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# Spin separation in self-organized quantum dots under optical orientation of electrons

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## Introduction

Optical methods, widely used in studying the arrays of self-organized semiconductor quantum dots (QDs), are even more effective when they are supplied with measuring polarization, which provides an access to additional degrees of freedom related to carrier spins [1]. In our previous work [2], we have shown that time-resolved picosecond techniques allow to observe transients of polarization of electrons in QDs under optical orientation conditions. The present work is aimed at studying the dynamics of population of discrete energy levels by spin-polarized electrons in QDs. In particular, we present a detailed study of "spin separation", i.e. redistribution of the average spin of electrons among their energy levels as a result of energy relaxation limited by Pauli's principle. Saturation of the QD ground state at high excitation intensities results in the rapid vanishing of the mean spin of electrons in this state. At the same time, the electron spin polarization at higher energy levels dramatically increases, reaching values close to 100%, which is normally forbidden by optical selection rules. We present time dependences of the circular polarization of luminescence, which reflects the electron spin state, measured over a wide spectral range under various excitation densities. A simple theoretical model is discussed that qualitatively describes the experimental results.

## Experimental

The results presented below are obtained on MBE-grown $In_{0.5}Ga_{0.5}As$ QDs embedded in a GaAs matrix. An active region, which consisted of 6 planes of QDs separated by 50 Å GaAs spacers, was inserted into the middle of a 0.2 $\mu$m undoped GaAs layer confined by AlAs(2 nm)/GaAs(2 nm) superlattices. Each QD plane was formed by deposition of 4 monolayers of $In_{0.5}Ga_{0.5}As$ [3]. Circularly polarized beam of a tunable Ti-sapphire laser producing 1.2-ps long light pulses with a repetition rate of 82 MHz was used to excite the investigated structure. The time resolution of the experimental setup, based on the up-conversion technique, was limited by laser pulse duration. The luminescence was registered along the growth axis in back-scattering geometry. The degree of circular polarization of luminescence, $\rho$, was measured. As follows from measurements of spin beats in a strong magnetic field [2], the polarization of holes in our experiments can be neglected, and the polarization of luminescence is determined by polarization of electrons, $P_e$, only: $\rho = P_e$.

Normalized luminescence spectra obtained under excitation of carriers in the GaAs matrix are shown in Fig. 1. Spectrum 1, recorded under continuous wave (cw) excitation with a low pump density $W = 0.1$ W/cm$^2$, results from the emission of ground-state electrons and holes [2]. We associate the presence of two strongly overlapping lines in

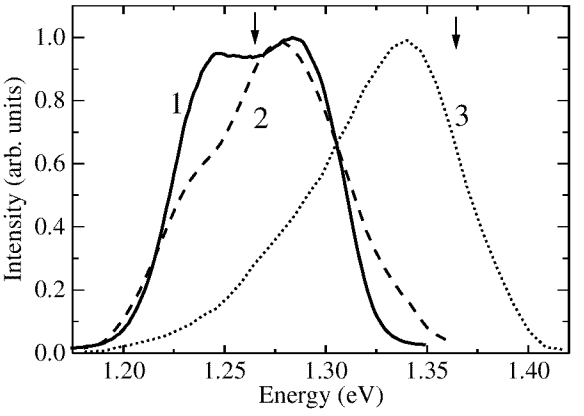


**Fig. 1.** Photoluminescence spectra of $In_{0.5}Ga_{0.5}As/GaAs$ QDs at excitation energy $E_{exc} = 1.531$ eV, $T = 10$ K. 1—cw excitation with power density 0.1 W/cm$^2$; 2 and 3—pulse pumping with power densities 0.1 and 3.0 MW/cm$^2$, time delay after excitation pulse equals 200 ps.

this spectrum with radiative recombination of two groups of dots having different mean sizes. Spectra 2 and 3 are registered under pulse excitation with $W = 0.1$ MW/cm$^2$ and $W = 3.0$ MW/cm$^2$, respectively. A significant blue shift of the luminescence line (up to 60 meV), arising with increase in pulse intensity, is due to the filling of the ground states of electrons and holes and to the appearance of an intense light emission from the excited states. Fig. 2 shows the time dependence $\rho(t)$ measured at two different energies of detection $E_{det}$ (marked by arrows in Fig. 1). It can be seen from Fig. 2 that at the high-energy edge of the luminescence line the polarization first increases up to the maximum value near 70% (curve 1), and then slowly decreases. On the contrary, at the low-energy edge, $\rho$ drops down to values which are close to zero at high excitation density (curve 2). Characteristic times of the fast increase and decrease coincide with the saturation time of the ground state. The latter, determined from the increase of the luminescence intensity (curve 3), appears to be 25 to 40 ps. Further slower decrease of polarization in curves 1 and $2'$ is governed by the spin relaxation time of electrons that is of the order of 300 to 400 ps.

