Stabilization of the Electron-Nuclear Spin Orientation in Quantum Dots by the Nuclear Quadrupole Interaction

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The nuclear quadrupole interaction eliminates the restrictions imposed by hyperfine interaction on the spin coherence of an electron and nuclei in a quantum dot. The strain-induced nuclear quadrupole interaction suppresses the nuclear spin flip and makes possible the zero-field dynamic nuclear polarization in self-organized InP/InGaP quantum dots. The direction of the effective nuclear magnetic field is fixed in space, thus quenching the magnetic depolarization of the electron spin in the quantum dot. The quadrupole interaction suppresses the zero-field electron spin decoherence also for the case of nonpolarized nuclei. These results provide a new vision of the role of the nuclear quadrupole interaction in nanostructures: it elongates the spin memory of the electron-nuclear system.

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An electron in a semiconductor quantum dot (QD) interacts with a large number $N \sim 10^5$ of the surrounding lattice nuclei. If one considers the electron spin as a solid-state quantum bit the spin decoherence should be overwhelmed. Hyperfine interaction (with hyperfine constant $A \approx 100$ μeV) imposes strict limits on the electron spin coherence time [1] at zero magnetic field: the initial dephasing in a random quasistatic nuclear field within a few nanoseconds [2] is followed by the decoherence over the microsecond time scale, because the nuclear field changes in time due to the precession of nuclear spins in the effective electron field (Knight field) of hyperfine interaction. An external magnetic field effectively decouples the electron and the nuclei, suppressing transitions with mutual electron and nuclear spin flips. Such a ''brute force'' technique extends the longitudinal electron spin relaxation time up to milliseconds [3]. A more refined approach involves the optical cooling of nuclei down to ultralow temperature $\theta \sim 10^{-6}$ K [4]: a relatively weak time-averaged Knight field induces a substantial nuclear polarization. Polarized nuclei create an effective magnetic field due to the hyperfine interaction that suppresses the electron spin relaxation similarly to the external field. The dynamic nuclear polarization in a low external field (1–10 G) was observed in charged InP/InGaP [5], InGaAs/GaAs [6], InAs/GaAs [7], and CdSe/ZnSe [8] QDs.

Here we propose another approach and show that the nuclear quadrupole interaction eliminates the restrictions imposed by hyperfine interaction on the spin coherence of the electron and nuclei in a quantum dot. The strain-induced quadrupole interaction suppresses the spin flip of nuclei and makes possible the zero-field dynamic nuclear polarization in quantum dots. The direction of the effective magnetic field of polarized nuclei is fixed in space, thus quenching the magnetic depolarization of the electron spin in the quantum dot. The quadrupole interaction suppresses the zero-field electron spin decoherence also for the case of nonpolarized nuclei. Therefore, the nuclear quadrupole interaction elongates the spin lifetime of the electron-nuclear system in quantum dots.

The structures were grown by MOVPE on a [001]-GaAs substrate and contained nanosized InP islands (120 nm in diameter with density $3 \times 10^9\,\mathrm{cm}^{-2}$) restricted by InGaP barrier. The layers contained donor impurities $\approx 10^{15}\ \mathrm{cm}^{-3}$. The samples were placed in a liquid-helium cryostat ($T = 4.2$ K) and pumped by a 10 W/cm^2 He-Ne laser (1.96 eV) above wetting layer. The photoluminescence (PL) circular polarization ρ_c was measured in the reflection geometry within the maximum of PL intensity. The external magnetic field $\vec{B}$ was applied in Voigt or in Faraday geometries. Measurements both in the presence and in the absence of dynamic nuclear polarization were carried out. To avoid the dynamic polarization of nuclei the helicity of light was alternated in sign at a frequency of 26.61 kHz with a photoelastic quartz modulator in the excitation channel and a fixed quarter-wave plate as analyzer. In this case there is not enough time for the nuclear spin to follow the polarization of electrons [9]. To initiate the Overhauser effect the polarizer and analyzer were interchanged.

