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Tartakovskii, A.I., Wright, T., Russell, A. et al. (10 more authors) (2007) Nuclear spin switch in semiconductor quantum dots. Physical Review Letters , 98 (2). 026806. ISSN 0031-9007

https://doi.org/10.1103/PhysRevLett.98.026806

Nuclear Spin Switch in Semiconductor Quantum Dots

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(Dated: February 6, 2008)

We show that by illuminating an InGaAs/GaAs self-assembled quantum dot with circularly polarized light, the nuclei of atoms constituting the dot can be driven into a bistable regime, in which either a threshold-like enhancement or reduction of the local nuclear field by up to 3 Tesla can be generated by varying the intensity of light. The excitation power threshold for such a nuclear spin "switch" is found to depend on both external magnetic and electric fields. The switch is shown to arise from the strong feedback of the nuclear spin polarization on the dynamics of spin transfer from electrons to the nuclei of the dot.

The hyperfine interaction in solids [1] arises from the coupling between the magnetic dipole moments of nuclear and electron spins. It produces two dynamical effects: (i) inelastic relaxation of electron spin via the "flip-flop" process (Fig.1a) and (ii) the Overhauser shift of the electron energy [2]. Recently, the hyperfine interaction in semiconductor quantum dots (QDs) has attracted close attention [3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14] fuelled by proposals for QD implementation in quantum information applications [15]. The full quantization of the electron states in QDs is beneficial for removing decoherence mechanisms present in extended systems [16, 17]. However, the electron localization results in a stronger (than in a bulk material) overlap of its wave-function with a large number of nuclei ($N \sim 10^4$ in small self-assembled InGaAs/GaAs dots and up to $10^5 \div 10^6$ in electrostatically-defined GaAs QDs), and the resulting hyperfine interaction with nuclear spins has been found to dominate the decoherence [3, 4, 5, 12, 13, 14] and life-time [9] of the electron spin at low temperatures.

In this Letter, we report the observation of a pronounced bistable behaviour of nuclear spin polarisation, S, in optically pumped self-assembled InGaAs/GaAs dots. In our experiments, spin-polarized electrons are introduced one-by-one into an individual InGaAs dot at a rate w_x (see Fig.1b) by the circularly polarized optical excitation of electron-hole pairs 120 meV above the lowest QD energy states. Due to hole spin-flip during its energy relaxation, both bright and dark excitons can form in the dot ground state. The former will quickly recombine radiatively with a rate $w_{rec} \approx 10^9$ sec^{-1}, whereas the dark exciton can recombine with simultaneous spin transfer to a nucleus via a spin "flip-flop" process (as in Fig.1a) at the rate $w_{rec} N p_{hf}$ [12, 18]. Here N is the number of nuclei interacting with the electron and p_{hf} is the probability of a "flip-flop" process, which from our perturbation theory treatment is given by:

$$p_{hf} = |h_{hf}|^2/(E_{eZ}^2 + \tfrac{1}{4}\gamma^2). \tag{1}$$

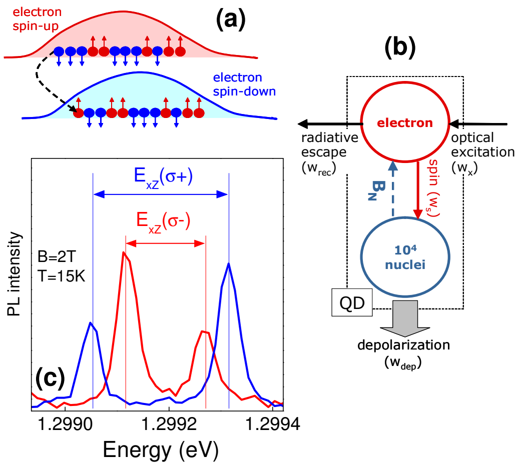

FIG. 1: (a) Schematic representation of the electron-nuclear spin "flip-flop" process. (b) The steps involved in nuclear polarization of a quantum dot (see text for detail). (c) X^0 photoluminescence spectra recorded for an individual InGaAs QD in an external magnetic field $B = 2$T at $T = 15$K. The spectrum excited with σ^+ (σ^-) light resonant with the wetting layer is plotted in blue (red). The horizontal arrows show the corresponding exciton Zeeman splittings.

