Spin synamics in quantum dots

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1. Introduction

Spin dynamics of carriers in semiconductors attracts considerable attention last four decades. The early studies of the spin dynamics are reviewed in Ref. [1.1] where the optical orientation of the spins and the spin relaxation mechanisms in bulk semiconductors are discussed in details. More recent reviews are present in Refs. [1.2, 1.3]. Optical orientation is the most-used method of spin polarization up to now. As an alternative, very promising method, we can mention so called spin injection and other spin-dependent electron transport effects [2.1–2.10] which are expensive developed now.

Last two decades, studies of spin dynamics were stimulated by invention of the low-dimentional heterostructures, in particular of the quantum dots (QDs). In the QDs, the spin dynamics is considerably modified [3.1–3.15]. First, one of the most effective relaxation mechanisms, namely Dyakonov-Perel mechanism [1.1], is suppressed due to the three-dimentional confinement of carriers. On the other hand, localization of the carriers in the QDs results in several other effects. E.g., electron-hole exchange interaction is considerably enhanced that results in simultaneous relaxation of the electron and hole spins. Anisotropic part of the interaction appearing due to anisotropy of real QDs causes relative fast relaxation of the spins because of splitting the exciton (electron-hole) basic states into linearly polarized eigenstates. Hyperfine interaction of the electron spins with those of nuclei is also enhanced which were evidenced by several interesting studies [4.1–4.5]. Theoretical estimates show [3.1] that the spin relaxation time for a single carrier is governed mainly by interaction with phonons and nuclei and may be very long.

One more interesting matter for study is the spin dynamics in the charged QDs, namely positively or negatively charged trions in the QDs consisting of two holes and one electron or two electrons and one hole, respectively [5.1–5.5]. The trions can be created optically in the singly charged QDs. Excitation of the dots by circularly polarized light allows one to polarize spins of resident carriers and to study their relaxation time.

Simultaneously with development of the QD fabrication technology, different kind experimental techniques were developed which allow one to investigate spin dynamics in many details. Historically, a method based on the Hanle effect was extensively used as the main method for this purpose [1.1, 1.3]. However last two decades, modern techniques of the photoluminescence (PL) kinetics measurements [3.1–3.15] as well as the Faraday or Kerr rotation, polarized pump-probe and four-wave mixing measurements [6.1–6.5] with picosecond (or even sub-picosecond) time resolution are developed which are capable to supply with direct information about spin dynamics in ensemble of QDs in real time.

Success in the QDs fabrications and experimental studies of spin dynamics together with the very promising theoretical prospects about spin memory and spin manipulations give rise to "dream" of new branch of electronics, called "spintronics" where the electron spin may be used for information storage and processing. Details of this activity can be found in many papers (see, e.g., [1.2, 1.3, 7.1–7.7]). We should also mention that "spintronics" has stimulated growing activity in studying of spin dynamics in the bulk material and in the quantum wells where new interesting results have been obtained (see, e.g., [8.1–8.5]).

In this work, we consider several examples of experimental study of spin dynamics in real time for so called self-assembled QDs which are widely studied due to their high quality and possibility to integrate in semiconductor devices. We restrict ourselves to PL kinetics measurements which have used in most publications due to their relative simplicity, high sensitivity as well as selectivity to signal from a QD layer. We discuss two types of experimental studies: 1) spin quantum beats (QBs) in neutral and charged QDs and 2) relaxation of the polarized spins of resident electrons in QDs. These examples allow one to understand physics underlying the spin dynamics in QDs.

