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Nuclear Spin Switch in Semiconductor Quantum Dots

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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 thresholdlike enhancement or reduction of the local nuclear field by up to 3 T can be generated by varying the pumping intensity. The excitation power threshold for such a nuclear spin "switch" is found to depend on both the external magnetic and electric fields. The switch is shown to arise from the strong feedback of the nuclear spin polarization on the dynamics of the 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. In quantum dots (QDs), the electron coupling to the underlying nuclei is stronger than in a bulk material due to the electron confinement [2–9]. This leads to the enhanced contribution of the hyperfine interaction to the electron spin decoherence [3] and relaxation [4,8,9] at low temperatures. In this Letter, we show that in optically pumped individual self-assembled InGaAs/GaAs dots the enhanced coupling between the electron and nuclear spins gives rise to a pronounced bistability of the nuclear spin polarization.

In our photoluminescence (PL) experiments, spin-polarized electrons are introduced one by one into an individual InGaAs dot at a rate w_x by the circularly polarized optical excitation of electron-hole pairs 120 meV above the lowest QD energy states [see Fig. 1(a)]. Both bright and dark excitons can form in the dot ground state due to hole spin-flip during its energy relaxation. The former will quickly recombine radiatively with a rate $w_{rec} \sim 10^9$ sec^{-1}, whereas the dark exciton can recombine with simultaneous spin transfer to one of N nuclei in a dot ($N \sim 10^4$) via a spin "flip-flop" process [as in Fig. 1(b)]. The rate of this process is $w_{rec}Np_{hf}$ [8,10], where p_{hf} is the probability of a spin "flip-flop" process and is given by:

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

Here γ is the electron lifetime 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 dependent on both external and effective nuclear (Overhauser) magnetic fields, where the latter, B_N, is proportional to the degree of nuclear polarization, S. This provides a feedback mechanism between the spin transfer rate, w_s, and nuclear polarization in the dot leading to the enhancement of w_s in the case when B_N acts to reduce the electron Zeeman splitting. The feedback gives rise to bi-

stability in the nuclear polarization and thresholdlike transitions between the spin states of 10^4 nuclei (referred to below as a nuclear spin "switch") leading to abrupt changes of B_N by up to 3 T.

We observe the nuclear spin switch in several different structures containing self-assembled InGaAs QDs with $\sim 3 \times 20 \times 20$ nm size. Below, we present results obtained at a temperature of 15 K for two GaAs/AlGaAs based Schottky diodes [11], where the dots are grown in the intrinsic region of the device. Figure 1(c) shows time-averaged (60 s) PL spectra recorded for a neutral exciton
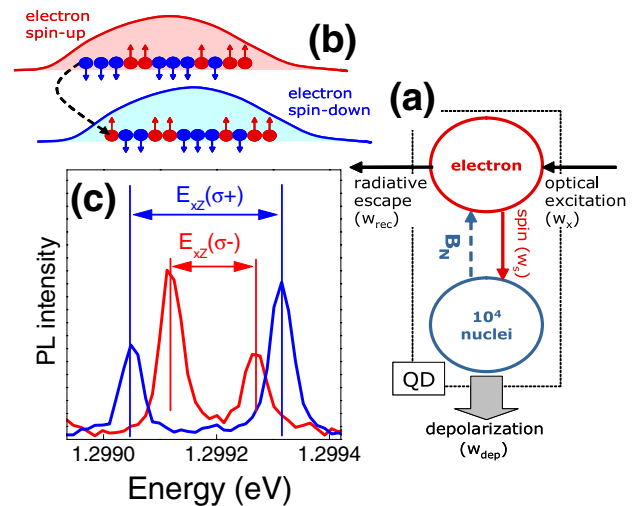

FIG. 1 (color online). (a) The steps involved in nuclear polarization of a quantum dot (see text for detail). (b) Schematic diagram of the electron-nuclear spin "flip-flop," in which the electron confined on the dot flips its spin simultaneously flipping the spin of a single nucleus. (c) X^0 PL spectra 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.

in a single QD in an external magnetic field of 2 T. Circularly polarized laser excitation at 1.425 eV 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. 1(c): $E_{xZ}(\sigma^+) = 260 \ \mu eV$ and $E_{xZ}(\sigma^-) = 150 \ \mu eV$. Such a dependence is a signature of dynamic nuclear polarization [2,6,7], which gives rise to the Overhauser field B_N aligned parallel (antiparallel) to B for σ^+ (σ^-) excitation.