Here γ is the exciton life-time broadening, h_{hf} is the strength of the hyperfine interaction of the electron with a single nucleus and E_{eZ} is the electron Zeeman splitting. E_{eZ} is strongly dependent on the effective nuclear magnetic field B_N generated by the nuclei. This provides a feedback mechanism between the spin transfer rate and the degree of nuclear polarization ($B_N \propto S$) in the dot [19]. The feedback gives rise to bistability in the nuclear polarization and threshold-like transitions between the spin states of 10^4 nuclei leading to abrupt changes of B_N by up to 3T in few nanometre sized QDs.

We observe such threshold-like transitions (referred to below as a nuclear spin 'switch') in several different structures containing self-assembled InGaAs/GaAs QD with

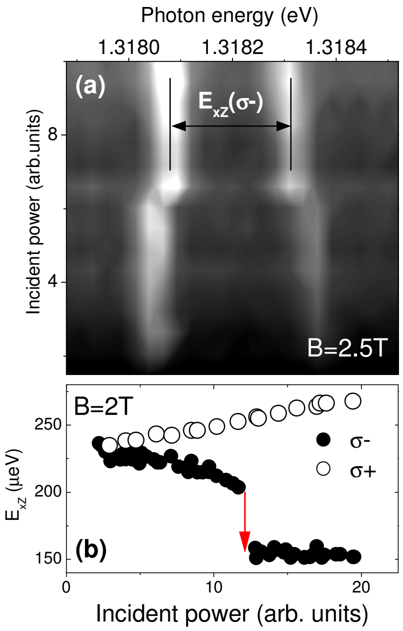

FIG. 2: (a) Grey-scale plot showing exciton PL spectra recorded for an individual InGaAs dot. The spectra are recorded at B=2.5T using unpolarized detection and σ^- excitation into the wetting layer. The spectra are displaced along the vertical axis according to the excitation power at which they are measured. (b) E_{xZ} power dependences measured at $B = 2$T for σ^+ and σ^- excitation polarizations.

~3x20x20 nm size. Below, we present results obtained at a temperature of 15K for two GaAs/AlGaAs Schottky diodes, where the dots are grown in the intrinsic region of the device. In these structures a bias can be applied permitting control of the vertical electric field, F [20]. For photoluminescence (PL) experiments, individual dots are isolated using 800 nm apertures in a gold shadow mask on the sample surface.

Fig.1c shows time-averaged (60s) PL spectra recorded for a neutral exciton in a single QD in an external magnetic field of 2T. Circularly polarized laser excitation in the low energy tail of the wetting layer (at 1.425eV) is employed and unpolarized PL from the dot is detected using a double spectrometer and a CCD. For each excitation polarization a spectrum consisting of an exciton Zeeman doublet is measured with the high (low) energy component dominating when σ^+ (σ^-) polarization is used. A strong dependence of the exciton Zeeman splitting (E_{xZ}) on the polarization of the excitation is observed in Fig.1c: $E_{xZ}(\sigma^+) = 260\mu$eV and $E_{xZ}(\sigma^-) = 150\mu$eV. Such a dependence is a signature of dynamic nuclear polarization [6, 7, 8, 10, 11], which gives rise to the Overhauser field B_N aligned parallel or anti-parallel to B for σ^+ or σ^- excitation, respectively.

The dependence of exciton PL at $B = 2.5$T on the power, $P \propto w_x$, of σ^- excitation is shown in the grey-scale plot in Fig.2a. At low P the Zeeman splitting $E_{xZ} = 310\mu$eV. As the power is increased, a threshold-like decrease of E_{xZ} to 225μeV is observed at $P = P_{up}$ indicating the sudden appearance of a large nuclear field. Fig.2b shows the power dependence of E_{xZ} measured at $B = 2$T for both circular polarizations of incident light. For σ^- excitation, E_{xZ} decreases below the threshold followed by a weak power dependence above the threshold. The σ^- behaviour contrasts the weak monotonic increase of E_{xZ} seen for σ^+ excitation over the whole range of powers similar to that reported in Ref.[21].