All the experimental data which we discuss here in details are obtained for InP QDs grown between $In_{0.5}Ga_{0.5}P$ barrier layers. Dots of this type are very suitable for optical studies because their photoluminescence lies in the near-infrared region (700–750 nm) where sensitive and fast photodetectors exist. Charged state of the QDs is easily varied by external voltage applied between n-doped GaAs substrate and semitransparent top electrode so that the dots could be neutral at negative bias on the top ($U < -0.5\,\mathrm{V}$), contain one electron per dot at small bias ($U \sim -0.1\,\mathrm{V}$) and several electrons at positive bias. This allows one to study spin dynamics of single electrons, electron-hole pairs, and negatively charged trions in one sample by varying of applied bias. The experimental data were obtained by means of set-up described in Refs. [3.5–3.8, 3.15] in which picosecond pulses of a laser were used for excitation and a synchro-scan streak-camera for detection of PL of the QDs.

2. Spin quantum beats

Observation of quantum beats (QBs) associated with interference between the spin states is known to be an efficient method for studying a fine energy structure and spin dynamics of carriers in quasi-2D structures. This method is attractive due to its ability to detect small energy splittings (fractions of meV) hidden within inhomogeneously broadened excitonic transitions. For observation of the QBs, at least a couple of energy states should be excited coherently by short light pulses with appropriate polarity to create a linear superposition of the states. Decay of the excitation results in QBs with a frequency which equals to difference of frequencies of optical transitions from these states.

2.1. Electron spin precession

First, we consider QBs in PL of neutral QDs where the fine energy structure of an electron-hole pair consists of the bright, $|\pm 1\rangle$, and dark, $|\pm 2\rangle$, exciton doublets [3.7, 3.14]. The doublets can be split by an external magnetic field directed across the QD layer and can be mixed by the oblique magnetic field. We consider the last case because QBs are determined under these conditions by the electron spin precession in the external magnetic + effective exchange fields. An example of the experimentally observed QBs of this type is shown in Fig. 1.

The QBs characterized by a relatively slow decay and sharp decrease in amplitude with decreasing θ, are observed in right-handed and left-handed co-circular-polarizations, and are absent in the cross-circular polarized PL. The beat frequencies in these two polarizations are practically the same in the Voigt geometry ($\theta = 90°$) and split apart upon deviation from this geometry, as shown in the Fig. 2(a). For theoretical analysis of the QBs, we used a model based on the spin Hamiltonian formalism. Transverse component of the magnetic field mixes the bright and dark excitons. The circularly polarized light can excite one of the bright (E_1 or E_2) and one of the dark (E_3 or E_4) components depending on the sign of the helicity. Interference of the excited states results in oscillations of the PL with the frequencies determined by the energy difference between the Zeeman components. Difference of energies shown in Fig. 2(a) can be easily understood if we consider the electron-hole exchange interaction as an effective magnetic field B_h affecting the electron spin. This field is directed along z-axis and is added to or subtracted from the z-component of external magnetic field depending on the hole spin orientation which is determined by the polarization of excitation.

As seen from the figure, this model adequately describes angular dependence of the beat frequentcies. Analysis shows that the dependences of both the smaller beat frequency on the tilt angle at different magnetic field strengths and on the

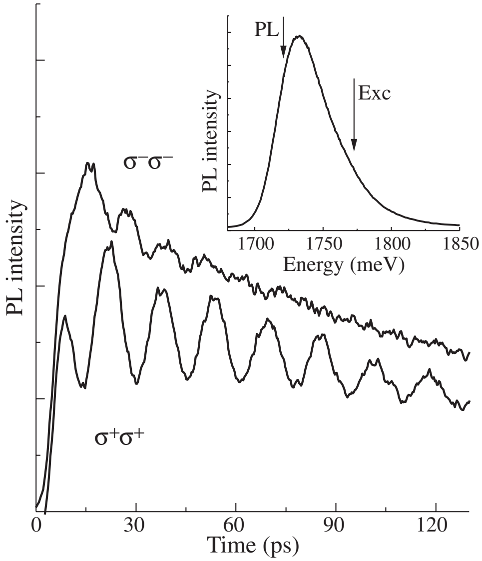

Figure 1. The co-polarized PL kinetics of InP QDs in σ^+ and σ^- polarizations at $B = 3\,\mathrm{T}$ and $\theta = 115°$. The inset shows PL band of the QDs excited above the barrier band gap. Arrows mark the excitation and PL detection energies in the PL kinetics measurements.

magnetic field strength at different tilt angles can be well fitted theoretically, which makes it possible to obtain the fine-structure parameters for the InP QDs: $g_{e,z} = g_{e,x} = g_e = 1.5$ and $\delta_0 = 0.14\,\mathrm{meV}$.

