ABSTRACT

This work present original results of investigation of nuclear spin dynamics in nanostructure with negatively charged InGaAs/GaAs quantum dots characterized by strong quadrupole splitting of nuclear spin sublevels. The main method of the investigation is the experimental measurement and theoretical analysis of the photoluminescence polarization as the function of the transverse magnetic field (effect Hanle).
The dependence of the Hanle curve on temporal protocol of excitation is examined. Experimental data are analyzed using an original approach based on separate consideration of behavior of the longitudinal and transverse components of nuclear polarization. The rise and decay times of each component of nuclear polarization and their dependence on transverse magnetic field strength is determined.
To study the role of the Knight field in the dynamic of nuclear polarization a weak additional magnetic field parallel to the optical axis is used. This allows us to control the efficiency of nuclear spin cooling and the sign of nuclear spin temperature. The standard nuclear spin cooling theory fails to describe the experimental Hanle curves in a certain range of control fields. This controversy is resolved by taking into account the nuclear spin fluctuations. The model allows us to accurately describe the measured Hanle curves and to evaluate the parameters of the electron-nuclear spin system of the studied quantum dots.
New effect of resonant optical pumping of nuclear spin polarization in an ensemble of singly charged (In, Ga)As/GaAs quantum dots subject to a transverse magnetic field is observed. Electron spin orientation by circularly polarized light with the polarization modulated at the nuclear spin transition frequency is found to create a significant nuclear spin polarization, precessing about the magnetic field. Nuclear spin resonances for all isotopes in the quantum dots are found in that way. In particular, transitions between states splitted off from the $\langle \pm 1/2|$ doublets by the nuclear quadrupole interaction are identified.

Keywords: quantum dots, nuclear spin, electron spin, polarization, Hanle effect, quadrupole splitting.

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1 INTRODUCTION

1.1 Intellectual merit

The spin dynamics in semiconductor quantum dots (QDs) has been an object of intense theoretical and experimental research during the past few decades (Dyakonov, 2008, Merkulov, 2002, Khaetskii, 2002, Gammon, 2001, Braun, 2006, Tartakovskii, 2007). In a QD, the spin of a confined electron is strongly coupled to the spins of the lattice nuclei. The coupling strength is given by the contact hyperfine interaction, which is enhanced due to strong localization of the electron in the QD (Merkulov, 2002, Khaetskii, 2002). The hyperfine coupling destroys electron spin polarization via interaction with random fluctuations of the effective nuclear magnetic field (Merkulov, 2002). A way to overcome this effect is to create a strong polarization of the nuclear spins (Khaetskii, 2002).

The dominant mechanism of the dynamic nuclear polarization (DNP) in semiconductors is the angular momentum transfer from optically oriented electrons to nuclei via electron–nuclear hyperfine interaction (Meier, Zakharchenya, 1984). This process is particularly effective in quantum dot heterostructures, where the electron wave function covers a limited number of nuclei, and the electron and nuclear spins make up a strongly coupled system. Since the spin polarized nuclei, in turn, generate effective magnetic field (Overhauser field) which splits electron spin sublevels, the state of the nuclear spin system can be examined by polarization and spectroscopic methods. The methods based on studying the polarization of photoluminescence (PL) were extensively explored for study of nuclear spin dynamics in bulk semiconductors (Meier, Zakharchenya, 1984, Kalevich, 2008). Nuclear spin relaxation times were found to be a few seconds or longer (Kalevich, 1982). Spectroscopic methods were widely used last fifteen years when the experimental technique was developed for study of PL spectra of single quantum (Brown, 1996, Gammon, 1996, Tartakovskii, 2007, Belhadj, 2008). These experiments allowed one to detect strong effect of nuclear magnetic field and to perform the first experiments on detection of nuclear magnetic resonance of single QD (Gammon, 1997). These measurements reported in recent years, have also shown that nuclear spin relaxation in QDs is much faster. In particular, nuclear spin relaxation in a magnetic field applied parallel to the optical axis (longitudinal field) was found to occur over times on the order of milliseconds (Maletinsky, 2007, Chekhovich, 2010, Belhadj, 2008).

