Hole spin relaxation in neutral InGaAs quantum dots: Decay to dark states

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Hole spin relaxation in neutral InGaAs quantum dots: Decay to dark states

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The authors report measurements of hole spin relaxation in neutral InGaAs quantum dots using polarization-dependent time-resolved photoluminescence experiments. The single-particle hole spin relaxation was isolated from other spin flip processes in the electron-hole system by detecting the initial transfer of population from optically active to dark states. The results indicate that electron-hole exchange interactions play a negligible role in the carrier spin kinetics, and are consistent with a mechanism of hole spin relaxation via phonon-mediated virtual scattering between confined quantum dot states. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2437063]

Spin states in semiconductor quantum dots (QDs) may form a suitable q bit for storing and manipulating quantum information in a scalable architecture.[1–3] This promise has led to a comprehensive research effort focused on electron spin dynamics in quantum dots, including measurements over a wide range of experimental conditions and in different types of quantum dots.[4–12] The measured spin relaxation times vary considerably, which has stimulated theoretical work to illuminate the operative spin relaxation mechanisms. Two primary processes have been identified: phonon scattering in the presence of spin-orbit coupling[10,13,14] and the hyperfine interaction with the lattice nuclear spins.[15]

In contrast, much less is known about the spin dynamics of holes in quantum dots. Theoretical treatment of hole spin relaxation, which is mediated by the spin-orbit interaction,[16] is complicated by the substructure of the valence band and the strong influence of interband coupling.[13,17–20] Measurement of hole spin dynamics is also more subtle than for electrons because, in the case of neutral quantum dots, these dynamics are not accessible using the standard approach of detecting the degree of circular polarization of the QD photoluminescence.[13,21] Hole spin decay was recently measured in quantum dots with a single excess electron by using the trion feature as a monitor[21,22] and in strongly negatively charged quantum dots.[10] In both cases, the ground electron level is occupied with both spins, so that the ground state trion luminescence indicates the spin of the optically injected hole. In these studies, the influence of the excess negative charge on the electronic structure and the hole spin kinetics is not known, making measurements in neutral quantum dots desirable. Such measurements would also elucidate the role of electron-hole exchange interactions in the carrier spin dynamics since these interactions vanish for trion-bound electrons and holes.[21,22]

Here we report measurements of hole spin relaxation in neutral self-assembled InGaAs/GaAs quantum dots. Instead of relying on quantum dot charging to access the hole spin information, we detect the initial transfer between bright and dark excitons following short circularly polarized optical excitation. Our findings indicate that single-particle scattering processes dominate the spin relaxation kinetics, with electron-hole exchange interactions playing a negligible role. The measured lifetimes suggest that electrons and holes lose their spin polarization through the spin-orbit interaction and phonon-mediated virtual scattering between quantum dot states of mixed spin.

Under circularly polarized optical excitation into wetting layer or barrier states, the bimolecular capture of spin-polarized electrons and holes will cause an imbalance in the occupation of the $|\pm 1\rangle$ optically active exciton states in the quantum dots (see the inset in Fig. 1). This nonequilibrium condition will decay through either consecutive single-particle spin flip processes of the electrons and holes (which involve an intermediate transition to a dark state) or simultaneous spin flip of the electron and hole within the exciton through the electron-hole exchange interaction. The degree of circular polarization of the quantum dot luminescence (ρ) is typically used as a monitor of the spin relaxation of carriers in the quantum dot ground state; however, ρ does not allow one to distinguish between simultaneous and sequential spin flip processes for the electron and hole spin. The decay time of ρ is determined only by the slowest spin relaxation process.[13,21]

Instead, one may exploit the transitions between bright and dark exciton states to access the individual carrier spin kinetics in neutral quantum dots. In this approach, which was first applied in intrinsic quantum wells,[23] one measures the ratio of the total luminescence intensities for linear (π) and circularly polarized excitation (σ_+),

$$R = \frac{S^+_{\sigma+} + S^-_{\sigma+}}{S^+_{\pi} + S^-_{\pi}}, \quad (1)$$

where $S^{\pm}_{\sigma+}$ ($S^{\pm}_{\pi}$) are the left and right circular polarization components of the photoluminescence under σ_+ (π) optical excitation. Other parameters governing the excitation condi-

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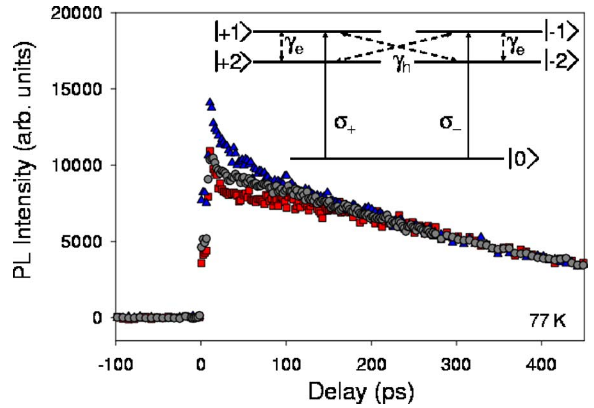

