite carriers near the GaAs band edge. Under circularly-polarized excitation, the optical selection rules[17] lead to the generation of 50% spin-polarized carriers. The PL from the ground-state optical transition in the QDs is time-resolved using sum frequency generation in a $KNbO_3$ crystal. Emission from the majority (minority) spin populations is distinguished using a quarter waveplate in the PL path. For our experiments, the excited carrier density corresponds to about one electron–hole pair per QD, as estimated using the measured fluence and the absorption coefficient of bulk GaAs.[22]

Results of PL experiments on charged and neutral QDs are shown in Figs. 1(a)–1(c). Due to the spin-sensitive optical selection rules in self-assembled InAs QDs for light emission along the (001) growth direction,[7] the degree of circular polarization of the QD ground state PL, given by

$$\rho(\%) = 100 \times \frac{\sigma_+ - \sigma_-}{\sigma_+ + \sigma_-}, \tag{1}$$

directly reflects the spin polarization of carriers in the QDs. The results in Fig. 1 illustrate the strong effect of the QD charge on the size of the carrier spin polarization: ρ is four times larger in +QDs than in neutral QDs, while for −QDs, no circular polarization is observed for any time delay.

The contrasting amplitudes of the circularly polarized emission for the charged and neutral QDs is caused by the influence of the built-in carrier population on: (i) The species of carrier captured (electron or hole), and (ii) the carrier capture rate. In charged QDs, the ground state will be full of electrons (−QDs) or holes (+QDs) prior to optical excitation in the GaAs. Ground state emission in charged QDs therefore originates from the capture and relaxation of only the opposite charge carriers. The degree of circular polarization of the PL from +QDs (−QDs) thus provides an indication of the residual spin polarization of captured electrons (holes). The study of spin dynamics in modulation-doped QDs with PL techniques, therefore, allows for the unambiguous separation of electron and hole spin dynamics, unlike resonant pump–probe experiments.[4] The early time dynamics of the PL under linearly polarized excitation [Fig. 2(a)] indicates that carriers are captured more rapidly into charged QDs than neutral QDs. This result is attributed to a carrier capture process mediated by the interaction with the built-in carriers in charged QDs.[15,16,23]

As shown in Fig. 2(b), the rapid electron capture into +QDs leads to a substantially larger spin signature than in neutral QDs: At the time delay corresponding to the peak of the QD PL, the degree of circular polarization is 50% in +QDs, compared to 10% in neutral QDs. The observation of 50% in +QDs, which is equal to the spin polarization of the carrier distribution initially injected into the bulk GaAs surrounding the QDs, indicates that no electron spin information is lost in the GaAs during the time required for capture into +QDs. In contrast, more than three quarters of the initial electron spin polarization has been lost during the longer 45 ps PL rise time in neutral QDs. The absence of circularly polarized PL from the −QDs indicates that hole spins are completely randomized prior to capture.[8,17] This result also indicates that the circularly polarized emission observed in neutral QDs may be attributed exclusively to the residual spin polarization of captured electrons.

Single exponential fits to the decay of the circularly polarized emission, which appear as the solid curves in Fig. 2(b), provide decay times of (120 ± 8) ps and (110 ± 5) ps for the neutral QDs and +QDs, respectively. Due to the random capture process for carriers into the QDs,[24,25] the decay of the circularly polarized PL reflects the combined dynamics of spin relaxation and spin redistribution among the QD ground and excited states.[4,8] The decay time of ρ was observed to increase sharply with decreasing optically injected carrier density down to the lowest densities accessible in these experiments (one electron–hole pair per QD), indicating that the effect of capture of opposite spin electrons is significant. Nevertheless, the low density decay times we observe are in line with previous room temperature-measurements on neutral QDs.[4] The most remarkable aspect of the results of Fig. 2(b) is the observation of similar decay times of the circularly polarized PL in neutral QDs and +QDs. It has been suggested that the presence of holes leads to an efficient spin relaxation channel for electrons in QDs through the electron–hole exchange interaction.[26] Indeed, excess holes were found to strongly reduce the electron spin decay time in *p*-doped bulk semiconductors[17] and in *p*-modulation-doped quantum wells.[18,19] The room-temperature experimental results we present here, which represent a measurement of electron spin dynamics in +QDs, indicate that the presence of even a large population of excess holes has little if any influence on the electron spin dynamics.

In order to examine the implications of the higher electron spin polarization in +QDs for a spin detection application, such as a spin LED,[2,3,6] we evaluated the steady-state circularly polarized luminescence for +QDs and neutral QDs by numerically integrating the measured ground-state PL over time delay for the two polarization geometries.[27] These findings indicate that the rapid spin-polarized electron capture into +QDs leads to a six-fold increase in the steady-state circularly polarized emission, demonstrating a substantial improvement in the electron spin detection efficiency in QDs with the incorporation of a large density of built-in holes.

In summary, we have applied time- and polarization-resolved PL experiments to the study of spin-polarized carrier dynamics in charged and neutral InAs QDs at room temperature. Compared to neutral QDs, shorter carrier capture and relaxation times were measured in charged QDs, attributed to electron–hole interactions involving the built-in carriers. In +QDs, these electron–hole scattering processes lead to the rapid transfer of optically injected spin-polarized electrons from the bulk GaAs to the QD ground state, resulting in a six-fold enhancement in the time-integrated circularly polarized photoluminescence compared to neutral QDs. Measurements of spin decay dynamics in +QDs and neutral QDs indicate that the large built-in hole population in the +QDs has little effect on the electron spin relaxation time. These factors, together with the high radiative efficiency of QDs, indicates that LEDs incorporating positively charged QDs offer superior performance as optoelectronic detectors of spin-polarized electrons.

This research is supported by the DARPA MDA972-01-C-0002 and the Natural Sciences and Engineering Research Council of Canada.

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