An alternative approach for detection of nuclear polarization in QDs is to measure the electron polarization created by optical pumping in an external magnetic field. This method does not require high spectral resolution and can be used to QDs ensemble characterized by extremely wide lines in the PL spectrum. As the nonequilibrium electron spin polarization is, in many cases, the magnetic-field dependent, the Overhauser field can be detected using its effect on the mean electron spin, for example, by observing the associated changes in the circular polarization of PL. In a magnetic field parallel to the optical axis (longitudinal magnetic field), the nuclear polarization created by the pumping may influence the PL polarization by suppressing electron spin relaxation (Cherbunin, 2009, Maletinsky, 2007, Maletinsky, 2007a). For optical pumping in a magnetic field perpendicular to the optical axis (transverse magnetic field), the electron spin polarization is usually destroyed with increasing magnetic field (Hanle effect). In this case, the Overhauser field modifies the width and shape of the dependence of the circular polarization of the PL on the magnetic field (Hanle curve), which can give rise to a nonmonotonous dependance, and even to a hysteresis (Meier, Zakharchenya, 1984, Cherbunin, 2009, Kalevich, 2008, Paget, 1977, Pal, 2009, Auer, 2009, Flisinskii, 2010, PIV, Masumoto, 2008, Urbaszek, 2013). In the presence of DNP, there is a specific feature in the Hanle curve near zero magnetic field (W-structure, **Fig. 1**) analyzed for the first time in Ref. (Paget, 1977). Qualitative interpretation of W-structure is typically done in the

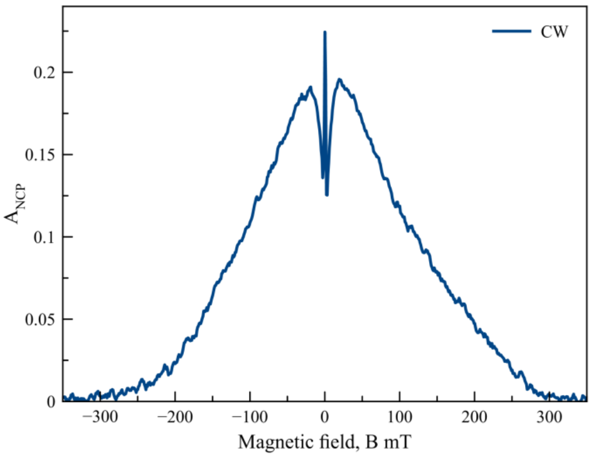

Fig. 1. Typical dependence of the degree of PL polarization in transverse magnetic field for QDs studied in present work (Hanle curve under continuous-wave pumping).

framework of the nuclear spin cooling model (A. Abraham, 1961, Meier, Zakharchenya, 1984). At the same time, this model based on thermodynamics approach does not always allow one to quantitatively analyze the dynamics of nuclear spin polarization and relaxation in QDs.

1.2 Goal of the work

The goal of present work is the investigation of main characteristics of optically created dynamical nuclear polarization in semiconductor QDs. We have performed the experimental study and theoretical modeling of the dynamics of electron-nuclear spin system in a set of nanostructures with InGaAs QDs. To quantitatively analyze the experimental data we have generalized the cooling spin model. The following tasks were solved:

- Investigation of the dynamics of the rise and decay of the spin polarization of nuclei in QDs under optical excitation (PII).
- Experimental study and theoretical modeling of the dependence of electron spin polarization in a transverse magnetic field in the QDs with strong electron-nuclear spin interaction (PIII).
- Investigation of nuclear magnetic resonance (NMR) by resonant optical pumping of nuclear polarization in the QDs under study (PIV).

1.3 Methods of investigation

In present work, the main experimental method was the investigation of the PL polarization of QDs excited by polarized optical pumping. For theoretical analysis, we used two phenomenological models: the geometric model (PII) and a generalization of the nuclear spin cooling model (PIII), allowing for adequate description the whole set of experimental data. To understand the nuclear spin dynamics, it is important to get understanding of the dynamics of electron spin. For this purpose at the first step we have studied the dynamics of polarization and relaxation of the electron spins in QDs induced by non-uniform electric field in a quantum well by the method of photo-induced magneto-optical Kerr effect (PI).

