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Suppression of Overhauser effect in the exciton-nuclear spin system of GaAs quantum dot

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The spin of a single electron localized on monolayer-high fluctuation of quantum well width (quantum dot) interacts with macroscopic number ($\sim 10^5$) of nuclear spins. As a result the tight-binding electron-nuclear spin system (ENSS) develops in nanostructures [1, 2]. Due to rather long spin relaxation time the nuclear spin could be used (at least in principle) for information storage and to control the electron spin. For example, the hysteresis phenomena have been found in ENSS of quantum wells [3].

Here we show that the presence of a hole in the quantum dot (i.e. exciton instead of a single electron) strongly affects the magnetic properties of the unified exciton-nuclear spin system (ExNSS). In spite of the absence of contact interaction between hole and nuclei the hole affects the ExNSS properties via the exchange coupling with electron. The interaction splits the quartet of exciton spin levels into pair of optically allowed states (bright excitons: electron and hole spins are antiparallel) and pair of optically forbidden states (dark excitons: electron and hole spins are parallel) [4]. This splitting (δ_0) suppresses the Overhauser effect. Indeed, an amount of energy δ_0 should be compensated to satisfy the energy conservation law under mutual electron-nuclear spin flip transitions. Under these conditions the role of dipole–dipole interactions between nuclei becomes anomalously large in comparison with bulk GaAs. This interaction destroys the nuclear polarization and acts as a leakage of nuclear spin. The external magnetic field eliminates the leakage. However, as a result of suppression of Overhauser effect the characteristic field value restoring the nuclear polarization is two orders in magnitude larger than that for the bulk GaAs.

Near-field magneto-photoluminescence spectroscopy [5, 6] has been used to study the ExNSS in single GaAs quantum dot. This method allows one to measure the splitting of exciton radiative levels and its magnetic field dependence under circularly polarized excitation directly. It is found that the splitting is minimal when external field compensates the internal nuclear magnetic field $B_N = 1.3$ T which is proportional to the nuclear polarization ($\sim$65%). This shifts the magnetic field dependence as a whole by the B_N value. The exception takes place in low magnetic fields $B < 1$ kG where the nuclear polarization is strongly reduced (the splitting is minimal for $B = 0$, too). The field range of the reduction is 300 times larger than that for bulk GaAs ($\sim$3 G [7]).

To explain the unexpected behavior of ExNSS we have deduced the average nuclear spin polarization determined by the balance between dynamical nuclear polarization (Overhauser effect) and depolarization (leakage). These processes are governed by fluctuations of the electron and nuclear polarization from their average values.

Polarization of the nuclear spins by optically oriented excitons arises from the "flip/flop" processes involving the simultaneous spin flip of a nucleus and electron. The electronic spin flip causes the exciton to transform between bright and dark states and requires emission or absorption of energy ($\delta_0 \approx 100\,\mu$eV) because of the large mismatch in electronic and nuclear Zeeman energies. Therefore some additional "assisting" process is necessary to satisfy energy conservation (e.g. emission or absorption of a photon or a phonon). Transition rates for flip/flop transitions calculated in second order perturbation theory are (δ_s^2/δ_0^2) smaller than those for the assisting processes with no spin flips, where $\delta_s \propto A/\sqrt{N}$ (A is hyperfine constant, N number of nuclei in the dot) is the splitting of electron spin states in a random nuclear magnetic field. If the dark exciton lifetime (τ_d) is much longer than the bright exciton lifetime (τ_b), nuclear polarization is determined by spin–flip assisted radiative recombination of the dark excitons [8]. We calculate the nuclear spin polarization rate to be $T_e^{-1} = (\delta_s^2/15N\delta_0^2)\tau_b^{-1} \approx 2.5\,\mathrm{s}^{-1}$ for $N \sim 10^5$ nuclei in the quantum dot [5] and $\tau_b \sim 0.1$ ns [9]. The estimation is in agreement with experiment ($T_{exp} \approx 3$ s).

Coupling of neighboring nuclear spins through the dipole–dipole interaction leads to the leakage of nuclear polarization. In a magnetic field B larger than the dipole field, $B_L \sim 1.5$ G, the energy of the nuclear dipole–dipole interaction is not enough to drive the transition between two nuclear spin sublevels split by the Zeeman energy. However, fluctuations of the z-component of the hyperfine magnetic field of the electrons acting on the nuclei (the mean value of the electron field determines the Knight shift in NMR [10]) can provide the necessary energy, leading to depolarization of the total nuclear spin, even in a relatively strong magnetic field. Calculations show that the nuclear dipole-dipole interaction weakly mixes the wave functions of different nuclear spin projection states, of order B_L/B. Transitions between these mixed states are induced by the hyperfine magnetic field of the electrons acting on the nucleus during the exciton lifetime. The dark exciton plays the main role in this process because $\tau_d \gg \tau_b$. This leads to a nuclear spin depolarization with a rate calculated in second order perturbation theory to be $T_{e-dip}^{-1} = 0.06\tau_d(\delta_s/\hbar)^2(B_L/B)^2N^{-1}$. Increasing of external field eliminates the role of leakage of nuclear spin. The polarization rate (Overhauser effect) and leakage rate becomes comparable when $T_e = T_{e-dip}$, i.e. at $B = \sqrt{\tilde{\xi}}B_L$. Parameter $\sqrt{\tilde{\xi}} \approx (\delta_0\sqrt{\tau_d\tau_b}/\hbar)$ is much greater than 1 in the experimental conditions. The steady state value of a mean nuclear spin

$$\langle I_z\rangle = \frac{I(I+1)}{S(S+1)}\langle S_z^d\rangle\frac{B^2}{B^2+\tilde{\xi}B_L^2}.$$

Here I, S are the nuclear and electron spins. A good agreement with experiment takes place when polarization of dark excitons $\langle S_z^d\rangle = 0.2$ and $\sqrt{\tau_d\tau_b} = 3.5$ ns.

Thus, the nuclear spin polarization tracking the electron spin polarization of the dark exciton does not depend on external magnetic field except in the strongly depolarized region around zero applied field with a Lorentzian lineshape with width that is two orders of magnitude greater than in bulk GaAs.

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