Excitation of QD nanostructures by circularly polarized light provides the optical pumping of resident QD electrons [10–12]. A plausible scenario under nonresonant excitation conditions is the capture of single carriers into the dots. For example, the hole is captured first, forming a neutral exciton (an optically allowed, bright exciton with angular momentum projection ± 1 or an optically forbidden, dark exciton with projection ± 2 onto [001]). The trapping of the photoexcited electron follows this. If the exciton recombines first, then the optically oriented electron becomes a resident. Thus the optical pumping takes place by replacement of nonpolarized electrons by the oriented ones, similar to the usual optical orientation in bulk n-type semiconductors [4]. The optical orientation of both neutral excitons and resident electrons can be detected by the degree ρ_c of the PL circular polarization of nega-

tively charged QD ensemble. The polarized PL of singly charged islands originates from the ground state of X^- trions—the complexes of two electrons with antiparallel spins and one hole. In turn, polarization of trions is determined by polarization of resident electrons and neutral excitons (both bright and dark) at the moment of trion formation [10]. Interplay between the polarizations of quasiparticles forming the trion makes usually the helicity of X^- emission opposite to the pump helicity (so-called negative circular polarization) [10–12].

The Hanle effect measurements (Voigt geometry) separate the contributions of neutral excitons and resident electrons to the circular polarization of the PL. In a low magnetic field ($B \sim 100$ G) only resident electron spins undergo Larmor precession [13], thus depolarizing PL. In contrast with the Hanle effect, the measurements in Faraday geometry reveal the fine structure of the neutral excitons forming trions. The electron-hole anisotropic exchange interaction [14] mixes $+1$ and -1 bright states (it comes from the QD anisotropy and induces splitting by $\delta_b = 10$–100 μeV), as well as dark states $+2$ and -2 (split by $\delta_d = 1$–2 μeV). As a result the depolarization of neutral excitons takes place. An external magnetic field in Faraday geometry eliminates the mixing, thus restoring their optical orientation.

Figure 1 shows the dependence $\rho_c(B)$ in Voigt [$\vec{B} \perp$ [001], Fig. 1(a)] and Faraday [$\vec{B}\|$[001], Fig. 1(b)] geometries. Polarization is negative, which is a striking signature of X^- trions. The Hanle effect of resident electrons [filled circles on Fig. 1(a)] is measured in the absence of the dynamical nuclear polarization (excitation by light of alternating helicity). The Hanle curve can be fitted by Lorentzian with the half-width $B_{1/2} \approx 50$ G. Taking into account the electron g factor $g = 1.6$ [15] one can estimate the electron spin lifetime according to formula $T_s = \hbar/\mu_B g B_{1/2} \approx 1.4$ ns [4,16]. One can see that a complete PL depolarization takes place. This means that the neutral excitons forming the trion are nonpolarized due to anisotropic electron-hole exchange interaction. An external magnetic field in Faraday geometry restores the optical orientation of excitons. Because of the inequality $\delta_d \ll \delta_b$ one restores the orientation of dark excitons in low fields, enhancing the circular polarization of PL [filled circles on Fig. 1(b)] followed by the restoration of orientation of bright states in larger (~10 kG) magnetic field (this range is not shown; see Ref. [10] for details). The dependences $\rho_c(B)$ are symmetrical with respect to the change of the B sign.

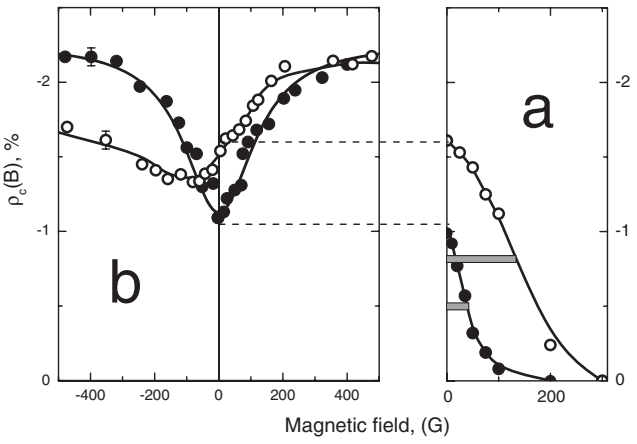

FIG. 1. The dependence of the circular polarization degree ρ_c on the external magnetic field B in Voigt (a) and Faraday (b) geometries. Closed circles are measured in the absence of nuclear polarization (excitation by light of alternating helicity), open circles in the presence of nuclear polarization (helicity is constant).