1.4 The main results

1. Behavior of the Hanle curves is studied experimentally using different temporal protocols of excitation and registration of photoluminescence of the InGaAs quantum dots.
2. The Hanle curves are analyzed by the use of an original approach based on separate consideration of behavior of the longitudinal and transverse components of nuclear polarization in QDs with strong quadruple splitting of nuclear spin states (PII).
3. This analysis made it possible to determine the rise and decay times of each component of nuclear polarization and their dependence on transverse magnetic field strength for QDs under study (PII).
4. Deformation of the Hanle curve as the function of small additional magnetic field parallel to the optical axis is studied experimentally (PIII).

5. To analyze this deformation a phenomenological model of the formation of the Hanle contour in such experimental conditions is developed. The model is based on the standard theory of nuclear spin cooling and takes into account the effect of nuclear spin fluctuations (PIII).
6. This generalized model allows us to adequately describe the experimental data and to evaluate the maximal value of the effective field of nuclear polarization created by optical pumping in QDs under study (PIII).
7. The analysis of experimental data using this model allowed us to determine the magnitude of effective field acting on the nuclei from the electron spin (Knight field) in the sample annealed at 980 ^{o}C to be of about 1 mT when the electron spin is almost totally polarized (PIII).
8. The effect of resonant optical pumping of the precessing transverse component of nuclear spin polarization in inhomogeneously broadened QD ensemble is observed for the first time under strong excitation of QDs by the circularly polarized light, which polarization is modulated with the nuclear spin precession frequency about external magnetic field (PIV).
9. Nuclear spin resonances for all isotopes in the quantum dots are identified using the calculated values of quadrupole nuclear spin splitting caused by the strain-induced gradient of crystal electric field at nucleus positions (PIV).

1.5 Practicability

The results can be applied to the study of coherent optical properties of nanostructures with quantum dots (In,Ga)As/GaAs, carrying out at the Technical University of Dortmund (Germany), Physico-Technical Institute (St. Petersburg), Novosibirsk State University and other research centers.

1.6 Appraisal of the work and publications

The results of this work were reported at the international conferences 16th International Symposium “Nanostructures: Physics and Technology” (Ekaterinburg, Russia — 2011, Saint-Petersburg, Russia — 2013), “International Conference of Spin-Optronics” (Saint-Petersburg, Russia — 2012, Toulouse, France — 2013), “International Conference on Optics of Excitons in Confined Systems” (Rome, Italy — 2013), “The 7th International Conference on Physics and Applications of Spin-related Phenomena in Semiconductors” (Eindhoven, the Netherlands — 2012), “7th International Conference on Quantum Dots” (Santa Fe, New Mexico, USA — 2012), “31st International Conference on the Physics of Semiconductors” (Zurich, Switzerland -2012), “NewMaRE: New Materials and Renewable Energy” (Tbilisi, Georgia — 2012), “School of Nanophotonics and Photovoltaics” (Maratea, Italy — 2013) and at the seminars of the Spin Optics Laboratory and the department of Solid State Physics (Saint Petersburg State University 2008 — 2013).

2 THE MAIN CONTENT

2.1 Samples and experimental setup

Heterostructures containing 20 layers of self-assembled (In,Ga)As/GaAs QDs separated by Si-δ-doped GaAs barriers were studied in this work (**Fig. 2**). Donor ionization supplies every dot with, on average, a single resident electron. The original structure was grown by molecular-beam epitaxy on a (100) GaAs substrate. Then it was separated into several pieces which were then thermally annealed at different temperatures. The annealing resulted in a reduction of the In content in the QDs due to interdiffusion of In and Ga atoms and in a high-energy shift of the lowest QD optical transition. The an-

