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We report direct measurement of the spin dynamics of electrons and holes in self-assembled InAs quantum dots (QDs) through polarization-sensitive time-resolved photoluminescence experiments on modulation-doped quantum dot heterostructures. Our measured hole spin decay time is considerably longer than in bulk and quantum well semiconductor systems, indicating that the removal of near degenerate hole states with different spin quantization axes through three-dimensional confinement slows hole spin relaxation in semiconductors. The electron and hole spin decay times we observe (electrons: 120 ps; holes: 29 ps) are consistent with spin relaxation via phonon-mediated virtual scattering between the lowest two confined levels in the QDs, which have a mixed spin character due to the spin–orbit interaction. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1857067]

Spin relaxation kinetics in semiconductor quantum dots (QDs) have gained considerable interest in recent years, and have been studied using a variety of optical techniques, including Hanle experiments,[1] pump probe methods,[2–4] and time-resolved photoluminescence.[5–8] Such studies are motivated by proposals for a scalable quantum computation architecture based on manipulation of QD spins.[9–11] The electron spin relaxation time has been found to vary over several orders of magnitude depending on the type of QD system studied[2–5] and even in similar QD structures depending on experimental parameters such as temperature and optical excitation energy.[1,5,6] These studies have stimulated several recent theoretical works aimed at identifying the operative relaxation mechanism for the QD electron spins.[12–14]

In contrast, much less is known about the spin kinetics of holes in QDs. Hole spin dynamics will play a crucial role in quantum computation schemes in which information is stored in spin-polarized excitons,[10,11] and have recently been shown to facilitate optical manipulation of electron spins in *n*-doped QDs through the electron–hole exchange interaction.[2] Hole spin kinetics were first studied several years ago in semiconductor quantum wells using an *n*-type modulation doped heterostructure to provide isolation of the hole spin decay,[15] and were examined in bulk recently using a nondegenerate pump probe experiment in which hole spin states were interrogated through intervalence band absorption.[16] Due to the spin–orbit interaction, hole states in III–V semiconductors have a mixed spin character. In this case, the spin quantization axis varies from state to state and any scattering process (with, e.g., phonons, other carriers, defects, etc.) will contribute to spin relaxation. Due to the large density of states near the band edge in bulk and quantum well semiconductors, such scattering processes are rapid, providing short hole spin flip times in the range of a few picoseconds or less.[15,16] In contrast, three-dimensional quantum confinement leads to a fully discrete energy spectrum, thereby eliminating near degenerate hole states with different spin quantization axes. The hole spin lifetime is therefore expected to be strongly enhanced in semiconductor QDs relative to higher dimensional semiconductor systems.

In this letter, we report the direct measurement of electron and hole spin dynamics in self-assembled InAs QDs. Separate measurement of the spin kinetics of electrons and holes is accomplished through the application of time-resolved photoluminescence (TRPL) experiments to modulation doped (*p* or *n*) QD heterostructures. The high doping density we have used, which corresponds to 20 carriers per quantum dot, results in occupation of the lowest hole (electron) levels in the *p* (*n*) modulation doped QDs over a wide temperature range, thereby allowing us to clearly isolate the ground state spin kinetics of the individual carrier species, in contrast to existing studies involving trion dynamics.[17] Our measured spin relaxation time (T_1) for electrons (120 ps) is only moderately longer than that for holes (29 ps), in stark contrast to the situation in higher-dimensional semiconductors.[15,16,18–21] We calculate the electronic structure of our InAs QDs using eight-band strain-dependent $k \cdot p$ theory. Together with the observation of weak or no temperature dependence in the carrier spin kinetics, our calculations suggest that a spin flip process mediated by virtual scattering with a nonequilibrium phonon population is responsible for our measured spin relaxation times for both electrons and holes. Such a mechanism is also consistent with the earlier observation of differing *electron* spin kinetics for conditions of resonant or nonresonant optical excitation of the QD ground state.[5,6] The hole spin decay time we observe is considerably longer than in bulk semiconductors[16] and quantum wells,[15,18] indicating that three-dimensional quantum confinement slows hole spin relaxation in semiconductors.

The modulation doped InAs/GaAs QD heterostructures were grown by molecular beam epitaxy. A corresponding undoped QD structure was also grown consecutively under identical conditions. Each sample contains a single layer of

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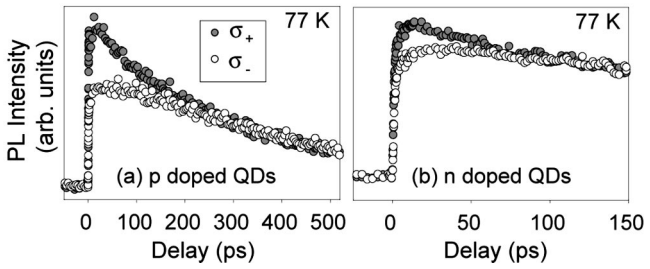

FIG. 1. Results of polarization-sensitive TRPL experiments on modulation-doped QDs: (a) *p*-doped; (b) *n*-doped. These data are used to evaluate the degree of circular polarization of the photoluminescence (ρ).