The presence of dynamical nuclear polarization (open circles in Figs. 1(a) and 1(b) are measured under excitation by light of constant helicity) changes the situation substantially. The curve of restoration of the trion optical orientation is shifted [Fig. 1(b)] by the nuclear magnetic field value about 80 G (the curve is broadened due to the nonuniform nuclear field distribution). Moreover, the zero-field polarization $\rho_c(B = 0)$ value is also affected [5]. It means that the dynamic nuclear polarization persists at zero field, too. For InAs QDs this effect was interpreted [7] as the Knight field-enabled nuclear polarization. In our case (at least), it is the result of electric quadrupole interaction suppressing the depolarization of In nuclei by local magnetic fields $B_L \sim 1$ G of the surrounding nuclei. The Hanle effect measurements [open circles on Fig. 1(a)] support this conclusion. The fact is that the Hanle curve (it is symmetrical with respect to the ordinate axis) half-width is 150 G. This is 3 times larger than the value $B_{1/2} = 50$ G in the absence [filled circles in Fig. 1(a)] of nuclear polarization [17]. This result contradicts the standard theory of optical orientation of electrons and nuclei in the nonstrained GaAs-type semiconductor (i.e., without quadrupole effects). Indeed, the nuclear field enhances the depolarizing effect of the external field on the spin of electrons [9] leading to the *narrowing* of the Hanle curve in striking contrast with the result in Fig. 1(a) [see also Fig. 2(a) for normalized data]. Thus the Hanle curve broadening cannot be explained by the Knight-field-enabled nuclear polarization that should enhance the low-field depolarization of PL.

It is important to note that the values of the nuclear-field-induced shift of the $\rho_c(B)$ in Faraday geometry [Fig. 1(b)] and the Hanle curve half-width [Fig. 1(a)] in the presence of nuclear polarization are comparable. Therefore, we have to suggest the existence of the nuclear field whose direction is fixed in space near the [001] axis and is nonzero even for $B = 0$. It both shifts the $\rho_c(B)$ dependence in Faraday geometry and blocks the Larmor precession of electron spin in Voigt geometry [Fig. 2(b)]. The fixing of the nuclear field direction in InP islands can be related with the quadrupole interaction of indium nuclei only [18] (95.5% of ^{115}In and 4.5% of ^{113}In, both with $I = 9/2$ [19]). It is essential in the low-field region when Zeeman splitting

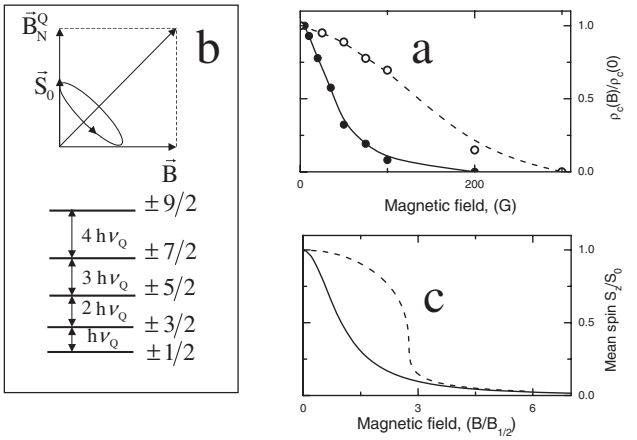

FIG. 2. (a) Normalized data from Fig. 1(a). (b) Illustration of mutual orientation of zero-field mean spin $\vec{S}_0$, nuclear field $\vec{B}_N^Q$, external magnetic field $\vec{B}$, and the energy level scheme of In in the presence of quadrupole interaction. (c) The result of calculation with the use of Eq. (1) under $a = 0$ (solid curve); $aS_0 = 5B_{1/2}$ (dashed curve).

of the nucleus $\gamma\hbar B$ (γ—nuclear gyromagnetic ratio) is less than the quadrupole splitting $h\nu_Q$.

