AIP | Applied Physics Letters

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Citation: Applied Physics Letters **90**, 153118 (2007); doi: 10.1063/1.2721380

View online: http://dx.doi.org/10.1063/1.2721380

View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/90/15?ver=pdfcov

Published by the AIP Publishing

APPLIED PHYSICS LETTERS **90**, 153118 (2007)

Time-resolved Faraday rotation measurements of spin relaxation in InGaAs/GaAs quantum dots: Role of excess energy

J. L. Robb, Y. Chen, A. Timmons, and K. C. Hall[a]
Department of Physics, Dalhousie University, Halifax, Nova Scotia B3H1Z9, Canada

O. B. Shchekin and D. G. Deppe[b]
Microelectronics Research Center, Department of Electrical and Computer Engineering, The University of Texas at Austin, Austin, Texas 78712-1084

(Received 1 November 2006; accepted 9 March 2007; published online 12 April 2007)

The authors report measurements of room temperature spin dynamics in InGaAs quantum dots using time-resolved differential transmission and Faraday rotation techniques. They observe an enhancement of the electron spin lifetime by an order of magnitude for direct optical pumping of the quantum dot ground state compared to optical pumping of the GaAs barriers. These findings indicate that the optical excitation conditions can have a critical influence on the spin kinetics, a result which may account for the wide variation of spin lifetimes reported to date. The enhancement in spin lifetime observed here is attributed to the reduction of phonon-mediated spin-flip scattering. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2721380]

Due to the strong potential of spin states in semiconductor quantum dots (QDs) for applications in quantum computation,[1–3] electron spin relaxation kinetics has been the focus of numerous experimental and theoretical studies in recent years with the aim to identify the operative spin relaxation mechanisms.[4–21] Theoretical proposals have largely centered on two possible mechanisms: (i) electron-phonon interactions in the presence of spin-orbit coupling[9,13,14] and (ii) the hyperfine interaction between electron and lattice nuclear spins.[15–17] However, measured electron spin relaxation times vary from ~100 ps to ~10 ms depending on the experimental conditions and the type of quantum dot system studied,[4–12,18–21] hindering conclusive comparisons with existing theoretical models. Experiments designed to clarify the relative strength of the two primary spin relaxation mechanisms under different experimental conditions are needed.

In this work, we report room temperature time-resolved differential transmission and Faraday rotation measurements on InGaAs QDs. We compare the electron spin relaxation time for optical excitation in the GaAs barriers with that for direct excitation of the ground state of the QD, with all other experimental parameters fixed. The nonequilibrium phonon population generated in the former case enhances phonon-mediated electron spin relaxation, thereby isolating this spin decay channel. Our findings reveal a dramatic reduction of the electron spin relaxation rate for resonant pumping of the QD ground state, indicating that the optical excitation conditions have a critical influence on the observed spin kinetics. This may at least partially account for the wide variation in spin lifetimes reported to date in similar QD systems.[4–12] The spin lifetime we observe for optical pumping of the barriers is similar to several earlier reports, in which time-resolved photoluminescence techniques were used under similar nonresonant pumping conditions.[4,7–10] The long room temperature electron spin relaxation time we observe for resonant excitation (1 ns) is promising for the future development of a scalable quantum computation architecture based on the control of QD spins.

The self-assembled InGaAs QDs were grown by molecular beam epitaxy. The heterostructure consists of two layers of QDs, separated by 100 nm, within a 200 nm GaAs region that is bounded on both sides by AlAs diffusion barriers. The QD layers were formed from a deposition of 10 ML with average composition $In_{0.5}Ga_{0.5}As$. Atomic force microscopy and transmission electron microscopy indicate that the QDs have an areal density of 1×10^{10} cm^{-2}, a lateral size of 35 nm, and a height of 11 nm after covering with GaAs. Results of room temperature continuous-wave photoluminescence (CWPL) experiments on these QDs, shown in Fig. 1, reveal several confined optical transitions.

