Nuclear spin pumping under resonant optical excitation in a quantum dot

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We demonstrate nuclear spin pumping in a single InGaAs/GaAs dot embedded in a p-i-n diode in the regime of resonant optical excitation of spin-polarized electron-hole pairs in the lowest energy states of the dot. A nuclear spin pumping mechanism is proposed relevant to the regime of high electric field where carriers escape from the dot by tunneling. The degree of nuclear spin polarization is shown to increase strongly with the applied electric field, controlling the carrier tunneling from the dot, since at low electric fields the dot is blocked for re-excitation due to the slow hole escape. © 2008 American Institute of Physics. [DOI: 10.1063/1.2958221]

In III-V semiconductor quantum dots (QDs) the hyperfine interaction between the electron and nuclear spins leads to dynamic nuclear polarization (DNP) that has significant influence on the properties of confined electrons. F-12 This dynamic process leads to a buildup of a significant nuclear polarization leading to occurrence of Overhauser magnetic fields and, as a result, modification of the electron energy spectrum.5,6,8-11 Overhauser fields, acting on the electron spin only, have a direct effect on the electron spin dynamics.7,13

In this work we consider a regime where the spin-polarized electrons are optically excited resonantly into the lowest energy states of an InGaAs/GaAs dot. Recently resonant optical excitation of QDs has been widely employed in ultrafast coherent control based on the Stark shift bias whilst fixing the laser excitation wavelength. This technique is based on the Stark shift [see Fig. 1(a)] exhibited by the Zeeman doublet excited with polarized light is shown by the common ultimate goal of all these experiments is manipulation of the electron spin in nanostructures. The combination of optical excitation with the control of the magnetic environment of a single electron spin is therefore of key importance.

Here we demonstrate voltage control of the nuclear spin in a resonantly optically pumped InGaAs dot in high magnetic field. A regime is considered where both spin-polarized holes and electrons escape from the dot via tunneling. We achieve tuning of the degree of nuclear spin polarization in a range of 0%-20% in external field Bext=4–5 T by changing the electric field controlling the rates of carrier tunneling from the dot. We propose a mechanism of the nuclear spin pumping on the dot, that is based on electron-tunneling-assisted spin flip-flops: the electron tunneling is accompanied by a simultaneous spin exchange flip-flop between a localized electron and one of the nuclei on the dot. We discuss that the nuclear spin pumping is limited by the slow tunneling of the hole at low electric fields reducing the rate of re-excitation of the dot with a spin-polarized electron.18,19

The p-i-n structure investigated was grown using molecular beam epitaxy on a n+ GaAs substrate. On top of n+ GaAs, undoped layers of AlGaAs (25 nm) and GaAs (400 nm) were deposited and a single dot layer (dot density of 5 × 109 cm−2) was then grown. The structure was completed with undoped layers of GaAs (180 nm) and AlGaAs (25 nm) and the top p+ contact formed with 300 nm of Be-doped GaAs. The structure was processed into diodes with the top surfaces covered with an opaque 80 nm thick Al mask with 400 nm clear apertures opened for optical access to individual QDs.

Photocurrent (PC) experiments were performed using a single-frequency cw Ti:Sapphire laser for excitation. The PC signal arises from the tunneling to the contacts of electron-hole pairs excited resonantly into the ground state of the dot. The PC measurements were performed by tuning the applied bias whilst fixing the laser excitation wavelength. This technique is based on the Stark shift [see Fig. 1(a)] exhibited by the celebrated exciton Zeeman doublet.

FIG. 1. (a) Stark shift of the ground state exciton peak. (b) Exciton Zeeman peak energies as functions of Bext. (c) PC spectra measured with circularly polarized excitation into the ground exciton state of an individual dot at Bext=5 T. The Zeeman doublet excited with σ+-polarized light is shown with solid (open) symbols and ΔE⁺ (ΔE⁻) denotes the corresponding exciton Zeeman splitting.

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the QD exciton states under applied electric field. This allows a direct conversion of the voltage [related to the electric field as \( F = (V + 1.5)/d \)] at which the PC signal is detected into the corresponding spectral position.

