Negative circular polarization dynamics in InP/InGaP quantum dots

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Abstract. Photoluminescence (PL) negative circular polarization (NCP) dynamics of InP/InGaP quantum dots (QDs) was studied. Time resolved measurements of PL demonstrated that NCP vanishes, when transverse magnetic field is applied, while oscillations of polarization (that are typical for both low-dimensional and bulk materials) do not occur. Hole g-factor spread in the QD ensemble was supposed to be the most probable reason for such NCP magnetic field behavior. The dependence of NCP dynamics on the repetition period of excitation laser pulses was investigated. In case of fairly small repetition period (T = 13.3 ns) long living NCP (13.3 ns < t < 133 ns) was detected, what was ascribed to resident electron spin orientation, accumulated during many laser pulses. In that regime more than one luminescence polarization decay time exist.

1. Introduction
Quantum dots (QDs) are promising objects for quantum information processing devices, owing to long spin memory [1]. Operation of such devices will be based on spin orientation utilizing. The point is that one of the most important spin relaxation mechanisms of bulk materials, Dyakonov-Perel mechanism, is suppressed in QDs [2].

In X-trion (two electrons and hole) luminescence unusual phenomena of circular polarization negative degree (NCP) takes place [3,4,5,6]. This fact means that the sign of luminescence circular polarization is opposite to the sign of excitation light circular polarization. In current research we have studied dynamics of NCP with and without external magnetic field (applied in Voigt geometry) to clarify NCP formation details in InP quantum dots.

2. Sample
The sample contains one layer of self-assembled InP quantum dots in In\textsubscript{0.48}Ga\textsubscript{0.52}P matrix, grown by metalorganic chemical vapour deposition (MOCVD). The distinctive feature of the sample is presence of two characteristic dot sizes, see figure 1. QDs are lens-shaped and mean lateral size of small dots is 100 nm, of big dots - 130 nm.

![Atomic force microscopy images](a and b, size 2.5x2.5 μm²) of the similar uncapped sample [7]. The In\textsubscript{0.48}Ga\textsubscript{0.52}P cap thickness in the studied sample equals 40 nm.

\textbf{Figure 1.} Atomic force microscopy images of the uncapped sample [7].
3. Experimental setup
Both CW and time-resolved studies of optical orientation were conducted. The sample was placed in a helium cryostat (T = 6 K or 10 K), luminescence excitation was provided using circular polarized emission of a Ti:Al₂O₃ laser with 700 nm wavelength. In case of time-resolved measurements a pulse picosecond laser was used. Luminescence was collected in backscattering geometry and passed sequentially through a polarization analyzer, a monochromator and a streak camera (in case of time-resolved measurements) or an avalanche diode (for time averaged studies). After that the signal was processed by a pulse counting card and a PC. In some experiments magnetic field in Voight geometry was applied. For certain time averaged studies modulation of the excitation light circular polarization sign at 50 kHz frequency was conducted, using a photoelastic modulator, in order to prevent orientation of nuclear spins [8].

4. Results
Spectra of intensity and polarization are presented on figure 2. Two intensity peaks correspond to two characteristic dot sizes. We perform studies of long-wavelength line (big QDs), where photoluminescence (PL) circular polarization degree is negative. NCP presence in big QDs luminescence is referred to resident electrons accumulation in those dots (InGaP layers have residual n-doping), where the ground energy state is lower, in comparison to small QDs. It should be recalled that NCP is specific feature of X-trion emission.

![Figure 2. Intensity and circular polarization (optical orientation) PL spectra. Temperature 10 K, CW excitation, 697 nm excitation wavelength.](image)

To clarify NCP formation details the PL dynamics with and without external magnetic field (applied in Voigt geometry) was studied. Time resolved measurements of the PL demonstrated that NCP vanishes with increasing of transverse magnetic field (figure 3 a,b), while oscillations of polarization (that are typical for both low-dimensional and bulk materials and are the consequence of the Larmor precession of electron spins) do not occur.

Three possible reasons for the polarization vanishing without oscillations have been proposed and examined:
(i) as far as NCP corresponds to X-trion emission, the orientation of resident electron spins is important for a trion formation process. If this spin orientation is accumulated with many laser pulses, the Larmor precession of resident electron spins, aligned by different pulses, starts at different times, and the coherence is lost;
(ii) the spread of nuclear magnetic fields acting on electron spins, localized in a QD, will result in the spread of Larmor precession frequencies of electron spins (inhomogeneous broadening), what may cause oscillations vanishing.
(iii) since the polarization of the X-trion emission is determined by hole spin orientation as well, the hole g-factor spread in the QD ensemble may result in vanishing of polarization degree in transverse magnetic field instead of oscillations.

Figure 3. Circular polarization time dependences at different magnetic fields in Voight geometry. Temperature 6 K, 700 nm excitation wavelength, 766±/17 nm detection wavelength, laser pulse repetition periods equal 13.3 ns (a) and 133 ns (b).