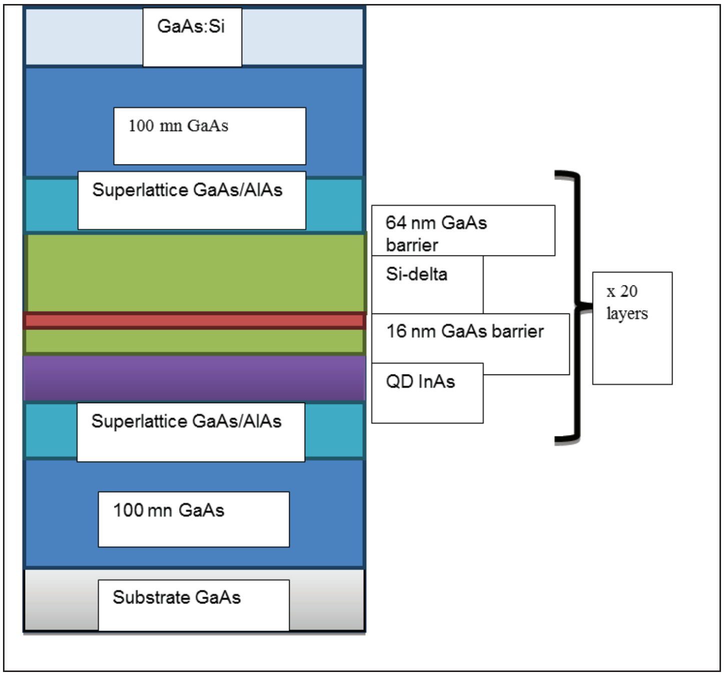

Fig. 2. Structure under study.

nealing also gave rise to the considerable decrease of mechanical stress in the QDs and, therefore, to reducing the quadrupole splitting of nuclear spin states. Besides, the enlarging of localization volume for resident electrons occurred due to the interdiffusion of In and Ga atoms.

The studied sample was immersed in liquid helium at a temperature T = 1.8 K in a cryostat with a superconducting magnet. Magnetic fields up to 100 mT were applied perpendicular to the optical axis (Voigt geometry) along to the [110] crystallographic direction of the sample. To create an additional magnetic field, perpendicular to the main magnetic field and parallel to the optical axis, a pair of small Helmholtz coils was installed outside the cryostat.

The PL of the sample is excited by circularly polarized light from a continuous-wave Ti:sapphire laser, with the photon energy tuned to the optical transitions in the wetting layer of the sample. An electro-optical modulator followed by a quarter-wave plate is used to modulate the polarization helicity of optical excitation. The degree of circular polarization of the PL is typically detected by a standard method using a photoelastic modulator and an analyzer (a Glan-Taylor prism). The modulator creates a time-dependent phase difference $\phi = (\pi/4)\sin(2\pi f \cdot t)$, between the linear components of the PL, thus converting each of the circular components (σ+ and σ−) into linear ones (x and y) at the modulation frequency, typically $f = 50$ kHz. The analyzer selects one of the linear components, which was dispersed with a 0.5-m monochromator and detected by an avalanche photodiode. The signal from the photodiode was accumulated for each circular component separately in a two-channel photon-counting system. The PL polarization was calculated using a standard definition, $\rho = (I^{++} - I^{--})/(I^{++} + I^{--})$, where I^{++} and (I^{--}) are the PL intensity for copolarization and cross polarization of excitation and detection, respectively. The PL polarization was recorded at the wavelength corresponding to the maximum of the PL band of the sample. The complete optical setup is shown in **Fig. 3.** In some experiments, the PL polarization is determined from the PL signals detected for a fixed helicity of the PL but at different helicities of excitation.

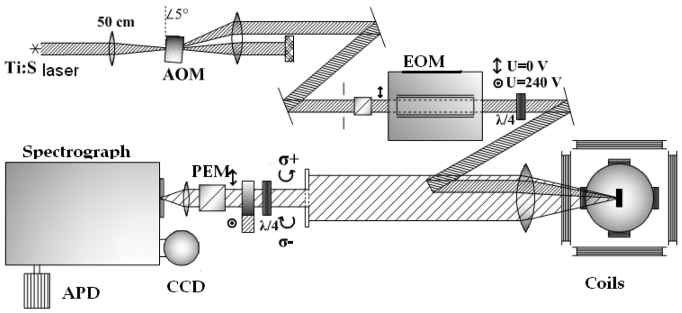

Fig. 3. Experimental setup.