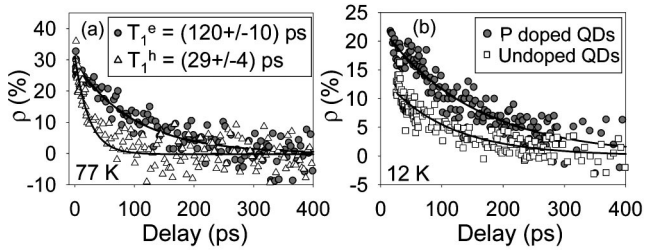

FIG. 2. (a): Degree of circular polarization (ρ) vs time delay for *p*-doped QDs (closed circles) and *n*-doped QDs (open triangles) at 77 K. (b): Circular polarization kinetics at 12 K, showing a comparison of results for *p*-doped and undoped QDs. For the data in (b), exponential fits provide decay times of (150 ± 15) ps (*p*-doped QDs) and (110 ± 20) ps (undoped QDs). The initial value of ρ is smaller for the undoped QDs due to slower carrier capture from the InAs wetting layer, (Ref. 24), which leads to a larger degree of spin decay prior to capture.

QDs in the center of a 30 nm layer of GaAs, which is clad 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 nm in diameter and 3 nm in height, with an areal density of 3×10^{10} cm^{-2}. For the modulation doped structures, the QDs were covered with 12 nm of undoped GaAs, followed by 3 nm of GaAs doped with Si (*n*-doped QDs) or Be (*p*-doped QDs) at a density of 2×10^{18} cm^{-3}. The associated free carriers (approximately 20 per QD) accumulate in the QD states. This high doping density ensures that, prior to optical excitation, the lowest electron levels (*n*-doped QDs) or hole levels (*p*-doped QDs) will be occupied over a wide temperature range with carriers that have a random spin orientation. Recombination of optically injected spin-polarized electrons (*p*-doped QDs) or holes (*n*-doped QDs) with the unpolarized background population allows us to separately interrogate the spin dynamics of the individual carrier species.[22]

In the TRPL experiments, spin-polarized electron–hole pairs are injected into the InAs wetting layer using circularly polarized (σ_+) 100 fs, 1.40 eV pulses from a Ti:sapphire laser. The photoluminescence from the QD ground state optical transition is time-resolved through sum frequency generation in a $KNbO_3$ crystal with a second linearly polarized Ti:sapphire pulse. A quarter waveplate in the photoluminescence path provides discrimination between the σ_+ and σ_- components of the PL, and the carrier spin polarization is given by the emission polarization.[23,24] For all experiments, the optically injected carrier density is $\leqslant1$ electron–hole pair per QD, corresponding to a factor of 20 below the background doping level.[25]

Results of polarization-dependent TRPL experiments on the modulation doped QDs are presented in Fig. 1. Due to the spin unpolarized background population in these modulation doped QDs the degree of circular polarization reflects the spin dynamics of the optically injected holes (*n*-doped QDs) *or* electrons (*p*-doped QDs). In contrast, for TRPL experiments on undoped QDs, emission results from captured electron–hole pairs, and therefore reflects only the spin kinetics of the carrier with the longer spin relaxation time, which is typically assumed to be the electrons. The strong circularly polarized emission we observe for both *n*-doped and *p*-doped QDs, which is positive for all temperatures and optical time delays, indicates that both electrons and holes retain spin information during capture from the InAs wetting layer and relaxation to the QD ground state. The ultrafast carrier capture in these modulation doped QD heterostructures[24,26] contributes to this efficient spin capture process. The rapid capture of spin-polarized holes (which occurs in approximately 1 ps at 77 K)[24] is especially crucial due to the fast relaxation of hole spins in barrier and wetting layer states.[27]

Single exponential fits to the decay of the circularly polarized emission [Fig. 2(a)] provide QD ground state spin relaxation times of 120 ps for electrons and 29 ps for holes. The electron spin relaxation time (T_1^e) is unchanged from 77 to 300 K, with a slightly longer lifetime found in experiments at 12 K (150 ps). In *n*-doped QDs, the degree of circular polarization decreases sharply with increasing temperature due to stronger relaxation of hole spins prior to capture into the QDs. This restriction limited the hole spin relaxation measurements to temperatures $\leqslant120$ K. The hole spin lifetime (T_1^h) remains constant between 77 and 120 K and increases to 45 ps at 12 K, similar to the temperature dependence of the electron spin kinetics. The decay rate of the circularly polarized emission is similar for the *p*-doped QDs and the corresponding undoped QD structure. [See Fig. 2(b) for results at 12 K; the corresponding comparison at 300 K was reported earlier.[24]] The dynamics of the circularly polarized emission from the undoped QDs reflect the electron spin kinetics. Our observations therefore indicate that T_1^e is unaffected by the large built-in hole population in the *p*-doped QDs, suggesting that electron–hole exchange scattering does not influence the carrier spin dynamics.