## Model and discussion

The qualitative explanation of the different behavior of polarization at the high-and low-energy sides of the QDs spectra is as follows. The electrons, generated in the GaAs barrier by circularly polarized light, have a spin polarization equal to 50% [3], and preserve it when trapped by QDs. By virtue of Pauli's principle each energy level in the QD can contain no more than two electrons with opposite spins. As at $P_e = 50\%$ there are three times more electrons with spin $-1/2$ than with spin $+1/2$, so energy relaxation will lead to a predominant population of higher energy levels by electrons with spin $-1/2$. This results in increased polarization of luminescence which may exceed 50% (see curve 1 in Fig. 2). On the contrary, with increase in the concentration of photoexcited carriers, the radiation from the ground state becomes non-polarized, since two electrons occupying this state have opposite spins. This is also observed experimentally (see curve 2 in Fig. 2). If the time of inter-level relaxation of electrons is finite, then under pulsed excitation the initial polarization of luminescence is governed by electrons coming from barriers: $\rho(t = 0) = 50\%$. This determines the initial increase of $\rho$ at high $E_{det}$ and decrease of $\rho$ at low $E_{det}$, corresponding to recombination from the ground state. The effect is demonstrated

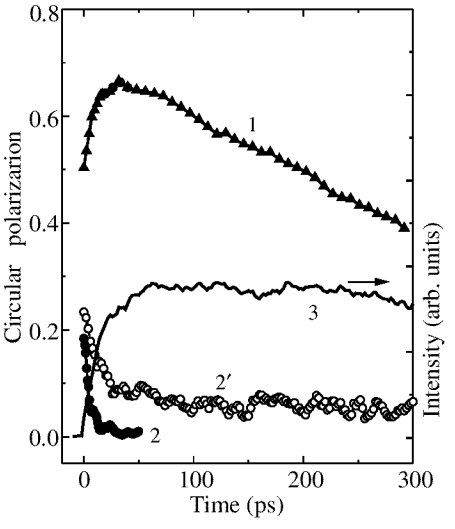


**Fig. 2.** Luminescence circular polarization dependence on time delay at two detection energies $E_{\rm det}$. $E_{\rm exc} = 1.531$ eV, $T = 10$ K. Solid lines are drown to guide the eye. $E_{\rm det}$ (eV): 1—1.363; 2, 2′, 3—1.265. $W$ (MW/cm$^2$): 1, 2—1.5; 2′, 3—0.8.

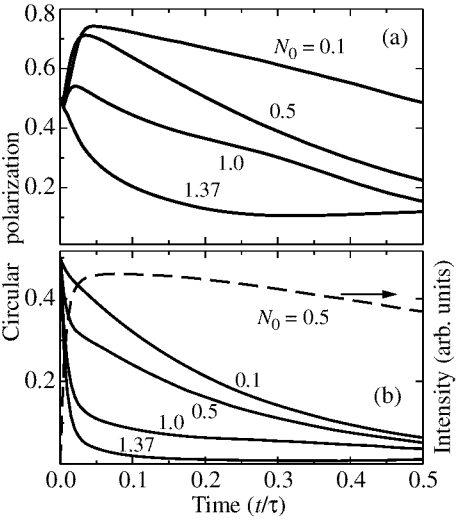


**Fig. 3.** Calculated dependence $\rho(t)$ at different excitation densities for: (a) excited state, (b) ground state. $\tau$ is the electron lifetime. $N_0$ are given with respect to the concentration of QDs. Dashed line is the luminescence intensity at $N_0 = 0.5$.

by experimental curves 1 and 2 in Fig. 2 and by theoretical curves in Fig. 3.

The main question arising when one attempts to describe the transient population of an ensemble of QDs, is to what extent the dots can be considered as isolated from each other. The answer determines the type of statistics applicable. In our case, QDs were vertically coupled, forming linear clusters, or short wires, 6 dots in each. In principle, electrons could move from one dot to another within the cluster, but probabilities of such transitions are not known. For this reason, we have considered two extreme cases, namely, isolated and strongly coupled QDs.

To describe an ensemble of dots isolated from each other, we have used the master equation approach suggested by Bimberg *et al.* [5]. In this approach, probabilities of occupation of all possible states of a dot are calculated as functions of time. As distinct from [5], we additionally distinguish these states by spin. Considering for simplicity dots with 2 electron levels, we get 16 possible electron configurations. In addition, there are two functions describing concentrations of spin-up and spin-down electrons in the reservoir (=barrier): $N_1$ and $N_2$. Dot states are coupled by single-electron transitions: 1) capture from the reservoir to the first or the second level, 2) recombination, 3) spin relaxation within a level, 4) inter-level energy relaxation (transition from the second level to the first without changing spin). In total, there are 18 differential equations describing time evolution of the spin system. This set of coupled equations has been solved numerically under initial conditions corresponding to empty dots and the reservoir filled according to optical selection rules: $N_1(t = 0) = 3N_0$, $N_2(t = 0) = N_0$. Polarizations and intensities of luminescence from the first and second levels calculated at different values of $N_0$, assuming realistic values of relaxation times, are presented in Fig. 3.

For the case of strongly coupled dots, the electron levels belonging to different dots

form narrow bands. This situation would be close to the one considered by Dyakonov and Perel' [6] for bulk semiconductors, but for delta-shaped density of states, typical of quantum-dot systems. In order to describe time dependent electron polarization in this case ("wire" model) , we have used rate equations for concentrations of electrons with both spin directions on the first and second levels. Numerical results of this model are similar to those shown in Fig. 3. The most pronounced difference between the models occurs at low exciting intensities. In the "dot" model the polarization of the second level decreases with increase in intensity, being limited by 80%. In the "wire" model the polarization of the excited level increases with increase of pumping, coming to 100%, and then decreases with further increase in carrier concentration due to the saturation of the second level.

Both models give qualitative agreement with the experiment. To conclude on validity of a specific model, additional experiments with isolated QDs are required.

It should be noted that the spin-dependent population of excited electron levels must lead to the dependence of their radiation intensity on the mean spin of carriers and, therefore, on the polarization of the exciting light. Within both our models, this effect does show up if the polarization of the exciting light is changed from the circular to the linear one.

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