To study the dynamics of nuclear polarization, we used a amplitude modulation of laser beam using an acousto-optic modulator to produce pulses with various bright and dark time durations. The spin polarization of the resident electrons is monitored through the effect of negative circular polarization (*NCP*), $\rho < 0$, of the PL observed for singly charged QDs. The mechanism of NCP of the PL of QDs has been extensively discussed in Refs. (Cortez 2002, Ignatiev2009, Dzhioev, 1998), where it was shown that the presence of *NCP* is the result of optical orientation of the resident electrons. The amplitude of *NCP* is proportional to the projection of electron spin onto the optical axis z, averaged over the QD ensemble, (Flisinski2010)

$$A_{NCP} \sim 2S_z. \quad (1)$$

The amplitude of the PL polarization, A_{NCP}, increases with rising excitation power at relatively low excitation levels (see **Fig. 4**). A further rise of the power results in saturation

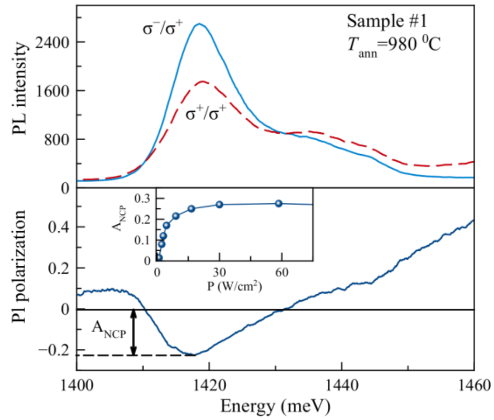

Fig. 4. Typical PL spectra (top curves) measured at σ+-excitation and co- and cross-polarized detection and the degree of circular polarization (bottom curve) for sample annealed at 980°C. The definition of amplitude of the negative circular polarization is illustrated by the arrow marked A_{NCP}. Inset shows the power dependence of A_{NCP}.

of the A_{NCP}, which indicates a high level of electron spin polarization. The pump powers used in our experiments were sufficiently high to totally polarize the electron spin.

2.2. Dynamics of nuclear polarization in a transverse magnetic field

Dynamics of nuclear polarization in quantum dots in the magnetic field perpendicular to the optical axis (in Voigt geometry), until recently, did not actually investigated. An application of the transverse magnetic field reduces the degree of circular polarization of photoluminescence from semiconductors (Hanle effect). This is the effect of electron (or exciton) spins precession about the field. The shape of Hanle curve can be changed by effective magnetic field of nuclear polarization (Paget, 1977, Krebs, 2010). This makes it possible to study the dynamics of nuclear polarization experimentally by measuring the Hanle effect in the kinetic regime, i.e., with time resolution.

The first observations of the time resolved Hanle effect in an ensemble of negatively charged InGaAs/GaAs quantum dots (Cherbunin, 2010) demonstrated that experiments of this kind would provide an effective tool for examining the dynamics of a nuclear spin system. In PII we demonstrate systematic experimental data presented and analyzed to estimate the nuclear polarization buildup and decay times for the structure under study. In our experiments, we used amplitude modulated optical pumping with various dark and excitation times, t_d and t_{exc}.

The Hanle curves obtained under strong pumping for the sample under study are largely similar in shape to those observed previously for donor-bound electrons, in qualitative agreement with predictions of a classical model of DNP in a transverse magnetic field (Meier and Zakharchenya, 1984, Kalevich, 2008). However, the classical model fails to explain an increase in Hanle curve width with optical pumping intensity observed in our experiments. Following (Cherbunin, 2010, Dzhioev, 2007), we suppose that the Hanle curve broadening is due to nuclear polarization stabilized by quadrupole splitting of nuclear spin states. An analysis of time resolved measurement data provides quantitative estimates of the rise and decay times for longitudinal and transverse nuclear fields in the structures under study.