The dependence of exciton PL at $B = 2.5$ T on the power, $P \propto w_x$, of σ^- excitation is shown in the gray-scale plot in Fig. 2(a) [12]. At low P the Zeeman splitting $E_{xZ} = 310 \ \mu eV$. As the power is increased, a thresholdlike decrease of E_{xZ} to 225 μeV is observed at $P = P_{up}$ indicating the sudden appearance of a large nuclear field. Figure 2(b) 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 [13]. The σ^- behavior contrasts to the weak monotonic

increase of E_{xZ} seen for σ^+ excitation over the whole range of powers.

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 (g_h) is the electron (hole) g factor [14], μ_B is the Bohr magneton]. B_N in its turn depends on the external field B. The triangles in Fig. 3 show the difference between $E_{xZ}(\sigma^-)$ at low and high powers (squares and circles in Fig. 3, 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 $B \approx$ 2.5–3 T. For a typical $In_{0.3}Ga_{0.2}As$ dot, the maximum ΔE_N in Fig. 3 corresponds to a nuclear polarization of 40% [7]. 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 such as 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 setup.
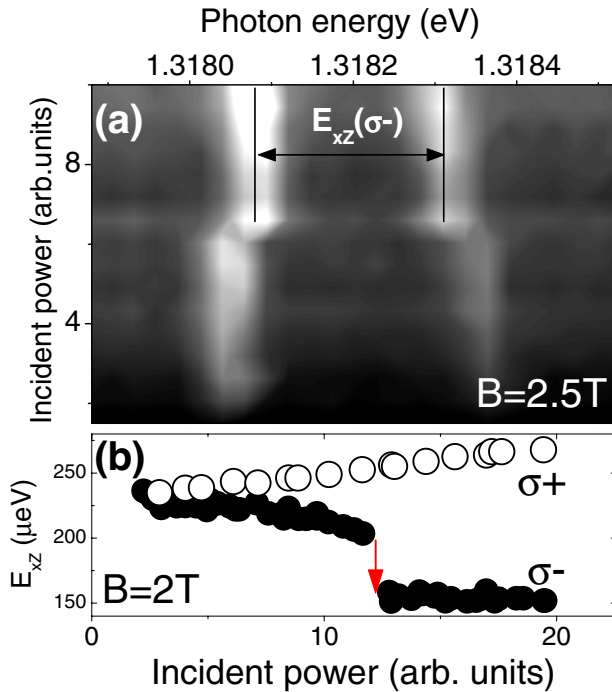

FIG. 2 (color online). (a) Gray-scale plot showing exciton PL spectra recorded for an individual InGaAs dot. The spectra are recorded at $B = 2.5$ T using unpolarized detection and σ^- excitation into the wetting layer. The spectra are displaced along the vertical axis according to the excitation power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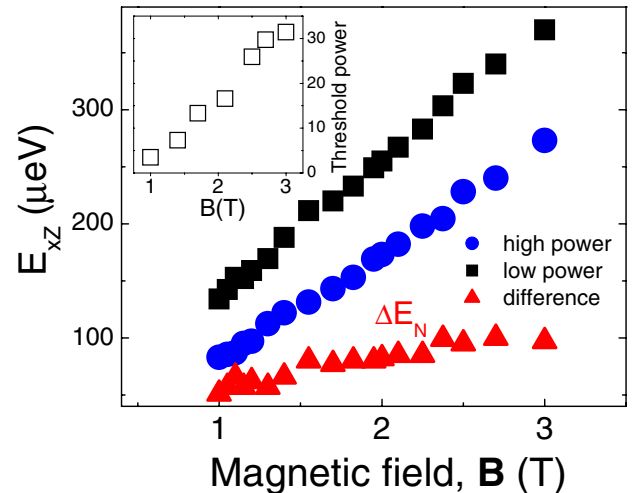