Figure 1(b) shows the energies of the two Zeeman split exciton states measured in PC as a function of the magnetic field. As seen in Fig. 1(c) for spectra measured at \( B_{\text{ext}} = 5 \) T the low (high) energy peak has the strongest intensity when excited with \( \sigma^- \) (\( \sigma^+ \)) circularly polarized excitation. Figure 1(e) shows that the splitting between the two Zeeman peaks is dependent on the sign of the circular (\( \sigma^- \) or \( \sigma^+ \)) polarization of excitation. At \( B_{\text{ext}} = 5 \) T the splitting under \( \sigma^- \)-polarized excitation, \( \Delta E = 398 \) \( \mu \)eV, while for \( \sigma^+ \) polarization \( \Delta E = 465 \) \( \mu \)eV is measured. The dependence of the Zeeman splitting on the polarization of excitation is a signature of DNP and is due to the build-up of the Overhauser field, \( B_N \), acting on the electron spin. In the presence of the Overhauser field, the exciton Zeeman splitting can be expressed as \( \Delta E^\pm = g_\lambda \mu_B B_{\text{ext}} \pm g_e \mu_B (B_{\text{ext}} \pm B_N) \), where \( g_e \) (\( g_\lambda \)) are the electron (hole) \( g \)-factors and \( \mu_B \) is Bohr magneton. The magnitude of the effective nuclear magnetic field, \( B_N \), is a linear function of the degree of nuclear spin polarization on the dot, \( S \). The sign of \( B_N \) observed in Fig. 1(e) is in agreement with that previously reported for InGaAs dots when photoexcited electrons are involved in the spin transfer.\(^{8,9,11,12}\) The Overhauser field \( B_N \) is parallel (antiparallel) to \( B_{\text{ext}} \) when \( \sigma^+ \) (\( \sigma^- \)) excitation is used.

Figure 2(a) shows the power dependence of the shift, \( \Delta \), between the \( \sigma^- \) (high energy) Zeeman peaks excited with \( \sigma^+ \) and \( \sigma^- \) polarized light for a dot at \( B_{\text{ext}} = 4 \) T. The peak energies in Fig. 2(a) and Fig. 3(a) are obtained by the fitting of the spectra with two Lorentzians [see an example of the fitting curves in Fig. 2(c)]. In Fig. 2(a) the high energy peak of the Zeeman doublet corresponds to a bias of 4.4 V. A clear saturation of \( \Delta \) is observed with increasing incident power (\( P \)). In Fig. 2 the maximum \( \Delta = 54 \) \( \mu \)eV, which corresponds to \( B_N \approx 1.86 \) T for the electron \( g \)-factor \( g_e = 0.55 \). For a typical In\(_{0.6}\)Ga\(_{0.4}\)As dot \( \Delta = 54 \) \( \mu \)eV corresponds to nuclear polarization \( S \approx 22\% \).

Figure 2(b) shows the integrated PC intensity, \( I_{\text{int}} \), as a function of \( P \). Even at high bias of 4.4 V the rate of increase in \( I_{\text{int}} \) with \( P \) is reduced after \( P \approx 150 \) \( \mu \)W since the ground state absorption is weakened by the increasing dot occupancy.\(^{13,14}\) Stronger saturation effects are observed at lower biases.

Figure 2(c) and 2(d) shows PC spectra of the high energy peak of the Zeeman doublet measured at 4.6 and 2.9 V, respectively. Spectra measured with \( \sigma^+ \) (\( \sigma^- \)) polarization are shown with solid (open) symbols. High pumping power is employed, in the range for which a weak dependence of \( \Delta \) on power is observed in Fig. 2(a). Gray lines in Fig. 2(c) show the line shapes of individual Zeeman components obtained from the fitting of each PC spectrum with two Lorentzians. Figures 2(c) and 2(d) show that the degree of nuclear polarization, \( S \), can be controlled by the applied bias. As seen in Figs. 2(c) and 2(d), at 4.6 V \( \Delta \) considerably exceeds that at 2.9 V: \( \Delta (4.6 \text{ V}) = 40 \mu \text{eV}, \Delta (2.9 \text{ V}) = 13 \mu \text{V} \).