To examine nuclear polarization buildup, we measure *NCP* as a function of time after a pump pulse had arrived using the multichannel photon counting system.

Nuclear spin relaxation was examined by detecting photoluminescence during a short interval $t_{det} = 1$ ms at the start of pumping after various dark times. Pumping was supposed to have a weak effect on nuclear polarization during the detection time. The degree of polarization was measured as a function of dark time by varying t_d from 20 μs to 50 ms. **Fig. 5a** shows the Hanle curves obtained for several dark times, and **Fig. 5b** shows their central portions. It is clear that the curves corresponding to short dark times are similar to that obtained under CW pumping (see **Fig. 1**). In particular, a pronounced W profile is observed, and the curve is wider. An increase in dark time smoothies out the W profile and reduces the width of the Hanle curve.

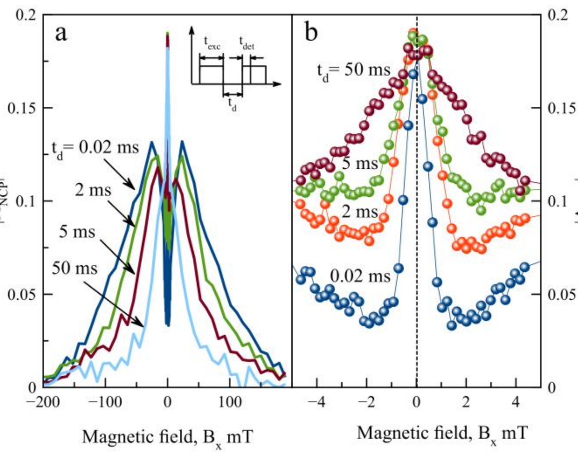

Fig. 5. Hanle curve at constant excitation time $t_{exc} = 100$ ms, parameterized by dark time t_d: (a) complete curves; (b) central portion of curves.

Our experimental findings suggest that the development of nuclear polarization generally leads to a decrease in electron spin polarization in weak transverse magnetic fields and its increase in strong fields. It is obvious that these effects are associated with two different processes.

This implies that dynamics of the longitudinal component $B_{DNP||}$ of nuclear field can be inferred from the time evolution of the dips around the central peak.

Information about behavior of the transverse component $B_{DNP\perp}$ of nuclear field can be extracted by analyzing the width of the Hanle curve. Its large width has been attributed to the formation of a transverse component $B_{DNP\perp}$ of nuclear field stabilized by quadrupole splitting of nuclear spin states along the optical axis (Dzhioev, 2007). Since the longitudinal component $B_{DNP||}$ plays no significant role in strong applied magnetic fields (Meier and Zakharchenya, 1984, Kalevich, 2008), dynamics of $B_{DNP||}$ and $B_{DNP\perp}$ can be inferred separately from behavior of electron spin polarization in weak and strong fields, respectively. Accordingly, to analyze experimental data, expressions are required that relate the degree of electron polarization to the magnitudes of the corresponding DNP components.

Measured degree of photoluminescence polarization is proportional to the electron spin projection on the viewing direction.

$$S_z = S * \cos \vartheta^2 = S \frac{B_{tot,z}^2}{B_{tot}^2}. \tag{2}$$

Here S quantifies the degree of optically induced spin orientation and ϑ is the angle between the viewing direction and the total field $B_{tot} = B + B_N$, which is the sum of the applied field B and the nuclear field, $B_N = B_f + B_{DNP}$, including the effective nuclear spin fluctuation field, B_f, generated by randomly oriented nuclear spins (Merkulov, 2002). Since the electron spin in a quantum dot interacts with limited number of nuclear spins, the contribution due to fluctuations is significantly larger than that for donor bound electron spins in a bulk material, amounting to several tens of milliteslas (Petrov, 2009). Therefore, we can evaluate only an ensemble averaged spin.