The dynamic nuclear polarization in the presence of a strong quadrupole interaction (QI) was studied intensively in AlGaAs bulk solid solutions [9]. Substitution of Ga atoms by Al atoms leads to the QI of As nuclei. The main axes of the QI were directed along {111}. In our case, the binary compound InP forms islands and QI is absent in zero approximation [20]. However, a very large strain (due to 3.7% lattice mismatch between InP and InGaP) takes place in the system under study [21] with the main axis being close to the growth direction. Strain will induce the QI of indium nuclei and will dominate in a low magnetic field. In the simplest case of uniaxial strain along the z axis ($e_{zz} \neq 0$) the QI Hamiltonian $h\nu_Q[\hat{I}_z^2 - I(I+1)/3]/2$ is determined by the only constant [22]

$$\nu_Q = \frac{3eV_{zz}Q}{2I(2I-1)h},$$

where $Q = 0.76 \times 10^{-24}$ cm^2 is the quadrupole moment of ^{115}In [19]. Strain induces the electric field gradient (EFG) $V_{zz} = S_{11}e_{zz}$, where the constant $S_{11} = 2 \times 10^{16}$ statcoulombs/cm^3 was measured experimentally for the case of ^{115}In nuclei in InP [23]. Estimating the z component of deformation tensor as $e_{zz} = 0.02$ (2%) we get ν_Q about 1 MHz. The constant ν_Q exceeds the precession frequency of In nuclei $\gamma B/2\pi = 933$ Hz/G [19] up to the field values $B \approx 1000$ G. Therefore, QI dominates over the nuclear Zeeman interaction for the entire field range in Figs. 1 and 2. In zero field an axially symmetric QI lifts the degeneracy of In nuclei, grouping the energy levels in pairs $m = \pm 1/2, \pm 3/2, \ldots, \pm 9/2$ with the same module of momentum projection m onto z axis [Fig. 2(b)]. In a weak magnetic field ($\gamma B \ll 2\pi\nu_Q$) the Zeeman energy is a small perturbation. Then nuclei in states $\pm m$ can be considered as quasiparticles with pseudospin $\frac{1}{2}$ [9]. Zeeman interaction of these quasiparticles is characterized by a strongly anisotropic g factor with the main axes of the g tensor coinciding with those of EFG. In the first order the transverse magnetic field does not split the states $+m$ and $-m$ for $|m| > 1/2$, which leads to zero transverse g-factor components (the longitudinal component $g_{zz} = 2|m|$). As a result, local magnetic fields B_L of surrounding nuclei do not destroy the z component of the mean nuclear spin $\vec{I}_N$ even in zero magnetic field [24]. A well-known example [9] is the increase of spin relaxation time of As nuclei due to the magnetodipole interaction up to $T_2' \sim T_2/g_\perp^2$ in an AlGaAs solid solution, where $T_2 \sim (\gamma B_L)^{-1} \approx 100$ μs, transverse the g factor Lande for nuclear quasiparticles $g_\perp \sim 0.1$ results from a small deviation of the EFG tensor from axial symmetry. Time $T_2' \sim 10$ ms becomes comparable with the spin-lattice relaxation time. Therefore, the stabilizing effect of QI should be carefully analyzed in *any* (self-organized) quantum dot system having nuclei with large spin $I > 1/2$. For example, the QI may explain qualitatively the zero-field nuclear polarization in InP [5] and InAs [7] dots as well as the surprising long-term conservation of spin polarization in the electron-nuclear spin system of InGaAs QD [6].