Time-resolved differential transmission and Faraday rotation experiments were performed using 150 fs pulses from a regenerative amplifier/optical parametric amplifier (OPA) laser system. Experiments were performed using both degenerate and nondegenerate pump-probe configurations with similar pump fluences ($100\ \mu J/cm^{-2}$). For the nondegenerate experiments, electron-hole pairs were excited in the GaAs barriers surrounding the QDs using 800 nm pump pulses from the regenerative amplifier. 1.31 μm pulses from

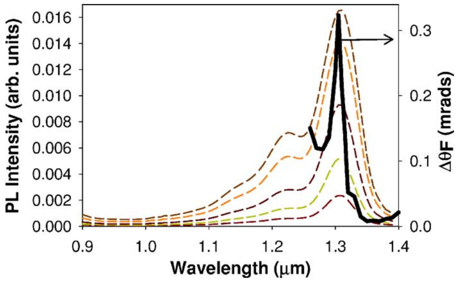

FIG. 1. (Color online) Results of spectrally resolved Faraday rotation measurements on InGaAs quantum dots for 800 nm optical pumping (solid curve). The peak Faraday rotation signal, which occurs at a probe delay of ≈1 ps, is shown for wavelengths near the ground state optical transition. Results of continuous-wave photoluminescence measurements for a range of optical powers between 20 and 300 mW are also displayed (dashed curves). All experiments were carried out at 295 K.

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[a] Electronic mail: kimberley.hall@dal.ca

[b] Present address: CREOL and FPCE-College of Optics and Photonics, University of Central Florida, Orlando, Florida 32816-2700.

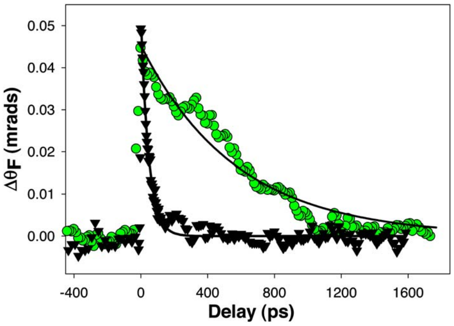

FIG. 2. (Color online) Results of Faraday rotation experiments at 295 K for optical excitation of the GaAs barriers using 800 nm pulses (triangles) and for direct optical pumping of the quantum dot ground state at 1.31 μm (circles).

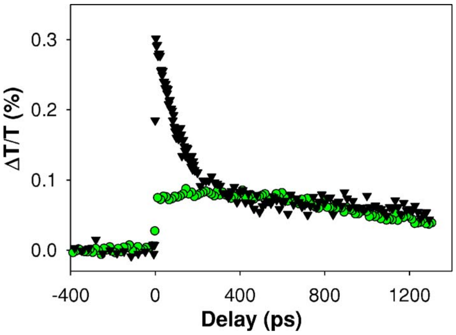

FIG. 3. (Color online) Results of time-resolved differential transmission measurements at 295 K for 800 nm pumping (triangles) and 1.31 μm pumping (circles).

the OPA were used as a probe to detect the population density and spin polarization of carriers in the ground state of the QDs. For degenerate experiments, both pump and probe pulses were 1.31 μm. In this case, electron-hole pairs are directly injected into the ground state of the quantum dots. In the Faraday rotation measurements, a photoelastic modulator was used to modulate the helicity of the circularly polarized pump beam at 50 kHz, and the probe beam was linearly polarized. The generation of spin-polarized carriers by the pump pulse leads to circular dichroism and rotation of the linear polarization of the probe. The pump-induced Faraday rotation was measured as a function of wavelength and probe delay using a polarization bridge. All experiments were carried out at room temperature.

Results of spectrally resolved Faraday rotation experiments under nondegenerate pumping conditions are shown in Fig. 1 (thick solid curve), together with the results of power-dependent CWPL experiments (dashed curves). For these measurements, spectral resolution was achieved by passing the wide-bandwidth OPA probe pulses through a monochromator following the sample. The maximum Faraday rotation signal, which occurs at a probe delay of ≈1 ps, is shown. The peak observed at 1.31 μm in Fig. 1, which coincides with the lowest energy peak observed in CWPL, is attributed to the optical response of carriers that have relaxed down into the ground state of the quantum dots following circularly polarized excitation in the GaAs barriers. Due to rapid hole spin relaxation at room temperature in GaAs,[8,9] the Faraday rotation signal is attributed to spin-polarized electrons. The onset of Faraday rotation in the first excited state of the quantum dots is also apparent in Fig. 1 at the short wavelength side of the probe spectrum.

The Faraday rotation signal from the QD ground state optical transition is shown versus probe delay in Fig. 2 for both degenerate and nondegenerate excitation conditions. For these spectrally integrated measurements, the broadband pulses from the OPA were passed through an interference filter centered at 1.31 μm with a full width at half maximum bandwidth of 10 nm. This spectral filtering, which increased the OPA pulse duration by ≈2 times, ensured selective optical excitation of the QD ground state for the degenerate experiments. The data in Fig. 2 indicate a much slower recovery of the Faraday rotation signal for degenerate pumping conditions in comparison with data for optical excitation of the GaAs barrier states.