In summary, the degree of electron spin polarization can be represented by the general expression

$$\rho = \left\langle S_z \right\rangle / S = \frac{\left(B_{DNP\perp}^2 + 0.5 B_{f\perp}^2 \right)}{\left(B + B_{DNP||} \right)^2 + B_{DNP\perp}^2 + B_f^2}, \tag{3}$$

where $\left\langle B_f^2 \right\rangle = \left\langle B_{f||}^2 \right\rangle + \left\langle B_{f\perp}^2 \right\rangle = 3 \left\langle B_{f||}^2 \right\rangle$. The last relation holds when dynamic nuclear polarization is insignificant and nuclear spin fluctuations are statistically isotropic.

Experimental data can be analyzed by simplifying expression (3) in two special cases of particular importance. According to (Meier and Zakharchenya, 1984), the longitudinal component $B_{DNP||}$ of nuclear field appears only in the W-profile region of the Hanle curve, where the applied field is negligible compared to the nuclear spin fluctuation field (Dzhioev, 2002, Merkulov, 2002). Then, it holds for this region that

$$\rho \approx \frac{\left(B_{DNP\perp}^2 + 0.5\left\langle B_{f\perp}^2 \right\rangle\right)}{\left(B_{DNP\|}\right)^2 + B_{DNP\perp}^2 + 3\left\langle B_{f\|}^2 \right\rangle}. \tag{4}$$

In strong applied magnetic fields (as $B_{DNP\|} \to 0$), the degree of polarization becomes

$$\rho \approx \frac{\left(B_{DNP\perp}^2 + 0.5\left\langle B_{f\perp}^2 \right\rangle\right)}{B^2 + B_{DNP\perp}^2 + 3\left\langle B_{f\|}^2 \right\rangle}. \tag{5}$$

Thus, we can examine the time dependence of ρ in strong and weak magnetic fields to determine the respective kinetics of the longitudinal and transverse components of nuclear polarization

For describing the rise and decay of the longitudinal component of nuclear polarization were found the following expression:

$$\rho \approx \frac{a^2 + 1}{c^2\left(1 - e^{-t/\tau}\right)^2 + a^2 + 3}, \tag{6}$$

$$\rho \approx \frac{a^2 + 1}{c^2 e^{-2t/\tau} + a^2 + 3}. \tag{6'}$$

Expression (5), valid for strong applied magnetic fields, can be written analogously to describe the rise and decay of the transverse component of nuclear polarization, respectively:

$$\rho \approx \frac{\left(1 - e^{-t/\tau}\right)^2 + c'^2}{a'^2 + \left(1 - e^{-t/\tau}\right)^2 + 3c'^2}, \tag{7}$$

$$\rho \approx \frac{e^{-2t/\tau} + c'^2}{a'^2 + e^{-2t/\tau} + 3c'^2}. \tag{7'}$$

Figure 6, 7 shows the results of an analysis of the time dependent Hanle curves, which are measured after the start of optical pumping. The values of ρ are refined by taking into account photoluminescence depolarization due to contributions from neutral quantum dots. Experimental data were processed to determine the time resolved degrees of polarization corresponding to several applied magnetic field strengths. **Figure 6** demonstrates a wide difference between kinetics of degree of polarization under weak and strong field conditions (few milliteslas and higher than 20 mT, respectively).

At $B = 2$ mT (the lowest point of a dip), where the dominant role is played by $B_{DNP||}$, the degree of electron spin polarization ρ decreases with time elapsed (**Fig. 6**), signifying an increase in $B_{DNP||}$. We found that the time dependent degree of polarization determined from experimental data can be fitted by Expression (6) only if the transverse component of nuclear field is sufficiently weak, $B^2_{DNP\perp} \ll \left\langle B^2_f \right\rangle$. Using the resulting approximation, we estimated the characteristic rise time for $B_{DNP||}$, $\tau_{||} \approx 6$ ms.