FIG. 3 (color online). Dependence of the QD exciton Zeeman splitting $E_{xZ}(\sigma^-)$ on the external magnetic field. Squares and circles show high and low power data, respectively, and triangles show their difference, ΔE_N. For all B shown in the figure the nuclear switch threshold was observed with the threshold power shown in the inset as a function of B.
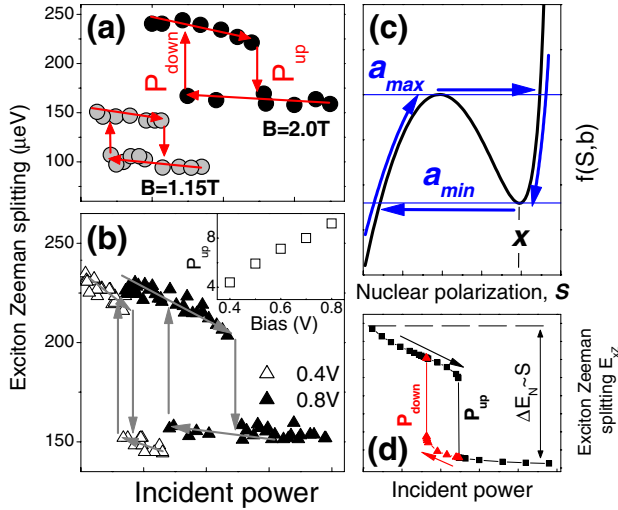
FIG. 4 (color online). (a) Power dependence of $E_{xZ}(\sigma^-)$ measured at $B = 2$ T and 1.15 T. The arrows show the direction in which the hysteresis loop is measured with two thresholds P_{up} and P_{down}. (b) $E_{xZ}(\sigma^-)$ power dependence measured at $B = 2.0$ T. The two hysteresis loops are measured at 0.4 and 0.8 V 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 $\theta = 0.1$.

We also show in Figs. 4(a) and 4(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 $d = 230$ nm is the width of the undoped region of the diode]. The inset in Fig. 4(b) shows the P_{up} reverse bias dependence for a p-type Schottky diode. 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 carrier population and hence on the electron spin dynamics in the dot [15], the latter also influenced by the interaction with the continuum of states in the contact [16]. These effects will be discussed in detail elsewhere.

In order to explain the nuclear spin switching and bistability, we employ a model based on spin-flip assisted e-h recombination [8,10]. 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 simulta-

neously flipping its spin due to the hyperfine interaction: the electron virtually occupies an optically active state with the opposite spin and the same energy [8,10] transferring spin to nuclei and, then recombines with the hole with the rate $w_{rec}Np_{hf}$, where p_{hf} 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 pumped into 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 the nuclei at a rate $w_s \propto \alpha w_x Np_{hf}$ competes with nuclear depolarization, $\dot{S} = -w_{dep}S$ [see Fig. 1(b)] due to spin diffusion away from the dot into the surrounding GaAs, at a rate $w_{dep} \sim 1$–10 s^{-1} [17,18]. 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 the 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 $B_N^{max} \approx 2.5$–3 T [19].

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 a threshold power $P_{down} < P_{up}$. This explains the hysteresis behavior in Fig. 4 which accounts for the existence of a bistable nuclear polarization state for $P_{down} < P < P_{up}$.

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,

$$\dot{S} = n_d w_{rec} p_{hf}(1 - S) - w_{dep}S,$$
$$\dot{n}_b = (1 - \alpha)w_x[1 - n_b - n_d] - w_{rec}n_b;$$
$$\dot{n}_d = \alpha w_x[1 - n_b - n_d] - \tfrac{1}{2}(1 - S)Nw_{rec}p_{hf}n_d,$$

where $1 - n_b - n_d$ is the probability that the dot is empty, and p_{hf} is given by Eq. (1). In the limit $\gamma \ll |g_e|\mu_B B_N^{max}$ we obtain the following equation for a steady state polarization, S, induced by the $\sigma^\pm$ excitation:

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

where $x = B/B_N^{max}$, and for $w_x \ll w_{rec}$ (low occupancy of the dot) both a and b are proportional to the excitation power: $a = 2\alpha w_x/N w_{dep}$ and $b = 2\alpha N w_x/w_{rec}$.

For a low excitation power, such that $b \ll 1$, Eq. (2) has a single solution $S \approx a$ for both σ^+ and σ^- excitation. In the σ^+ excitation case, $f(S, b)$ is a monotonic function and for all a and b a single solution to Eq. (2) 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. 4(c). 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 Eq. (2) has three solutions, two of which are stable with an unstable one in between.

We find [using Eq. (2)] 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 $\theta_{exp} \sim 1$–10 from $w_{rec} \sim 10^9$ sec^{-1}, $w_{dep} \sim$ 1–10 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 $\theta_c = \frac{1}{16}(3 - \sqrt{9 - 8x})(4x - 3 + \sqrt{9 - 8x})^2/(1 + \sqrt{9 - 8x}) \leq 0.1$, which is 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 the critical θ_c), is shown in Fig. 4(d).

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 3 T 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. The nuclear spin switch has been observed for InGaAs/GaAs QDs at temperatures $T =$ 15–30 K and external magnetic fields $B = 1$–3 T. The effect arises due to the strong feedback of the nuclear spin polarization on the dynamics of the electron-nuclear spin transfer accompanying the radiative recombination process, which is accelerated when the Overhauser and external magnetic fields cancel each other.

Note added.—Another report on similar nuclear effects has been published recently [20].

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