FIG. 1. (Color online) Results of polarization-dependent time-resolved PL experiments on InGaAs QDs at 77 K. Triangles (squares) indicate the dynamics of the σ_+ (σ_-) components of the photoluminescence under excitation with σ_+. Circles indicate results for linearly polarized excitation, in which the same linear component of the photoluminescence is detected. Inset: Selection rules for the ground state optical transition in the quantum dots, which is primarily heavy-hole in character (Refs. 25 and 26). The states $|J_z\rangle$ indicate the total angular momentum projection of the exciton spin along the growth direction of the quantum dots. The degenerate optically active states $|\pm 1\rangle$ are separated from the dark states $|\pm 2\rangle$ by the electron-hole exchange interaction energy, which is typically ~100–200 μeV (Refs. 25 and 26) and has been exaggerated for clarity. Single-particle hole (γ_h) and electron (γ_e) spin flip transitions, which connect dark and bright exciton states, are indicated by dashed lines.

tions (intensity and wavelength) are kept constant. Measurement of R isolates the decay of an excess population of optically active states ($|+1\rangle$ under σ_+ excitation) caused by the first single-particle spin flip processes, assumed to be due to holes.[10,21] R is unaffected by electron-hole exchange interactions that cause scattering between the $|\pm 1\rangle$ optically active states *or* to the slower single-particle spin relaxation of electrons, allowing hole spin relaxation to be studied independently of any other spin decay processes. This allows the relative importance of electron-hole exchange interactions and other spin flip mechanisms to be assessed in neutral quantum dots. This information has important implications for recent studies of optical[11,21] and electrical[22] control of trion spin dynamics.

The InGaAs/GaAs self-assembled quantum dots[24] were formed by depositing 10 ML of $In_{0.5}Ga_{0.5}As$, resulting in cylindrically symmetric, lens-shaped dots with a lateral size of ~35 nm and a height of ~11 nm after covering with GaAs. The structure contains two layers of quantum dots, each with an areal density of ~10^{10} cm^{-2}. AlAs carrier confinement layers separated by 0.2 μm were grown above and below the quantum dot layers. Four well-defined peaks separated by 60–70 meV are observed in continuous-wave photoluminescence measurements. Polarization-dependent time-resolved photoluminescence experiments[27] were performed using 100 fs, 870 nm pulses from a Ti:sapphire laser, which injects electron-hole pairs into low energy states in the GaAs barriers. The photoluminescence from the QD ground state optical transition is time resolved through sum frequency generation in a $KNbO_3$ crystal with a second Ti:sapphire pulse. For circularly polarized excitation, a quarter wave plate in the photoluminescence path, in conjunction with the type I phase matching conditions in the $KNbO_3$ crystal, provides discrimination between the left and right circular components of the resulting photoluminescence. The temperature dependent absorption coefficient at 870 nm was

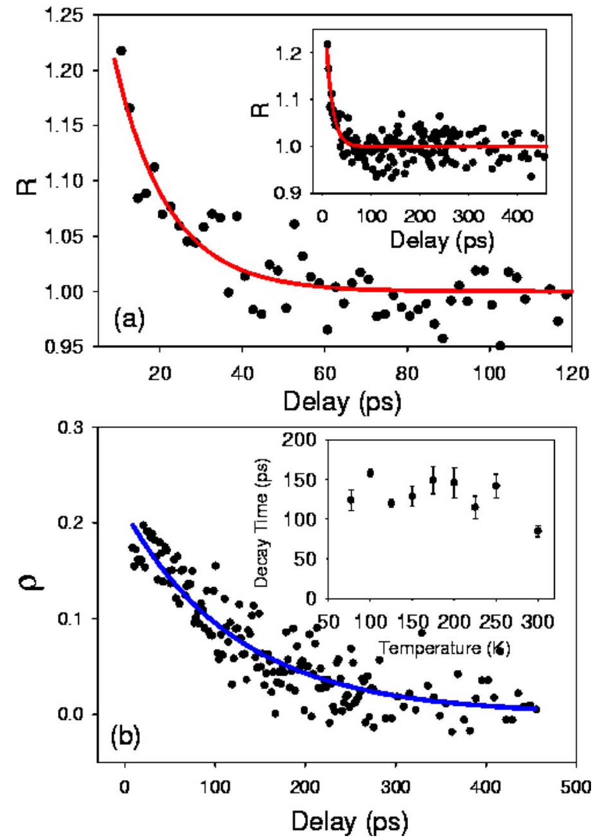

FIG. 2. (Color online) (a) Time dependence of the ratio R. The decay of R to unity indicates spin relaxation of holes in the QDs. The exponential fit (solid curve) indicates a decay time of 15±4 ps. Inset: Same data for larger optical time delays. (b) Decay of the degree of circular polarization (ρ), indicating the spin dynamics of electrons in the QDs. The exponential fit (solid curve) provides a decay time of 125±15 ps. Inset: Temperature dependence of the exponential fit results for ρ.

estimated from photoluminescence excitation experiments. For all experiments, the excitation density was ~0.5 electron-hole pairs per quantum dot.

