OPTICAL ORIENTATION AND SPIN POLARIZATION OF THE ELECTRONS AND NUCLEI IN SEMICONDUCTOR QUANTUM DOTS

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A short review of the results concerning the optical orientation and spin polarization effects in semiconductor quantum dots obtained by a group of investigators of the Department of Physics and Measurement Technology of the Linkoping University in Sweden with the participation of Evgenii S. Moskalenko is written below in his memory.

The possibility of polarizing the nuclei in a metal by an electron spin system in equilibrium was first pointed out by Overhauser [1]. The spin polarization effects of the localized electrons in semiconductor quantum dots (QDs) can be understood on the base of investigations initiated by Dyakonov and Perel [2] and prolonged in a series of theoretical [3-8] and experimental investigations [9-12], which elaborated the basic concepts in this field. They were performed and prolonged in numerous further investigations including [13-18]. Most of these effects belong to the optical orientation phenomena arising under the action of the circularly polarized laser radiation including the spin polarization of electrons and nuclei and optical alignment of excitons in semiconductors.

The interest in electron spin polarization effects in QDs is due to the fact that they are not subjected to classical spin relaxation mechanisms known for free carriers [9]. The recent theoretical studies [2-7] predict that the dominant mechanism of electron spin relaxation in QDs at low temperatures is due to its hyperfine interaction with nuclei.

The comparison with the experiments confirms this point of view. The long spin dephasing time of an electron in a QD suggests elaborating a model of quantum bit on this ground [9]. The influence of the electron spin polarization on the spins of nuclei takes place through the hyperfine Fermi constant interaction, which is described by Hamiltonian (2) in [4]:

$$H_{hf} = \frac{\mathrm{v}_0}{2}\sum_j A^j \left|\Psi(\vec{R}_j)\right|^2 \left(\hat{I}^j_\alpha \cdot \hat{\sigma}_\alpha\right), \quad (1)$$

where the sum goes over all nuclei in the QD, v_0 is the volume of the unit cell, $\Psi(\vec{R}_j)$ is the electron envelope wave function at the *j*-th nucleus, $\hat{I}_\alpha$ and $\hat{\sigma}_\alpha$ are the spin projections on the coordinate axes $\alpha = x, y, z$. The electron spin in a QD interacts with a large but finite number of nuclei $N_L \approx 10^3 - 10^5$ [9]. The constant of the Fermi contact interaction A^j has the expression [4]:

$$A^j = \frac{16\pi\mu_B\mu_j}{3I^j}\left|U_c(\vec{R}_j)\right|^2. \quad (2)$$

It includes the Bohr magneton μ_B, the nuclear magnetic moment μ_j, and the periodic part

$U_c(\vec{R}_j)$ of the electron Bloch wave function at the point $\vec{R}_j$. As a result of the nuclear spin polarization, a nuclear hyperfine magnetic field $\vec{B}_N$ arises. It is known as Overhauser field. Its value $\vec{B}_N$ can be determined from equation (3) in [4]:

$$\mu_B g_e \vec{B}_N = v_0 \left\langle \sum_j A^j \left|\Psi(\vec{R}_j)\right|^2 \vec{I}^{\,j} \right\rangle. \tag{3}$$

The symbol $\langle\ \rangle$ denotes the quantum mechanical average over the nuclei wave functions, and g_e is the electron Lande *g*-factor. The magnitude and direction of this field are randomly distributed over the ensemble of QDs. The density distribution function has a Gaussian probability [4]:

$$W(B_N) = \frac{1}{\pi^{3/2}\Delta_B^3} \exp\left[\frac{-B_N^2}{\Delta_B^2}\right]. \tag{4}$$

The parameter Δ_B determines the dispersion of the density distribution.