The beat amplitude strongly depends on the angle θ and the beats are not seen at $\theta<20°$. By using the procedure described in Ref. [3.7], one can obtain the theoretical expression for ratio of the beat amplitudes, $I_+(0)$ and $I_-(0)$, in the σ^+ and σ^- co-polarizations, respectively. Figure 2(b) shows the experimental as well as theoretical ratios of the beat amplitudes for different circular polarizations as a function of the tilt angle θ at $B = 3\,\mathrm{T}$. No fitting parameters were used. The good agreement gives convincing evidence of the analysis of the QBs in oblique magnetic field.

2.2. Hot trion quantum beats in charged QDs

As the negative voltage decreases, the energy of the lowest electronic level in the QDs decreases and becomes lower than the Fermi level of the GaAs substrate. In this case, electrons start to move from the doped substrate to QDs, rendering them charged. The beats shown in Fig. 1 rapidly decrease in amplitude and vanish. At

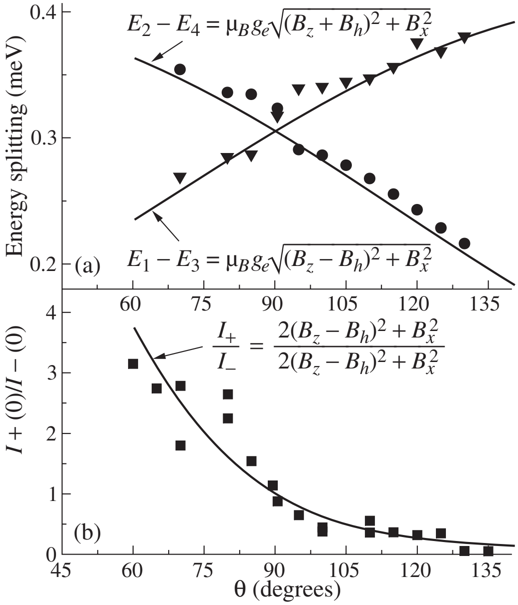

Figure 2. (a) The dependence of the beat frequency on the angle θ. The symbols are the experimenttal data and solid lines are calculations based on energy differences described. $B_z = B\cos\theta$, $B_x = B\sin\theta$, and $B_h = \delta_0/\mu_B g_e$. (b) The ratio of the beat amplitudes in two circular co-polarizations of the PL. The symbols are the experimental data, and a solid line is the calculation based on the equation described.

the same time, under certain conditions, new type of the beats arises.

At small ($U_{\text{bias}} \approx -0.2\,\text{V}$) negative voltage (slightly dependent on spectral point of PL detection), when the QDs predominantly contain one excess electron per dot, the PL kinetics exhibit well pronounced beats, as shown in Fig. 3. These beats, similar to those described above, are observed under linearly polarized excitation and under PL detection in the co- and cross-linear polarizations. The main specific feature of the beats, shown in Fig. 3, is, however, the fact that they can be observed in the absence of magnetic field. This fact, which seems, at first sight, paradoxical, was studied in detain in Ref. [3.5]. It was shown in that paper that these beats are related to interference of the states of a hot trion, comprised of a photoexcited electron-hole pair and a resident electron. In a simplified way, we can imagine that the exchange interaction with the excess electron operates as a build-in magnetic field, which splits the bright state of the exciton into the $|+1\rangle$ and $|-1\rangle$ components. The coherent excitation of these states by a linearly

