Time-resolved Hanle effect in (In,Ga)As/GaAs quantum dots

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Abstract. The circular polarization degree of (In,Ga)As/GaAs quantum dot photoluminescence is experimentally studied as function of magnetic field applied perpendicular to the optical excitation direction (Voigt geometry). The measurements are performed using various modulation timing protocols for the excitation polarization and intensity. The experimental data obtained allow us to estimate the characteristic times of polarization and relaxation processes for the nuclear spin system.

Introduction

The hyperfine interaction of electron and nuclear spins in semiconductor quantum dots (QDs) has recently become a matter of extensive theoretical and experimental research [1]. The electron spin dynamics is found to depend sensitively on the state of nuclear system. On the other hand, the oriented electron spin can generate a dynamical nuclear polarization (DNP) through the hyperfine interaction. The steady-state properties of DNP in semiconductors are under study for at least three decades [2], however experimental and theoretical studies of the DNP dynamics in QDs have been developed only during the last several years [3, 5, 6]. In particular, it has been predicted theoretically that the dynamics in QDs should be quite complicated [7, 8]. Experiments reveal a large variety of nuclear spin relaxation times from milliseconds [3, 6] to seconds [4, 5].

Most experimental data about the DNP in QDs have been obtained in magnetic field applied along the axis of optical excitation and signal detection (Faraday geometry), see, e.g., Ref. [1] and references therein. In this case the magnetic field prevents the nuclear spin system from angular momentum leakage due to the dipole-dipole interaction between the nuclear spins. Such experiments provide results, which allow a straightforward analysis of the processes studied.

In magnetic field perpendicular to the optical axis (Voigt geometry), the polarization of the electron spin decreases due to its precession about the magnetic field (Hanle effect). Strong optical pumping accompanied by relaxation processes in the nuclear spin system gives rise to an effective DNP field aligned along the external magnetic field and, correspondingly, normal to the direction of optical orientation. The presence of this DNP field may considerably modify the Hanle curve [9, 10].

In the present work, we report on experimental results of the time-resolved Hanle effect for a (In,Ga)As/GaAs quantum dot ensemble. These results allow us to estimate the characteristic times of processes occurring in the nuclear spin system.
Figure 1. Hanle curves in relatively strong (a) and weak (b) magnetic fields for excitation with constant intensity but modulated circular polarization. The left inset in panel (a) shows the modulation protocol and the right inset shows the change of HWHM of the Hanle curves with rise of modulation period $T_{\text{mod}}$. Hanle curves measured in relatively strong (c) and weak (d) magnetic fields for excitation with constant circular polarization and modulated intensity are shown to the right. The left inset in panel (c) shows the excitation protocol and the right inset shows the change of HWHM with rise of dark time $t_d$.

1. Experimental details
A heterostructure containing 20 layers of self-assembled (In,Ga)As QDs sandwiched between n-delta modulation doped GaAs barriers has been studied. Donor ionization supplies every QD with on average a single resident electron.

Photoluminescence (PL) was excited by a continuous-wave Ti:Sapphire laser. Special timing protocols for the optical excitation which are schematically shown in Fig. 1 were developed to study the dynamics of nuclear polarization. An acousto-optical modulator was used to form the excitation pulse sequences. Modulation of excitation polarization was realized by an electro-optical modulator. The PL polarization was measured using a photoelastic modulator operated at a frequency of 50 kHz and a multi-channel photon counting system. The experiments were done at temperature $T = 1.8$ K. The electron spin polarization was determined from the degree of circular polarization of the QD PL excited at the energy of the wetting layer optical transition. We analyzed the degree of the negative circular polarization (NCP) effect, which is provided by the spin orientation of the resident electrons in singly-charged QDs. Further experimental details can be found in Ref. [6].

2. Experimental results
To study the dynamics of the nuclear polarization rise we have measured a set of Hanle curves at modulated polarization of the excitation. The modulation period was varied over a wide range. For large
modulation period ($T_{\text{mod}} > 100 \text{ ms}$), the Hanle curve is found to coincide almost with that for continuous-wave (CW) measurements. It consists of a wide Lorentzian-like part and a w-shaped peculiarity in its central part. Decrease of the modulation period is accompanied by a strong decrease of the half width at half maximum (HWHM) of the Hanle curve. Besides, a remarkable modification of the curves is observed in the range of small magnetic fields. As seen in Fig. 1(b), the width of the central peak increases, the symmetrical dips around the peak are smoothed, and the curve approaches the standard form of a Hanle curve. According to Ref. [9], the w-shape of Hanle curve with narrow central peak is caused by DNP whose effective magnetic field is directed along the external magnetic field. Our results presented in Fig. 1 show that for the excitation conditions used DNP develops in the time range of tens of milliseconds.

To study the relaxation of DNP, we used amplitude modulation of the optical excitation keeping the helicity of polarization as shown in the inset of Fig. 1(c). We used relatively long and intensive pump pulses to excite the electron-nuclear spin system up to a stationary state, which does not depend on the history of excitation, and in particular on the dark interval duration. The dark interval, $t_d$, was varied in the time range from 0.02 ms to 50 ms. The NCP signal and, correspondingly, the spin polarization were detected during the first millisecond just after the dark interval when the nuclear spin polarization was still not restored by the pumping. Results of these experiments are shown in Fig. 1(c,d). The Hanle curve measured after a very short dark interval is similar to that measured in CW regime. In particular, the central peak related to DNP is very pronounced. However already at $t_d > 2 \text{ ms}$, a noticeable deformation of the peak becomes observable. This means that the nuclear spin relaxation time is units of milliseconds. This conclusion agrees well with the results on DNP dynamics measured in longitudinal magnetic field [6].

![Figure 2](image_url)

**Figure 2.** Hanle curves measured in CW regime at different excitation densities. Inset shows the dependence of HWHM of Hanle curve on excitation density.
In Figure 2, Hanle curves measured in CW regime at different pump powers are shown. A pump power increase is accompanied by increase of both the amplitude and the HWHM of the curves. At the maximal excitation power \( P_{\text{exc}} = 100 \text{ W/cm}^2 \) used in the experiment, HWHM exceeds 150 mT and is almost four times larger than the one obtained in the absence of DNP [compare with Fig. 1(a), \( T_{\text{mod}} = 0.04 \text{ ms} \)]. We believe that the HWHM increase is related to the increase of DNP field and its value exceeds 100 mT at the largest power density used. Recently, a very large broadening of the Hanle curves was observed for single QDs and was assigned to the DNP effects [10].

3. Conclusion
The time resolved measurement of the Hanle effect in (In,Ga)As QDs gives two new results. Firstly, the rise and decay times of nuclear spin polarization are estimated for the weak magnetic field range where DNP appearance may be explained by a classical mechanism. Secondly, a sharp rise of the Hanle curve width with excitation power increase has been found. We can conclude that the broadening of the Hanle curve is a result of DNP formation.

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