The equation of motion of the spin $\vec{S}$ in a fixed magnetic field is given by equation (9) in [4], which describes the coherent electron spin precession in a single QD due to the magnetic field $\vec{B}_N$ oriented in the unit vector $\vec{n}$ direction giving rise to the precession frequency ω_e equal to

$$\omega_e = \frac{\mu_B g_e B_N}{\hbar}; \qquad \vec{n} = \frac{\vec{B}_N}{B_N}. \tag{5}$$

Each electron spin is moving in the frozen fluctuation of the nuclear hyperfine magnetic field $\vec{B}_N$ in its own QD. But these fields are randomly distributed among the dots of the ensemble. Averaging the coherent precession over the magnetic field distribution (4) the ensemble averaged time dependence (10) in [4] was obtained:

$$\left\langle \vec{S}(t) \right\rangle = \frac{\vec{S}_0}{3}\left\{1 + 2\left[1 - 2\left(\frac{t}{T_\Delta}\right)^2\right]\exp\left[-\left(\frac{t}{T_\Delta}\right)^2\right]\right\}, \tag{6}$$

where $\vec{S}_0$ is the initial spin. It consists of two terms. One of them is a constant $\vec{S}_0/3$, whereas the other term describes the Gaussian decreasing dependence with the dephasing time T_Δ related with the dispersion parameter Δ_B by formula (11) in [4]:

$$T_\Delta = \frac{\hbar}{\mu_B |g_e| \Delta_B}. \tag{7}$$

This spin dephasing mechanism in the frozen fluctuation model with the inhomogeneous broadening of the local magnetic fields B_N is termed as spin relaxation [2-5, 9]. For repeated measurements on a single quantum dot the hyperfine interaction has the same effect as for an ensemble with the same time dependence. The Overhauser field in its turn influences the electron spin. This influence is measured by the Zeeman effect with energy $\hbar\omega_e$ known as Overhauser shift [2, 3]. The electron spin creates its own magnetic field $\vec{B}_e$ known as Knight field [2, 3]. Merkulov, Efros, and Rosen [4] analyzed three main processes, which determine the electron spin relaxation. One of them is the precession of the electron spin around the Overhauser hyperfine field with the frequency ω_e (5). The second process is the precession of the nuclear spin around the Knight field $\vec{B}_e$. Its frequency ω_n is determined by

$$\omega_n \approx \frac{B_e \mu_n}{\hbar}, \quad \mu_n = \hbar \gamma I,$$

where μ_n is the nuclear magnetic moment, γ is the gyromagnetic ratio, and I is the nuclear spin. The third process is determined by the precession of the spins of nuclei around the magnetic fields created by another neighbour nuclei. This process is due to the dipole-dipole interaction between the nuclear spins and can be characterized by the frequency ω_{d-d}.

Supposing the number of nuclei in a QD equal to $N_L \cong 10^5$ in [9] the values $\omega_e \approx 10^{10} s^{-1}$, $\omega_h \approx 10^7 s^{-1}$ and $\omega_{d-d} \approx 10^3 s^{-1}$ were estimated. These so different frequencies permitted introducing the adiabatic hypothesis into the calculations considering that the electron precession takes place under the condition of the frozen nuclear spins, because they do not succeed to change their orientation during the precession period of the electrons. The value of the Overhauser shift with the precession frequency $\omega_c \approx 10^{10} s^{-1}$ is two order of magnitude smaller than the thermal energy kT at $T = 10\,\mathrm{K}$. The possibility to observe the hyperfine electron-nuclei interaction effects under these conditions is a wonderful event in condensed matter physics. It is due to the small spin-spin nuclear interaction, which is equivalent to a very low effective nuclear spin temperature in comparison with the lattice temperature related with the orbital motion of the nuclei and with their phonon branches of the spectrum.

Braun et al. [9] have investigated the spin time dynamics of positively charged X^+ excitons in InAs/GaAs QDs by cw and time-resolved photoluminescence (PL) spectroscopy. The excitons X^+ consist of one electron and two holes forming a spin singlet. The exchange interaction between the electron and the holes is canceled in the X^+ ground state. Under these conditions, the analysis of the circular polarization of the X^+ luminescence probes directly the spin polarization of the electron. If the excitation pulse is circularly polarized, for example σ^+, and the luminescence intensity is recorded in the copolarized I^+ and in the counterpolarized I^- geometry as regards the excitation laser, then the degree ρ_c of the PL circular polarization is determined as [9]:

$$\rho_c = \frac{I^+ - I^-}{I^+ + I^-}. \tag{8}$$

Merkulov et al. [4] and Semenov et al. [6] have predicted that the electron spin dephasing induced by the inhomogeneous broadening of the nuclear field B_N can be suppressed in an external magnetic field $\vec{B}_{ext}$, which must be larger than the dispersion Δ_B. In Ref. [9] assuming the electron *g*-factor as $|g_e| = 0.8$ [10] the dispersion Δ_B was determined as equal to 28 mT, whereas the dephasing time T_Δ as 500 ps. It was shown that the spin polarization decays within the first 800 ps down to 1/3 of its initial value. After that, the polarization remains stable with no measurable decay on the radiative lifetime scale [9], in full accordance with the theoretical description [4]. The application of a small external magnetic field $\vec{B}_{ext} =$ 100 mT is very unusual in nonmagnetic semiconductors, as was underlined in [4]. As was mentioned in [9], the photogenerated electrons in the undoped QDs feel a strong effective magnetic field due to exchange interaction with the holes [9, 10]. This exchange field has the order of magnitude of a few Teslas, much stronger than the Overhauser fields. In the case of X^- excitons with two electrons and one hole, the total spin of electrons is zero and no effect of hyperfine interaction was observed [9, 11]. The hole spin in *p*-type valence band does not interact with nuclei [9, 12]. In a series of papers published by the Linkoping University group of investigators [13-18] it was established that even for the case of neutral excitons, a nonzero nuclear magnetic field builds up in the In(Ga)As/GaAs QDs. To explain these experimental observations, the exciton formation in a QDs was considered as a process of separated capture of electrons and holes, rather than a mechanism according to which the electron and hole are captured into the QD as an entity. This variant takes place in experiments where the electrons and holes are created in the wetting layer (WL) on the surface of which the QDs are situated, and not directly into the QDs. The charge carriers undergo a transport in the WL prior to the capture into the QDs. The transport and capture processes are characterized by the parameter $\Delta\tau_{e-h}$, which denoted the difference in the capture times of electrons and holes into the QDs. These processes taking place under the influence of electric and magnetic fields were studied in [19-38]. According to [16-18], the electrons are captured first because of their smaller effective mass. The efficiency of the dynamic nuclear polarization by spin-oriented electrons depends on the fraction of time $\Gamma_e = \Delta\tau_{e-h}/\tau_{rc}$, where τ_{rc} is the recycling time, i.e., the average time between two adjacent events of exciton formation in QDs. $\vec{B}_N$ and ρ_c are predicted to be directly proportional to τ_{rc}^{-1} as in fact was observed in experiments. Electron is alone in the QD during $\Delta\tau_{e-h}$ and can then polarize the nuclei. Within a certain number of recycling events, the nuclear field builds up. As a result, the electron spin $\vec{S}$ is influenced not only by a “destructive” magnetic field $\vec{B}_{exch}$ arising due to the exchange electron-hole interaction, but also by the “stabilizing” nuclear field $\vec{B}_N$ [13-17]. It is the joint influence of both of these factors that determine the value of the projection of $\vec{S}$ onto the *z* axis, and hence ρ_c, as measured in the experiment. The present model is very similar to that developed for the exciton pseudospin and the role of $\vec{B}_{ext}$ is played by $\vec{B}_N$. The model adopted in [13-17] based on the separate capture of electrons and holes into the QDs is widely accepted, as one can see from [39, 40]. The studies of the denser ensembles of the same-type QDs performed in crossed internal electric and external magnetic fields, have directly demonstrated that the capture process of electrons and holes into the QDs should in fact be separately considered [15, 16]. In [13-17] a high degree of spin polarization is achieved for the neutral exciton at a zero external magnetic field in individual InAs QDs excited by circularly polarized laser radiation and studied by micro-photoluminescence (μ-PL) methods.