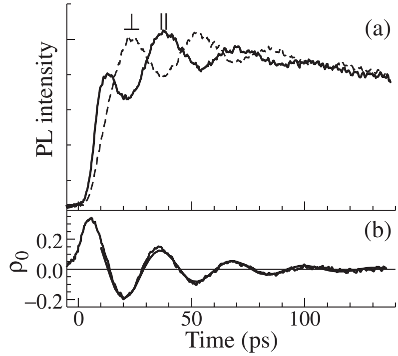

Figure 3. (a) PL kinetics in two linear polarizations for the singly charged QDs. No magnetic field is applied. $\Delta E_{\mathrm{St}} = 15\,\mathrm{meV}$, $U_{\mathrm{bias}} = -0.2\,\mathrm{V}$. (b) Temporal profile of the degree of linear polarization. Trionic QBs are clearly seen.

polarized light induces the trionic beats.

The observed period of the trionic beats $T \approx 30\,\mathrm{ps}$ can be used to estimate the exchange coupling in the trion, $\delta_{0t} \approx 120\,\mu\mathrm{eV}$. From the decay time of the beats ($\tau \approx 30\,\mathrm{ps}$), we can estimate the upper limit for the spread of the exchange splitting $\Delta\delta_{0t} < 40\,\mu\mathrm{eV}$. The estimate of $\Delta\delta_{0t}$ is by an order of magnitude smaller than $\Delta\delta_{0X}$ for the exciton. Such small spread of the exchange coupling in the trion can be explained by a small spread of parameters of the sub-ensemble of the QDs containing exactly one excess electron for the given value of U_{bias}. It is clear that quantum dot may contain a single excess electron provided that the lowest electronic level exactly coincides with the Fermi level of the substrate. Any deviation of the size or shape of the dot gives rise to a shift of the electronic level and changes the number of resident electrons in the QD. This suggestion is supported by the small range of the bias voltages $\Delta U_{\mathrm{bias}} = 0.2\,\mathrm{V}$, within which the trion beats are observed [3.5].

3. Electron spin relaxation

Here we discuss the experimental results showing that the lifetime of the spin orientation in QDs can be very long, $T_1 > 100\,\mu\mathrm{s}$, at temperatures up to 10 K and small magnetic fields [3.15]. The PL pump-probe method was used to study the electron spin memory in self-assembled InP QDs. This method involves detection of the effect of a circularly polarized pump on polarization of the PL excited by a probe beam. The PL pump-probe method makes it possible to overcome the limitation of the experimental study of spin dynamics related to finiteness of the electron-hole pair lifetime. At the same time, this method allows one to retain the high sensitivity and spectral selectivity characteristic of the conventional PL techniques. Most measurements were made in a longitudinal magnetic field $B = 0.1\,\mathrm{T}$ to suppress the effect of fluctuations of the nuclear field on the electron

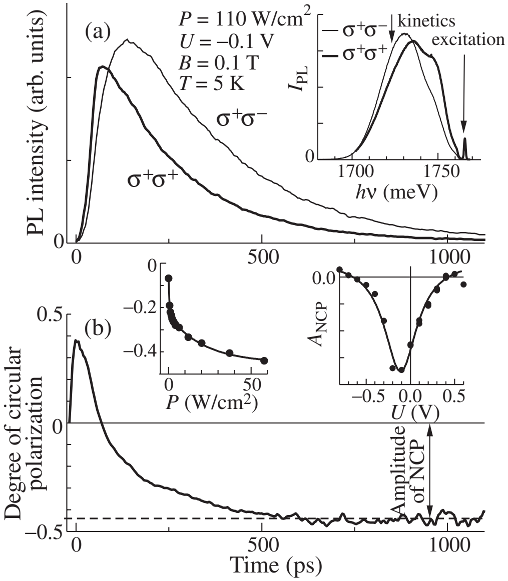

Figure 4. (a) PL kinetics under strong σ^+-excitation in co- $(\sigma^+\sigma^+)$ and cross- $(\sigma^+\sigma^-)$ polarizations. Inset shows PL spectra in the co- and cross-polarizations under excitation at $E_{\text{exc}} = 1771\,\text{meV}$. (b) Kinetics of the degree of circular polarization. Left inset shows the dependence of the NCP amplitude on the pump power density. Right inset shows the dependence of the NCP amplitude on the bias voltage.

spin orientation [4.3].