Scenario (i) - NCP accumulation with many laser pulses - was studied experimentally. Figure 4 shows time averaged NCP degree dependence on laser pulses repetition rate. The circular polarization degree doesn’t depend on frequency of laser pumping, at periods from 133 ns and higher, while for 13.3 ns period the value of NCP is substantially bigger. We propose that in 13.3 ns laser pulse repetition period accumulation of resident electron spin orientation with many laser pulses takes place, thus mean spin orientation of resident electrons and consequently the degree of NCP increase. Spin relaxation time of resident electrons ($\tau_s$) can be estimated as follows: $13.3 \text{ ns} < \tau_s < 133 \text{ ns}$. It was concluded that scenario (i) doesn’t cause the effect, because oscillations are absent both in the NCP accumulation with many laser pulses regime (figure 3 a) and the separate laser pulses regime (figure 3 b).

Figure 4. The time integrated circular polarization dependence on the period of excitation laser pulses. Temperature 6 K, 700 nm excitation wavelength, 766 nm detection wavelength
Our analysis has shown also that assumption (ii) – the nuclear fields spread in the QD ensemble - is unlikely, because in that case oscillations are expected to appear in big magnetic fields (which are much bigger than nuclear fields), what does not happen. First of all, presence of nuclear magnetic fields, acting on electron spins, will be demonstrated. Figure 5 shows (CW) NCP dependence on magnetic field in Faraday (5 a) and Voigt (5 b) configurations. The Hanle curve (5 b) in case of excitation with the constant polarization (black dots) is two times wider than in case of modulated polarization (red dots). The polarization modulation frequency equals 50 kHz and prevents nuclear spin orientation. NCP field dependence in Faraday geometry is moved relatively y axis by a value of 300 G. One can conclude that nuclear fields, acting on electrons, persist, and nuclear quadrupole interaction manifests itself [9]. Indeed, in the standard theory of optical orientation Knight field enabled nuclear fields increase the spin depolarization rate in transverse magnetic field, what leads to narrowing of the Hanle curve. That contradicts the obtained results. Presence of the nuclear electric strain-induced quadrupole interaction causes fixing of nuclear field in the structure growth direction, what explains the Hanle curve broadening (external magnetic field has to overcome nuclear field) and the longitudinal field shift. The value of nuclear magnetic fields of approximately 300 G can be estimated from NCP field dependences. A simulation has shown that oscillation vanishing takes place, only when the typical value of nuclear magnetic field fluctuations equals to the value of total magnetic field (which is the sum of nuclear and external fields). Thus in external magnetic fields much bigger than nuclear fields oscillations are expected to be recovered. But they are not recovered even in 5000 G external magnetic fields, what can be seen in figure 3 a.

The heavy hole g-factor scattering (scenario (iii)) is supposed to be the main candidate for explanation of the oscillations lack and requires further research. It will be shown why considerable g-factor spread in the QD ensemble may be realized. Heavy hole g-tensor is asymmetric, and in case of quantum dots with high enough symmetry (axial or D<sub>2d</sub>) g-tensor components in the structure plane equal zero to a first approximation (actually they are nonzero, but very small [10],[11]). In-plane anisotropy of QDs will result in admixture of light and heavy hole states [12]. Thus g-factor in the structure plane appears. So in case of considerable in-plane anisotropy relatively high g-factor value can be achieved. Because of QDs forms scattering in the ensemble, transverse g-factor of heavy hole will scatter as well.
In order to figure out influence of NCP accumulation with many laser pulses on a negative polarization formation process we studied the NCP dynamics at different laser pulses repetition periods. Figure 6 shows the time dependence of PL circular polarization at two different pulse frequencies. Red curve (13.3 ns repetition period, corresponds to accumulation of negative polarization with many laser pulses) shows complicated dynamics of NCP – two polarization decay times. In case of 133 ns pumping repetition period (black curve), what corresponds to the separate pump pulses regime, long living NCP is absent. We conclude that such pulse frequency dependence is determined by presence of resident electron spin orientation in case of frequent pulses and absence, when pulses are rare. One can notice that the relatively high NCP degree is achieved without resident electron spin orientation, what is unusual. It should be noticed that observation of such long spin relaxation times (tens of nanoseconds) in luminescence requires long carrier lifetime as well. Appearance of nonradiative dark excitons due to hole spin relaxation during carriers thermalization process is proposed. Their radiative recombination can happen, if spin flip of a single carrier in a dark exciton takes place or, after forming a trion with a resident electron. Latter process also may require an electron spin flip for relaxation from triplet to singlet trion spins and further recombination. Characteristic times of these processes can be rather long.

Figure 6. PL circular polarization time dependences at different laser pulse repetition periods. Temperature 6 K, 700 nm excitation wavelength, 766+/−17 nm detection wavelength.

5. Conclusions
To summarize, the photoluminescence negative circular polarization dynamics of InP quantum dots in InGaP matrix was studied. Time resolved measurements of PL demonstrated that NCP vanishes with increasing of external transverse magnetic field, while oscillations of polarization do not occur. Three reasons of the phenomenon were proposed and examined. The hole g-factor spread in the QD ensemble was supposed to be the most probable cause of such NCP magnetic field behavior. However, this scenario requires further studies. The dependence of NCP dynamics on the repetition period of excitation laser pulses was studied. In case of frequent laser excitation pulses (T = 13.3 ns) more than one polarization decay time exist. Presence of long living NCP (13.3 ns < t < 133 ns) in this excitation regime was ascribed to resident electron spin orientation, accumulated during many laser pulses.

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