A high degree of polarization $\rho_c \approx 60\%$ observed in the case of a neutral exciton is a remarkable event, because it is well known that the bright states $|+1\rangle$ and $|-1\rangle$ of the heavy-hole neutral exciton are mixed due to in-plane asymmetry of a QD. The electron-hole pair state $|+1\rangle$ is created by electron spin projection $|s_z\rangle = |-1/2\rangle$ and heavy-hole moment projection $|j_z\rangle = |+3/2\rangle$, whereas the state $|-1\rangle$ by the states $|s_z\rangle = |+1/2\rangle$ and $|j_z\rangle = |-3/2\rangle$. The states $|+1\rangle$ and $|-1\rangle$ are mixed forming two linearly polarized states $|x\rangle$ and $|y\rangle$, which emit light along the $\langle 110\rangle$ and $\langle 1\underline{1}0\rangle$ crystallographic directions and are split by the anisotropic e-h exchange interaction energy $\hbar\omega_{exch}$. At external magnetic field $\vec{B}_{ext} = 0$, the neutral exciton is expected to reveal two orthogonal linearly polarized components of the PL spectrum and only at elevated values of $\vec{B}_{ext}$ parallel to *z* axis the mixed states $|x\rangle$ and $|y\rangle$ will transform into the pure $|+1\rangle$ and $|-1\rangle$ states giving rise to circular-polarized emission [13].

Upon excitation with linearly-polarized light σ^x at $\vec{B}_{ext}=0$, two orthogonal and linearly polarized PL lines separated by $\hbar\omega_{exch}=25\,\mu\text{eV}$ were in fact monitored [13, 14]. Following the exciton pseudospin model proposed in [8, 41] the anisotropic e-h exchange interaction is viewed as an in-plane magnetic field $\vec{B}_{exch}$ perpendicular to axis *Oz*. The precession period of the exciton pseudospin around the field $\vec{B}_{exch}$ equals to $T_{exch}=2\pi/\omega_{exch}\approx 160\text{ ps}$ [13, 14].

Taking into account that the exciton decay time equals $\tau_d=800\text{ ps}$, one may conclude that the exciton pseudospin will accomplish many turns around $\vec{B}_{exch}$ before recombination. Hence ρ_c being proportional to the projection of the exciton pseudospin onto the *z* axis is predicted to be negligible. An external magnetic field of sufficient strength applied in Faraday geometry $\vec{B}_{ext}\parallel Oz$ will restore the polarization of the neutral exciton because of the decoupling of the $|+1\rangle$ and $|-1\rangle$ states. This decoupling in the condition $\left|\vec{B}_{ext}\mu_B g_{ex}\right|>\hbar\omega_{exch}$, where g_{ex} is the neutral exciton *g*-factor and $\mu_B=58\,\mu\text{eV/T}$ is the Bohr magneton, will give rise to nonvanishing values of ρ_c. In the vector model [8, 41, 13] the application of $\vec{B}_{ext}$ initiates the precession of the exciton pseudospin around the total magnetic field $\vec{B}_{tot}=\vec{B}_{ext}+\vec{B}_{exch}$ as is represented in Fig. 1 in [13]. This precession leads to a nonzero value of $\left|\vec{S}_z^{ex}\right|$ and hence to $\rho_c\neq 0$. Despite $\vec{B}_{ext}=0$, at the circularly polarized excitation of the InAs QDs, the obvious energy separation of about 50 μeV was recorded between the σ^+ and σ^- components of the PL spectrum [13]. It was the signature of the appearance of an effective magnetic field $\vec{B}_N$ in a sample with a projection onto the *z* axis. When the excitation helicity was reversed the polarization-resolved PL components exchange their spectral positions, clearly demonstrating that $\vec{B}_N$ has reversed its direction. The contribution of $\left|\vec{B}_N^z\right|$ to the Zeeman splitting was estimated as 42 μeV. The build-up time of the $\vec{B}_N$ was measured in [13, 14] using the light with alternating σ^+ and σ^- polarizations with frequency *f*, whereas the detection of the PL components was performed within the time intervals corresponding to only σ^+ and σ^- excitation windows. The polarization degree ρ_c remains approximately the same for $f\lesssim 100Hz$, and the time $\Delta\tau\simeq 10\text{ms}$ was sufficient for $\vec{B}_N$ to be build-up. The value $\left|\vec{B}_N^z\right|$ determining ρ_c was estimated to be on the order of a few Teslas. To understand the origin of $\vec{B}_N$ in [13] a concept of dynamical polarization of lattice nuclei by optically oriented electrons was employed following [1-7] discussed above. As in the case of X^+ exciton studied by Braun et al. in [9], in the case of X^0 exciton in the In(Ga)As/GaAs QDs, the interaction between a single electron and a large number 10^4-10^5 of nuclei results in a dynamical nuclear spin polarization leading to the appearance of the spin-oriented nuclei. As was mentioned above, the equivalent effective magnetic field $\vec{B}_N$ acting on the electron localized in the QD gives rise to the Overhauser shift defined as $\Delta E=\left|B_N^z\mu_B g_{ex}\right|$. Adopting $|g_{ex}|=0.5-0.6$ and