As an indication of the resident electron spin orientation, it is used the effect of negative circular polarization of PL in the charged InP QDs observed in the PL spectrum for the Stokes shift exceeding 30 meV [see inset in Fig. 4(a)]. The negative circular polarization (NCP) of the PL is revealed even stronger in the PL kinetics measured with a Stokes shift of 45 meV [Fig. 4(a)]. The degree of polarization calculated in a standard way, $\rho = (I_+ - I_-)/(I_+ + I_-)$, where I_+ (I_-) is the intensity of the co- (cross-) polarized PL, changes its sign in approximately 100 ps after the excitation and then comes to a negative steady-state value further referred to as the NCP amplitude (A_{NCP}) [Fig. 4(b)].

The experiments have shown that the NCP amplitude strongly depends on the bias voltage applied to the sample, U_{bias} [right inset in Fig. 4(b)]. The greatest NCP is reached in a narrow range of the bias voltages near $U_{\text{bias}} \approx -0.1\,\text{V}$ practically coincident with the range where, according to Ref. [3.5], the trionic quantum beats are observed. Based on these data, we can conclude that it is the trionic states

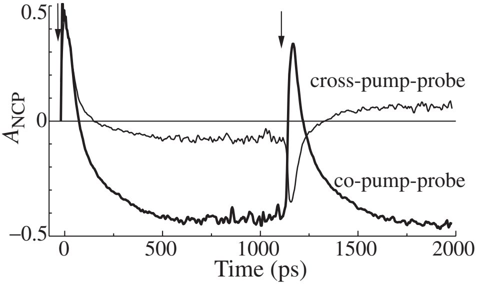

Figure 5. Kinetics of the degree of circular polarization for the co- and cross-polarized pump and probe pulses. Time positions of the pulses are indicated by arrows.

that are responsible for the arising NCP.

In order to study the electron spin lifetime, kinetics of the PL excited by laser pulses separated in time was measured. For this purpose, the laser beam was split into two beams and one of them was optically delayed by 1 ns so that the PL excited by each of the pulses could not overlap in time with that excited by adjacent pulses. Polarization of each beam could be varied independently. The results of the experiments for the case of equal intensities of the probe and pump beams are shown in Fig. 5. When the QDs are excited by circularly co-polarized beams, a large NCP of both PL pulses is observed. However, in the case of excitation by the cross-polarized beams, the PL polarization virtually vanishes. It follows from these results that the spin orientation created by the pump pulse affects the polarization of the PL excited by the probe one and vice versa even when the exciting pulses are separated in time by an interval substantially exceeding the PL decay time ($\tau_{\mathrm{PL}} = 250\,\mathrm{ps}$). It is evident that, after recombination of the electron-hole pairs, the information about polarization of the excitation can be stored only in the orientation of the resident electron spin.

It follows from the data presented in Fig. 5 that the relaxation time of the electron spins substantially exceeds the laser pulse repetition period, 12 ns. Indeed, the NCP amplitude at co-polarized excitation is approximately the same both after the probe and after the pump pulses, in spite of the fact that the former arrives 11 ns after the latter, while the latter comes 1 ns after the former.