The bias dependence of \( \Delta \) at \( B_{\text{ext}} = 5 \) T is summarized in Fig. 3(a). In the whole range of biases in Fig. 3(a) the measurements are performed at a high \( P = 500 \) \( \mu \)W. As seen from the figure the Overhauser shift \( \Delta \) exhibits a nearly linear dependence on the bias and varies from \( \approx 6 \) to 40 \( \mu \)eV in the range of 2.2–4.4 V. The variation of \( \Delta \) corresponds to increase of \( S \) in a range of 0%–17% controlled by applied electric field. As seen in Fig. 1(a) similar but smaller shift is observed for the low energy peak of the Zeeman doublet. A smaller magnitude of this shift is related to a lower bias at which the low energy peak is measured in each PC spectrum.

We now discuss the origin of the bias dependence observed in Fig. 3(a). A significant difference with previously reported nuclear spin pumping for nonresonant optical exci-
tiation lies in the nonradiative carrier escape in the conditions of this experiment. We propose the following mechanism of the nuclear spin pumping: the photoexcited electron virtually occupies the inverted spin state while remaining at the same energy, flops the spin of a single nucleus and tunnels out of the dot into a continuum of states in the contact.

The degree of nuclear spin polarization will depend on the rate of re-excitation of the dot with a spin-polarized electron.\textsuperscript{11} In the low voltage regime this is limited by the radiative lifetime of the electron-hole pair, whereas in our experiment this is fully controlled by the magnitude of the applied voltage. As shown in previous studies on similar structures, the re-excitation of the dot may be blocked by the slow tunneling of the hole due to its heavy effective mass.\textsuperscript{15} Under conditions of resonant excitation this in turn will lead to suppression of the flow of spin-polarized electrons through the dot and a low rate of the electron-nuclear spin flip-flop. The steady state nuclear polarization will then remain low due to nuclear spin depolarization through spin diffusion into the bulk.\textsuperscript{12}

The observed increase with the bias of the nuclear polarization correlates well with the bias dependence of the integrated PC. Figure 3(b) shows that as the applied voltage increases in the range from 2 to 4 V, the integrated PC increases by a factor of 3 for a fixed excitation power, indicating an increasing rate of re-excitation of the dot with spin-polarized e-h pairs. The increased re-excitation rate is a direct consequence of the reduced carrier lifetime on the dot at high bias. For the electron, which tunnels out much quicker than the hole, this can be estimated from the exciton PC linewidth shown in Fig. 3(c). The exciton linewidth in Fig. 3(c) changes from 90 to 240 μeV. Taking into account the bias-independent contribution to the peak width (90 μeV) we deduce that at 4.6 V the exciton lifetime, defined by the fastest tunneling process involving electron,\textsuperscript{18,19,22} is ≈2 ps. The re-excitation rate can be estimated from the magnitude of the PC in Figs. 1 and 2, where a maximum of 150–200 pA is observed. This corresponds to a re-excitation rate of 10⁹ Hz. This is in reasonable agreement with the experiments involving nonresonant optical excitation, where the re-excitation is limited by the radiative recombination time of order 1 ns.\textsuperscript{13,5,6,8–12}

As predicted by our calculations,\textsuperscript{22} at high biases the fast electron tunneling may result in reduced efficiency of the nuclear spin pumping due to a low time-averaged electron occupancy of the dot. In the range of biases in our experiment, the detrimental effect of the fast electron tunneling does not prevail the contribution from the fast hole escape, enabling the re-excitation of the dot.

In conclusion, we have shown that a sizable nuclear spin polarization can be pumped in individual InGaAs dots by resonant optical excitation into the lowest exciton Zeeman states. We demonstrate tuning of the degree of nuclear spin polarization by varying the vertical electric field, controlling the carrier tunneling rates. Overhauser fields in excess of 1.5 T can be optically pumped in the regime of fast electron and hole tunneling from the dot, where the dot re-excitation rates of the order of 10⁹ Hz can be achieved.

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