Under optical orientation conditions the mean spin of quadrupole-split nuclei is $\vec{I}_N \propto (\vec{S}\cdot\vec{n})\vec{n}$, with its direction being determined by the unit vector $\vec{n}$ along quadrupole axis rather than by the external magnetic field $\vec{B}$. These nuclei create a fixed-in-space effective magnetic field $\vec{B}_N^Q = a(\vec{S}\cdot\vec{n})\vec{n}$ acting on the QD electron spin (below a is considered as a phenomenological fitting parameter). The nuclear field $\vec{B}_N^Q$ quenches the electron spin magnetic depolarization and enables one to explain threefold broadening of the Hanle curve in the presence of the nuclear polarization [Fig. 2(a)]. Within the simplest model the steady state average electron spin $\vec{S}$ in the InP islands is governed by the Bloch equation

$$\frac{\vec{S}_0 - \vec{S}}{T_s} + \frac{\mu_B g}{\hbar}[\vec{B} + \vec{B}_N^Q] \times \vec{S} = 0, \qquad (1)$$

where $\vec{S}_0 \| [001]$ is the zero-field mean spin; spin lifetime T_s takes into account phenomenologically all contributions coming from spin relaxation and recombination [16]; the second term of Eq. (1) describes spin precession [Fig. 2(b)] in the sum of external ($\vec{B} \perp [001]$) and nuclear ($\vec{B}_N^Q \| \vec{n}$) magnetic fields (below we put $\vec{n} \| [001]$). We neglected the contribution into the nuclear field from phosphorous nuclei whose spin ($I = 1/2$) is 9 times less than that of indium nuclei. The PL degree is $\rho_c \propto S_z = (\vec{S}\cdot\vec{n})$. Figure 2(c) shows the dependence $S_z(B)/S_0$ calculated from Eq. (1) in the absence ($a = 0$, solid curve) and in the presence ($aS_0 = 5B_{1/2}$, dashed curve) of the nuclear field. It is seen that the nuclear field of QI-perturbed nuclei does stabilize electron spin and broaden the Hanle curve by 3 times. Thus

the experimental data can be explained by the nuclear quadrupole interaction within the standard approach [9] worked out for bulk semiconductors. Bloch equation (1) is applicable when the resident electron dwell time (correlation time) within the QD is short: electron spin has no time to make a turn about a random nuclear field. Such a situation can be easily realized under optical excitation, when the electron spin is renewed fast due to recombination or photoinduced spin relaxation processes.

Finally, we show that the QI will also stabilize the spin of the electron that spends a relatively long time τ_c before its renewal (time τ_c should be much longer than the precession period $\hbar N/A \sim 1$ μs of nuclear spin in the Knight field). For the nonpolarized nuclei case, the first step of electron spin dephasing in a quasistatic random nuclear field remains in the presence of QI, too. Indeed, the transverse (x, y) spin components of quadrupole-split nuclei rotate about z axis with characteristic angular frequency $2\pi\nu_Q \sim 10^7\ \mathrm{s}^{-1}$. This is much slower than the electron spin precession frequency $A/\hbar\sqrt{N} \sim 10^9\ \mathrm{s}^{-1}$ in random nuclear field with $N \sim 10^5$ [1], so that the nuclear field is still "frozen" and distributed isotropically at this time scale. However, the second step—the decoherence due to the Knight-field-induced precession of the nuclear spin (and the corresponding nuclear field evolution) with frequency $A/\hbar N \sim 10^6\ \mathrm{s}^{-1}$ [1] will be suppressed because the (x, y) components of nuclear spin rotate about z axis much faster than about the Knight field. Quantum mechanically one can say that the transverse components of the Knight field do not split in the first order $+m$ and $-m$ projections of a given nuclear spin for $|m| > 1/2$. Doublet degeneracy remains. As a result, the z component of both nuclear and electron spin is conserved much longer than 1 μs [1].

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