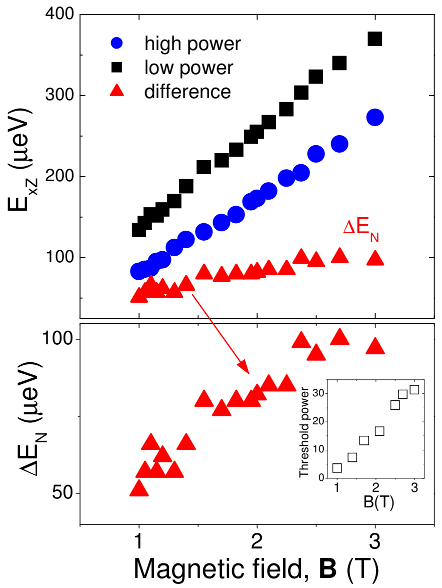

FIG. 3: Dependence of the QD exciton Zeeman splitting $E_{xZ}(\sigma^-)$ on the external magnetic field. Squares and circles show high and low power data, respectively, and triangles show their difference, ΔE_N. For all B shown in the figure the nuclear switch threshold was observed with the threshold power shown in the inset as a function of B.

The variation of the Zeeman splitting in Fig.2 reflects the change in the nuclear field B_N: $E_{xZ}(\sigma^\pm) = |g_e + g_h|\mu_B B \pm |g_e|\mu_B B_N(\sigma^\pm)$ (where g_e is the electron g-factor [22], μ_B is the Bohr magneton). B_N in its turn depends on the external field B. Triangles in Fig.3 show the difference between $E_{xZ}(\sigma^-)$ at low and high powers (squares and circles in Fig.3a, respectively), $\Delta E_N = |g_e|\mu_B B_N(\sigma^-)$, as a function of B. ΔE_N increases linearly with B at low fields and then saturates at $B \approx 2.5 \div 3$T. The inset in Fig.3 shows that the threshold-power for the switch also increases nearly linearly with B. No switch could be observed at $B > 3$T in the range of powers employed in our studies.

For $B < 3$T, when the excitation power was gradually reduced from powers above the switch, E_{xZ} was found to vary weakly with power until another threshold was reached, where the magnitude of the exciton Zeeman splitting abruptly increased (at $P = P_{down}$), as shown in Fig.4. This increase of E_{xZ} corresponds to depolarization of the nuclei and hence reduction of B_N. The observed hysteresis of nuclear polarization shows that two significantly different and stable nuclear spin configurations can exist for the same external parameters of magnetic field and excitation power. We find that high nuclear polarization persists at low excitation powers for more than 15 min, this time most likely being determined by the stability of the experimental set-up.

We also show in Figs.4a,b that the size of the hysteresis loop depends on the external magnetic or electric fields (the electric field is given by $F = (V_{rev} + 0.7V)/d$, where V_{rev} is the applied reverse bias and $d = 230nm$ is the width of the undoped region of the device). The inset in Fig.4b shows the P_{up} reverse bias-dependence. In general, both P_{up} and P_{down} increase with B and reverse bias, but also the difference between the two thresholds increases, leading to a broader range of incident powers in which the bistability occurs. The threshold bias dependence arises from the influence of the electric field on the charge state of the dot [20], and will be discussed elsewhere.

In order to explain the nuclear switching and bistability, we employ a model based on spin-flip assisted e-h recombination [12, 18]. We assume that the electron spin is defined by the sign of the circularly polarized excitation ($\sigma^{\pm}$), whereas the hole spin is partially randomized during the energy relaxation. Thus, dark and bright excitons can be formed in the dot ground state, with the rates αw_x and $(1-\alpha)w_x$, respectively. A bright exciton recombines with the rate w_{rec} without spin transfer to the nuclei. In contrast, a dark exciton can recombine with the electron simultaneously flipping its spin due to the hyperfine interaction: the electron virtually occupies an optically active state with the opposite spin and the same energy [12, 18] transferring spin to nuclei and, then, recombines with the hole with the rate $w_{rec}Np_{hf}$, where p_{hf} (given by Eq.(1) depends on the electron Zeeman splitting, $E_{eZ} = |g_e|\mu_B[B \pm B_N(\sigma^{\pm})]$. For the case of σ^- excitation, polarization of the nuclei leads to a decrease of E_{eZ}, and thus a positive feedback and speeding up of the spin transfer process: the more spin is transferred to the nuclear system the faster becomes the spin transfer rate. By contrast for σ^+ excitation, spin transfer leads to an increase of E_{eZ}, leading to the saturation of S (and B_N) at high power.