$\Delta E = 42\,\mu\text{eV}$ in [13], the value $\left|\vec{B}_N^z\right| = (1.2-1.4)\ \text{T}$ was evaluated to be in satisfactory agreement with previous estimations [42, 43]. To explain the experimentally observed fact that a relatively strong nuclear magnetic field was build-up in the QD even for the case of a neutral exciton at $\vec{B}_{ext} = 0$, in [13-15] the preceding steps to the formation of the exciton were considered. They are related with the processes of separate capture of electrons and holes as was demonstrated in many previous publications [15, 19-30]. The capture times $\tau_{e(h)}$ for electrons and holes excited into the wetting layer (WL) could be estimated as $\tau_{e(h)} = L_{e(h)} / V_{e(h)}$. Here $L_{e(h)}$ corresponds to collection lengths for individual charged carriers into the QD. V_e and V_h are their velocities in the WL plane. As was deduced earlier [12], they are $V_e = 1.6\cdot 10^7$ cm/s and $V_h = 3.1\cdot 10^6$ cm/s. For experimental conditions with the QD located within the area of the laser spot, the value $L_e = L_h = 1\,\mu\text{m}$ can be assumed and hence the difference in the capture times $\Delta\tau_{e-h}$ equals 26 ps. It is an expected time rise for the PL in the QD. According to this concept adopted in [13-15] before the recombination of an exciton, the QD is assumed to be populated with only one electron for 26 ps. This value is smaller than the average time τ_{rc} between two subsequent exciton formation events. The recycling τ_{rc} is supposed to be comparable with the exciton decay time $\tau_d \approx 800$ ps. When the parameter $\Delta\tau_{e-h}$ is decreased by exciting directly in the QD with the photon energy smaller than excitation energy of the WL, the gradual reduction of ρ_c was observed [13-15]. The nuclear spins being polarized continue to conserve their polarization at least 10^{-3} s and in this period of time the Overhauser magnetic field is build-up being continuously supplied by other spin polarized electrons appearing successively in the QD under the influence of the circularly polarized laser radiation. A strongly coupled electron-nuclear spin system gives rise to two effective magnetic fields: a nuclear Overhauser field $\vec{B}_N$ acting upon the electrons ant the electron magnetic field $\vec{B}_e$ called the Knight field acting upon the nuclei [2-4]. The typical local effective magnetic fields $\vec{B}_L$ have the magnitudes of about $B_L \approx (0.1-0.3)$ mT [2-4]. They are caused by the nuclear spin-spin interactions and lead to the disappearance of the optically oriented nuclear spins. If $\left|\vec{B}_e\right| > \left|\vec{B}_L\right|$ a dynamic nuclear polarization is predicted at $\vec{B}_{ext} = 0.$ To determine the value of $\left|\vec{B}_e\right|$ one can represent the average interaction energy of an electron spin $\vec{S}$ with N nuclei as the nuclear spin energy in an effective electron field $\vec{B}_e$ as follows:

$$A\left(\vec{I}_{av}\cdot\vec{S}\right) = -N\hbar\gamma\left(\vec{I}_{av}\cdot\vec{B}_e\right);\ \vec{B}_e = -\frac{\vec{S}\cdot A}{N\hbar\gamma};\ B_e = b\left|S_z\right|;$$
$$b = \frac{A}{N\hbar\gamma};\ \mu_n = \hbar\gamma I. \qquad (9)$$

Here γ is the gyromagnetic ratio, μ_n is the magnetic moment of nucleus with spin *I*. I_{av} is the average spin of nuclei, *A* is the hyperfine contact interaction constant.