The observation of comparable electron and hole spin decay times (which only differ by a factor of 4) is in stark contrast to the situation in higher-dimensional semiconductors,[15–17,19–21] and provides insight into the mechanism governing the carrier spin flip kinetics. The rapid relaxation of hole spins compared to electrons in bulk semiconductors (e.g., in bulk GaAs at 300 K, $T_1^{\text{hole}}\sim100$ fs,[16] while $T_1^{\text{electron}}\geqslant50$ ps[21]) is due to the degeneracy between the heavy-hole and light-hole valence bands, which leads to strong mixing of the hole spin states. Confinement lifts this degeneracy in semiconductor quantum wells, leading to weaker hole spin mixing,[15,17] however rapid quasielastic scattering processes still provide an efficient spin flip channel for holes. (For example, in quantum wells, T_1^{hole} is typically a few picoseconds at cryogenic temperatures.[15,17]) Three-dimensional quantum confinement in a semiconductor QD leads to a fully discrete energy spectrum, which drastically reduces the availability of energy-conserving scattering processes; In self-assembled InAs QDs, the hole states are separated by tens of milli-electron-volts. T_1^h in QDs is there-

fore expected to be enhanced relative to higher-dimensional semiconductors, consistent with the results in Fig. 2(a).

In order to gain further insight into the carrier spin kinetics, we calculated the electronic structure of the InAs/GaAs QDs using eight-band strain dependent $k\cdot p$ theory.[28–30] The QD dimensions chosen matched the measured values from atomic force microscopy and transmission electron microscopy on similar QD samples (height 3 nm; diameter 25 nm). Due to the spin–orbit interaction, the electron and hole states in the QDs have a mixed spin character. As a result, scattering from the pseudospin up ground state to the pseudospin down ground state is possible via virtual absorption of a phonon to the first excited state. The spin relaxation rate is given by

$$\frac{1}{T_1} \propto \frac{\sum_{s=\uparrow,\downarrow} |M(g\uparrow,es)M(es,g\downarrow)|^2}{(\Delta E_{g,e})^2}, \tag{1}$$

where $M(gs,es')$ are the matrix elements of the electron–LO phonon interaction between the ground and first excited energy level, $s,s'=\uparrow,\downarrow$ are the pseudospin states, and $\Delta E_{g,e}$ is the ground to excited state energy separation. Using our calculated QD states, we estimate $M(g\uparrow,e\downarrow)/M(g\downarrow,e\downarrow) \sim 0.05$ for these QDs for both electrons and holes, leading to hole spin flip times of the order of 50 ps. This is consistent with calculated hole dephasing times $\sim$100 fs from a similar, *spin-conserving* virtual phonon scattering process.[31] Furthermore, the calculated $\Delta E_{g,e}$'s are 54 meV (electrons) and 24 meV (holes), providing an expected ratio between the electron and hole spin flip times of $T_1^{\text{electron}}/T_1^{\text{hole}} \approx (54/24)^2 = 5.1$, in reasonable agreement with the measured ratio of 4.1. The relaxation rate for both electrons and holes exhibits weak or no temperature dependence, thus a nonequilibrium phonon population may contribute to the virtual scattering process leading to spin decay. Such a hot phonon population would be generated during carrier relaxation within the InAs wetting layer and through possible multiphonon emission processes within the QDs. Virtual scattering with a nonequilibrium phonon population would also account for the earlier observations in undoped InAs QDs of much faster *electron* spin relaxation for nonresonant optical excitation into the GaAs barrier states compared to findings for optical injection of spin-polarized carriers directly into the ground state of the QDs.[5,6,32]

We note that the hyperfine interaction between the electron and nuclear spins, which has been the focus of recent theoretical work,[13,14] is inoperative for holes. The similar spin relaxation times and electronic structure characteristics for holes and electrons in these InAs QDs favor a common mechanism for spin decay for both carriers, such as the phonon-mediated virtual scattering process discussed here.

In summary, we have separately measured the spin dynamics of electrons and holes in modulation doped self-assembled InAs QDs using time-resolved photoluminescence experiments. Our findings indicate that the discrete energy level structure in QDs, which eliminates scattering among near degenerate states with different spin quantization axes, slows hole spin relaxation compared to bulk semiconductors and quantum wells. Our electronic structure calculations suggest that virtual scattering processes between the confined QD states, which have a mixed spin character due to the spin–orbit interaction, dominates the spin decay process for both electrons and holes.

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