The spin transfer to nuclei at a rate $w_s \propto \alpha w_x N p_{hf}$ competes with nuclear depolarization, $\dot{S} = -w_{dep}S$ (see Fig.1b) due to spin diffusion away from the dot into the surrounding GaAs [23, 24], at a rate $w_{dep} \sim 1\div 10\text{s}^{-1}$. At high power of σ^- excitation w_s may exceed w_{dep} and then a stimulated nuclear polarization will take place due to the positive feedback mechanism described above leading to an abrupt increase of the nuclear spin (at $P = P_{up}$). To achieve the condition $w_s = w_{dep}$ a higher w_x (power) will be required at higher B in agreement with observation in the inset of Fig.3. The stimulation at $P \approx P_{up}$ stops when either (i) $|E_{eZ}|$ starts increasing again since $B_N > B$, causing reduction of w_s or (ii) the maximum achievable $B_N = B_N^{max}$ in the given dot is reached. This explains the dependence in Fig.3, where ΔE_N, and hence the nuclear field, increases at low B and saturates at high fields, from which we estimate B_N^{max} ≈2.5-3T [25].

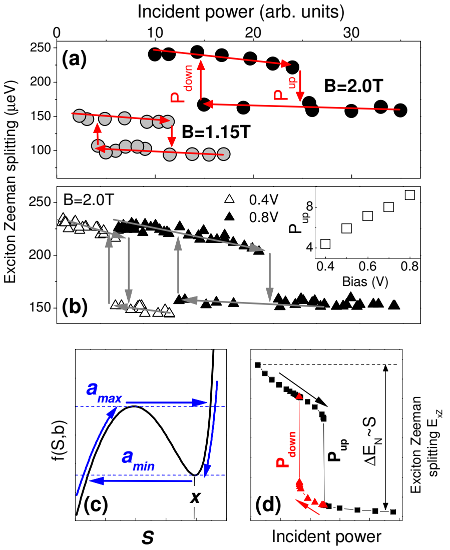

FIG. 4: (a) Power dependence of $E_{xZ}(\sigma^-)$ measured at $B = 2$T and 1.15T. The arrows show the direction in which the hysteresis loop is measured with two thresholds P_{up} and P_{down}. (b) $E_{xZ}(\sigma^-)$ power dependence measured at $B = 2.0$T. The two hysteresis loops are measured at 0.4 and 0.8V applied bias. The inset shows the P_{up} dependence on the reverse bias applied to the diode. (c) The full line shows the function $f(S, b)$ from Eq.2. Arrows show how the hysteresis loop is formed when the parameter a ($\propto w_x$) is varied for a fixed b. (d) Hysteresis loop of the exciton Zeeman splitting as a function of incident power calculated using Eq.2 for $x = 0.7$ and $\theta = 0.1$.

When the power is reduced from beyond the threshold P_{up}, and the condition $w_s < w_{dep}$ is reached at sufficiently low w_x, a strong negative feedback is expected: further nuclear depolarization will lead to even lower w_s due to the increase in the electron Zeeman energy E_{eZ}. Thus, an abrupt nuclear depolarization will take place (at the threshold P_{down}). This explains the observed hysteresis behavior in Fig.4, and also accounts for the existence of a bistable state in the nuclear polarization at intermediate powers, $P_{down} < P < P_{up}$.