For the parameters $N = 5\cdot 10^4$, $A = 56\,\mu\text{eV}$, γ=$5.86\cdot 10^7$ $\text{T}^{-1}\text{s}^{-1}$ the constant *b* was estimated as $b = 30$ mT. At $\left|S_z\right| = 1/2$ the Knight magnetic field equals $B_e = 15$ mT, which is well above the typical magnitude of B_L. This fact justifies the assumption that an essential $\left|\vec{B}_N\right|$ could be developed in the QD at $\vec{B}_{ext} = 0$ under the conditions described in [13-18].

To better understand the photoluminescence (PL) from the self-assembled InAs QDs (QDs) its dependence on some parameters, such as excitation power, pump-photon energy, crystal temperature [19-22], and the role of acceptor impurities [23, 24] were investigated.

Two new lines red-shifted relative to the ground state energy were identified as charged exciton complexes X^- and X^{--}.

It was observed that the PL spectra are very sensitive to the excitation energy and crystal temperature due to the effective diffusivity of photogenerated particles, which depends essentially on the excess excitation photon energy over the barrier band gap energy. The oscillating dependence of the QD exciton PL intensity on the photon excitation energy was explained in terms of the phonon relaxation processes [19-22]. The excess energy of the photogenerated carriers is released by the emission of LO phonons. If the excess energy exactly fits with an integer number of $\hbar\omega_{LO}$ phonons, it is quickly released, whereby the carriers become motionless at the band gap edge energy shortly after generation. It leads to low effective diffusivity. The carriers with higher diffusivity are moving in a spatially large region and can achieve the capturing centers. Due to different masses the excess energy is distributed in a non-equal way between the electrons and the heavy-holes, which results in different diffusivities. The capture centers primarily trap the high diffusivity carriers. Subsequently the attraction of low diffusivity carriers of opposite charge is increased by the Coulomb interaction [19-22]. The acceptor-induced threshold energy for the optical charging of InAs single QDs was observed [23, 24]. This threshold energy lies in the interval between the wetting layer ground state energy and the GaAs barrier energy and it is ascribed to the quantum transitions between the acceptor states and the conduction band states in GaAs [23, 24].

The use of an infrared laser excitation in addition to the optical excitation permitted to effectively manipulate the emission efficiency of the QDs [25-27]. At low temperatures the supplementary charges created by the infrared laser lead to the screening of the built-in electric fields, which play an important role in the carrier capture mechanism. The influence of the separate generation of the excess electrons and holes into the QDs at dual-laser excitation was investigated in [31, 32]. New interesting results concerning the properties of the self-assembled InAs/GaAs QDs were obtained subjecting the samples to the action of the external lateral electric field as well as of the magnetic field perpendicular and parallel to the wetting layer (WL) [32, 38]. When an external lateral electric field was applied, a considerable enhancement of the PL intensity of the QDs was observed. This severalfold increase depends on the magnitudes of the external electric field and the internal built-in electric fields. The latter could be altered by an additional infrared illumination of the sample. At low temperatures the magnetic field perpendicular to the layer alters the in-plane transport properties due to the localization of the carriers in the WL potential fluctuations. The decreased transport in the WL leads to a reduced capture in the QDs and consequently to a weakened dot-related emission. In addition, with increasing fields oriented parallel to the growth direction and perpendicular to the layer, i.e., in Faraday geometry, the redistribution of the intensities of the spectral lines was observed.

Conclusions

It has been established that, even for the case of neutral excitons, a nonzero nuclear magnetic field is building up in the In(Ga)As/GaAs QDs when the electrons and holes are photo-generated on the surface of the wetting layer and not directly into the QDs [13-18]. To explain these experimental observations, the exciton formation in QDs was considered as a process of the separated capture of electrons and holes, rather than a mechanism according to which an electron and a hole are captured into the QD as an entity [19-38].

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