To check the conservation of the spin orientation at much longer times, the time delay between the pump and probe excitation pulses has been radically increased by means of selection of the laser pulses using acousto-optical modulators (AOM) placed into the pump and probe paths. The repetition period of the pump and probe pulse trains, their duration and the delay were controlled by a functional generator connected with the AOMs. To obtain a noticeable spin polarization cre-

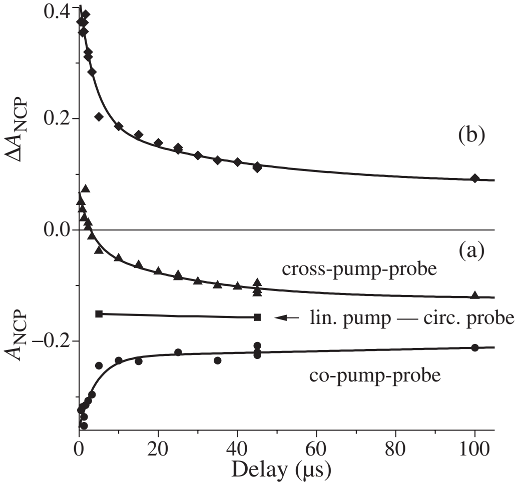

Figure 6. (a) Delay dependences of A_{NCP} for circularly polarized probe with linearly-polarized pump (squares) and with co- (circles) and cross- (triangles) circularly polarized pump. $T = 2\,\mathrm{K}$. (b) The time delay dependence of $\Delta A_{\mathrm{NCP}} = A_{\mathrm{NCP}}(\mathrm{cross}) - A_{\mathrm{NCP}}(\mathrm{co})$ (diamonds) and multi-exponential fit (solid line).

ated by the pump beam, the pump power density was chosen to be several W/cm^2. The intensity of the probe beam was by a factor of 10 . . . 20 lower, because only under these conditions the probe pulse train did not erase the spin polarization created by the pump.

The NCP amplitude of the PL excited by the probe beam was measured in dependence on time delay between the pump and probe pulse trains [see Fig. 6(a)]. Under linear polarization of the pump beam, the NCP amplitude slightly exceeded 10% and almost did not depend on the time delay. The circularly polarized pump substantially affected the value of the NCP. These changes were of opposite signs for the co- and cross-polarized pumping and were approximately equal in magnitude. Therefore, we attributed the NCP changes to the optical spin orientation rather than to other effects like, e.g., heating of the structure by the high-power pump. The difference between the NCP amplitudes at the co- and cross-polarized pumping can serve as quantitative characteristics of spin orientation of the resident electrons.

The results of the measurements have shown that, as the time delay between the pump and probe trains increases, this difference decreases, i.e., the orientation created by the pump decays in time. The dependence of the NCP on the time delay cannot be described by a simple exponential function [see Fig. 6(b)]. For the delays of about units of microseconds, one can observe a relatively fast decrease of the orientation and, then, for the time delays exceeding 50 μs, the orientation comes to a virtually constant level. This result shows that the average spin of the resident electron in the ensemble of the QDs does not completely relax at least during 100 μs. The reason for the non-exponential decay of the spin polarization is most likely related to a spread of the spin relaxation rates in the inhomogeneous ensemble of the QDs, resulting from random distribution of uncontrollable paramagnetic defects. Coupling with these defects speeds up the loss of the spin orientation.

A fundamental question is associated with the reason for so long-lived spin polarization of the resident electrons. Along with suppression of the spin relaxation in QDs, predicted theoretically [3.1], the persistence of the spin polarization can be ascribed, in principle, to the arising dynamic nuclear polarization [1.1, 4.2] capable of polarizing the electron spin. The nuclear polarization created by the light gives rise to the appearance of an effective magnetic field, which splits the electron spin states. If the splitting energy is larger that the thermal energy and the electron spin relaxation is fast, the electrons will be frozen out to the lowest Zeeman level (will be polarized), and this polarization will hold as far as the nuclear field exists. Polarization of electron spins due to the freezing is well known phenomenon in paramagnetic systems. Assumption that the electron polarization can be maintained in our structure by the dynamic nuclear polarization was checked by two kinds of experiments [3.15]. Details of the experiments will be present at the lesson.

4. Conclusion

The examples presented above demonstrate high efficiency of the direct kinetics measurements for study of spin dynamics in QDs. In the lesson, various experimental data will be present together with discussion of the conclusions which can be done from these data.

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