To model this bistability, we solve the rate equations for the nuclear spin polarization S, and populations of bright and dark excitons, n_b and n_d, ($1 - n_b - n_d$ is the probability that the dot is empty):

$$\begin{aligned} \dot{S} &= n_d w_{rec} p_{hf}(1-S) - w_{dep}S, \\ \dot{n}_b &= (1-\alpha)\, w_x[1 - n_b - n_d] - w_{rec}n_b, \end{aligned}$$

$$\dot{n}_d \;=\; \alpha w_x[1-n_b-n_d] - \frac{1}{2}(1-S)N w_{rec} p_{hf} n_d.$$

In the limit $\gamma \ll |g_e|\mu_B B_N^{max}$ we obtain the following equation for a steady state polarisation induced by the $\sigma^{\pm}$ excitation:

$$f(S,b) \equiv S\left[1 + b\frac{(x \pm S)^2}{1-S}\right] = a, \;\; x = \frac{B}{B_N^{max}}, \qquad (2)$$

where for $w_x \ll w_{rec}$ (low occupancy of the dot)

$$a = 2\alpha w_x/N w_{dep} \quad , \quad b = 2\alpha N w_x/w_{rec}. \qquad (3)$$

In Eq.(3), both a and b are proportional to the excitation power. For low excitation powers such that $b \ll 1$, for both σ^+ and σ^- excitation, Eq.(2) has a single solution for the degree of nuclear polarization, namely $S \approx a$. In the σ^+ excitation case, $f(S,b)$ is a monotonic function and for all a and b a single solution to Eq.(3) is obtained. On the other hand, for σ^- excitation, for higher powers such that $b \gtrsim 1$, $f(S,b)$ acquires an N-shape, as illustrated in Fig.4c. As shown in the diagram, an abrupt transition to $S > x$ ($S \approx a$) will be obtained when a_{max} (a_{min}) is reached at the local maximum (minimum) of $f(S,b)$. The transitions at a_{max} and a_{min} correspond to the P_{up} and P_{down} thresholds in Fig.4, respectively, whereas for $a_{min} < a < a_{max}$, the polarization degree S enters a regime of bistability in which the cubic Eq.(2) has three solutions, two of which are stable with an unstable one in between.

We find that the occurrence of the switch to $S > x$ depends on the dimensionless ratio $\theta = a/b = w_{rec}/N^2 w_{dep}$, since at small θ, a will grow more slowly with w_x than the magnitude of $f(S,b)$ at the local maximum. θ is determined by the dot parameters only, and can be estimated for the dots studied in our experiment: we obtain $\theta_{exp} \sim 1 \div 10$ from $w_{rec} \sim 10^9$ sec^{-1}, $w_{dep} \sim 1 \div 10$ sec^{-1} and $N \sim 10^4$. Using Eq.(2) we find that for $x \leq 0.8$ the spin switch is possible for any $\theta_{exp} > \theta_c$, where $\theta_c = \frac{1}{16}(3 - \sqrt{9-8x})(4x - 3 + \sqrt{9-8x})^2/(1+\sqrt{9-8x}) \leq 0.1$, consistent with our observations. A hysteresis loop calculated using Eq.(2) for $x = 0.7$ (with $\theta_c \approx 0.07$) and $\theta = 0.1$ (close to critical θ_c), is shown in Fig.4d.

To summarize, we have observed a strong optically induced bistability of the nuclear spin polarization in self-assembled InGaAs QDs. We show that nuclear magnetic fields up to 3T can be switched on and off in individual dots by varying one of three external controlling parameters: electric and magnetic fields and intensity of circularly polarized excitation. We have found that the nuclear spin switch effect is a general phenomenon and has been observed in several different InGaAs/GaAs quantum dot samples at temperatures $T = 15 - 30$K and in the range of external magnetic fields $B = 1 \div 3$T. The effect arises due to the strong feedback of the nuclear spin polarization on the dynamics of the electron-nuclear spin interaction accompanying the radiative recombination process, which is enhanced when the Overhauser and external magnetic fields cancel each other.

We thank R. Oulton, A. Imamoglu, C. Marcus, D. Loss, and S. Tarucha for discussions. This work has been supported by the Sheffield EPSRC Programme grant GR/S76076, the Lancaster-EPSRC Portfolio Partnership EP/C511743, the EPSRC IRC for Quantum Information Processing, ESF-EPSRC network EP/D062918, and by the Royal Society. AIT acknowledges support from the EPSRC Advanced Research Fellowship EP/C54563X/1 and research grant EP/C545648/1.

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