Time-resolved photoluminescence from the ground state optical transition in the quantum dots is shown in Fig. 1. The convergence of the photoluminescence intensity for the circular and linear polarization excitation geometries within 200 ps indicates that all spin information within the electron-hole system has decayed on this time scale. The single-particle hole spin relaxation time (τ_h) is extracted from these data by evaluating R using Eq. (1) versus pulse delay, and is shown in Fig. 2(a). For optical pumping of carriers into the bulk GaAs barrier, R will have the form

$$R(\tau) = 1 + 0.5e^{-t/\tau_h}. \quad (2)$$

Here the coefficient of the exponential results from the optical selection rules for a bulk III-V semiconductor,[28] and assumes that no spin information is lost prior to carrier capture into the quantum dots. This point is discussed further below. The data in Fig. 2(a) show that R decays to unity in under 50 ps, indicating *complete* decay of hole spins on this time scale. The exponential fit in Fig. 2(a) corresponds to $\tau_h = 15\pm 4$ ps. The observation of rapid single-particle hole spin relaxation implies that the electron-hole exchange interaction does not contribute significantly to the carrier spin dynamics in these quantum dots. Using this result, we can infer the electron spin relaxation time from a measurement of ρ, as shown in Fig. 2(b). We extract a decay time of 125±15 ps,

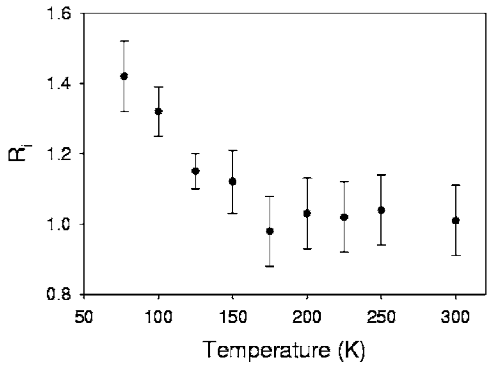

FIG. 3. Temperature dependence of the initial value of R (extrapolated to zero time delay) obtained from the exponential fit results.

which is relatively insensitive to temperature, as shown in the inset to Fig. 2(b).[29]

The temperature dependence of the initial value of R obtained from the fit results is shown in Fig. 3. A value less than 1.5 indicates some loss of hole spin polarization prior to and/or during hole capture into the quantum dots. For temperatures above 175 K, the initial value of R is unity, indicating that captured holes have no residual spin polarization. For these experiments, electron-hole pairs were optically injected into low energy states in the bulk GaAs barriers. Hole capture into the quantum dots is expected to occur on a subpicosecond time scale.[10,24,30] Hole spin relaxation in bulk III-V semiconductors is rapid due to the strong degree of spin mixing in the valence band states and the efficiency of momentum scattering processes. A hole spin relaxation time of 110 fs was recently measured in bulk GaAs at 300 K.[31] The spin lifetime is expected to increase with decreasing temperature due to the reduced occupation of large-wavevector hole states, which have a larger degree of spin mixing. This is consistent with our observation of a decrease in the spin polarization of captured holes with increasing temperature.

Electron and hole spin dynamics were recently measured in strongly charged InAs quantum dots.[10] The dominant mechanism of spin decay for both electrons and holes was attributed to phonon-mediated virtual scattering between quantum dot states of mixed spin. In the experiments we report here on *neutral* InGaAs quantum dots, the electron spin relaxation time is similar to these earlier results in charged quantum dots, but the hole spin lifetime is somewhat smaller ($\approx$2 times). This finding is expected from the larger size of the quantum dots studied here, which leads to a stronger degree of mixing in the quantum dot hole states.[12,13,17] This result therefore lends further support to a mechanism of single-particle spin decay based on phonon-mediated virtual scattering.[10]

In summary, we have measured the hole spin relaxation time in InGaAs quantum dots using time-resolved photoluminescence techniques. In contrast to existing studies on charged quantum dots, single-particle hole spin decay was isolated from other spin relaxation processes in our *neutral* quantum dots by detecting the initial transfer of holes out of optically active states. This method allows single-particle and exciton spin relaxation to be distinguished, and our findings indicate that electron-hole exchange interactions have a negligible influence on the carrier spin dynamics. Our measured spin relaxation times for electrons and holes are consistent with spin decay via virtual phonon scattering between quantum dot levels, which have a mixed spin character due to the spin-orbit interaction.

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