The decay of the Faraday rotation signal fits well to a single exponential function for both degenerate and nondegenerate pumping conditions. Fits to the data in Fig 2 provide time constants of 560 ± 20 ps (degenerate pumping) and 47 ± 5 ps (nondegenerate pumping). The recovery of the Faraday rotation signal is due to the combined effects of spin relaxation and carrier recombination. The Faraday rotation signal is proportional to $N^{+} - N^{-}$, where $N^{\pm}$ are the optically excited populations of spin up (+) and spin down (−) electrons in the ground state of the quantum dots. The spin relaxation time T_1 may be estimated using[22]

$$\frac{1}{T_1} = \frac{1}{\tau_{\mathrm{FR}}} - \frac{1}{\tau_R}, \tag{1}$$

where τ_{FR} is the decay time of the Faraday rotation signal and τ_R is the carrier recombination time. Differential transmission experiments were performed under the same conditions as the Faraday rotation measurements to obtain information about the spin-independent carrier kinetics. For these experiments, the pump and probe were linearly polarized. The pump-induced change in transmission, expressed as a percentage of the unsaturated probe transmission, is shown versus delay in Fig. 3. After the first few hundred picoseconds, the recombination dynamics are similar for degenerate and nondegenerate excitation geometries, revealing a recombination lifetime of 1–2 ns. For earlier time delays, a much faster (120 ps) recovery is observed in the data for 800 nm pumping, attributed to Auger recombination involving the large density of carriers ($\sim 10^{18}\ \mathrm{cm}^{-3}$) in the GaAs barriers.[23,24] Using these results, together with the decay times observed in the Faraday rotation measurements, we obtain $T_1 = 1$ ns for optical pumping at 1.31 μm and $T_1 = 75$ ps for optical pumping at 800 nm.

Due to the spin-orbit interaction, the confined energy states within the quantum dots have a mixed spin character. As a result, scattering with phonons can provide an efficient spin relaxation channel.[9,13,14] For optical excitation of the GaAs barriers, electron-hole pairs must lose ~600 meV of excess energy before reaching the ground state of the quantum dots. Electron and hole energy relaxation will involve a cascade of phonon emission processes within the barrier, wetting layer, and QD states.[25] This relaxation will lead to a nonequilibrium phonon population[26] that will enhance spin relaxation via phonon scattering, providing a way to highlight this spin decay channel while having little effect on the rate of spin flip via hyperfine coupling to the lattice nuclei. A similar nonequilibrium phonon population was recently found to enhance Mn spin heating in diluted magnetic semiconductor quantum dots.[27] Our observation of an enhancement in the electron spin relaxation time by an order of magnitude with resonant optical excitation of the QD ground state suggests that spin relaxation is strongly dominated by scattering with this nonequilibrium phonon population for nondegenerate pumping conditions. This is consistent with earlier theoretical estimates of spin relaxation rates via virtual scattering with phonons in similar quantum dots.[9] The spin relaxation time we observe for nondegenerate pumping conditions is similar to that observed in time-resolved photoluminescence studies on InAs quantum dots, in which excitation was carried out by optically pumping the GaAs barriers,[4,7–10,18–21] lending further support to the conclusions drawn here. Using Hanle effect measurements, Epstein *et al.* observed faster spin relaxation for GaAs barrier excitation compared to pumping the InGaAs wetting layer,[19] findings that are also consistent with spin scattering with nonequilibrium phonons. Long spin relaxation times (~1 ns) were previously observed at low temperatures (40 K and below) in InAs quantum dots for strictly resonant excitation of the quantum dot ground state using PL techniques[6] and four wave mixing experiments.[28] However, the findings we report here provide the first comparison of electron spin lifetimes for vastly different carrier excess energies *in the same quantum dots*.

In summary, time- and wavelength-resolved Faraday rotation studies on self-assembled InGaAs quantum dots reveal a strong (≈10 times) enhancement of the electron spin relaxation time with resonant optical pumping of the quantum dot ground state compared to optical pumping of the GaAs barriers. This finding highlights the dramatic influence that experimental conditions can have on the measured spin lifetimes, a result that may partially account for the wide variation in spin lifetimes reported to date in similar QD systems.[4–12] Our measurements also indicate that, under appropriate conditions, scattering with phonons can strongly dominate spin relaxation in quantum dots. The long room temperature spin lifetime we observe for resonant excitation is promising for the prospect of future applications of quantum dot spin q bits in quantum computation.

This research is supported by the Canadian Foundation for Innovation and the Natural Sciences and Engineering Research Council of Canada.

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