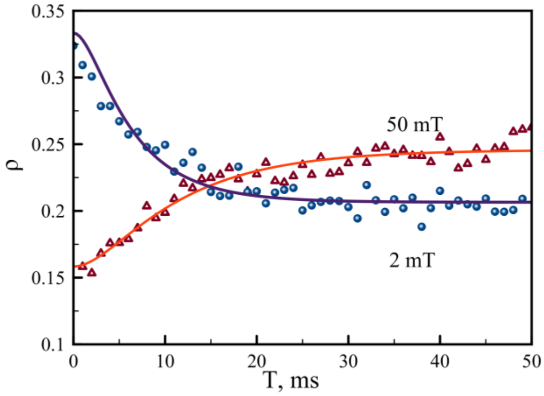

Fig. 6. Example of time-dependent degree of polarization. Symbols represent experimental results; solid curves are approximations by functions (6) and (7).

Figure 7 shows the rise time τ of the transverse component of nuclear polarization. It demonstrates that the time linearly increases from approximately 2.5 to 15 ms with an applied field between 20 and 100 mT.

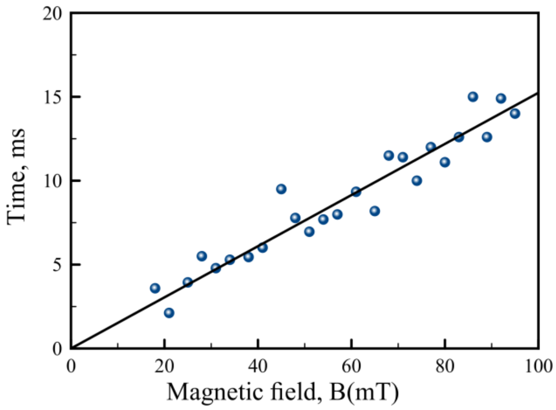

Fig. 7. Field dependence of the build-up time of the transverse DNP field component.

An analogous procedure was used to analyze the shape of the Hanle curve as a function of dark time (**Fig. 5**). Measurement results were converted into spin polarization kinetics for several values of applied magnetic field strength (as in **Fig. 6**), and the resulting curves were fitted by (6′) and (7′). The curves in **Fig. 8** are examples of such fits. The fitting parameters were used to evaluate the initial longitudinal and transverse nuclear fields, as well as the corresponding decay times. The decay time of the longitudinal component calculated by using the data for $B = 2$ mT was found to be $\tau \approx 5.5$ ms, which is close to the corresponding rise time reported above.

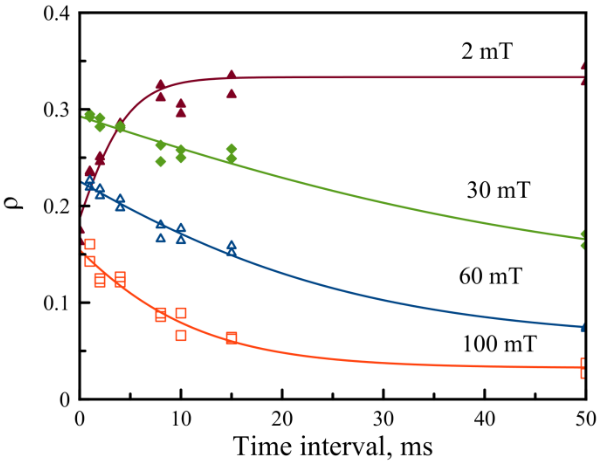

Fig. 8. Kinetics of degree of polarization at several magnetic field strengths, obtained by analyzing measurements results for various dark times. Symbols represent experimental data, solid curves are approximations by functions (6′) and (7′).

However, the decay time of the transverse component of nuclear polarization differs significantly from its rise time. Moreover, its time variation in an applied magnetic field exhibits an opposite trend: whereas the rise time increases with field strength (**Fig. 7**), the decay time rapidly decreases (**Fig. 9**). Accordingly, these times are approximately equal in strong magnetic fields but differ by a factor of several tens at B = 20 mT.

Our analysis shows that the longitudinal and transverse components of nuclear polarization in the quantum dots under study exhibit widely different dynamical patterns. The behavior of longitudinal polarization is relatively simple. After the start of optical pumping, this component increases with a characteristic time of approximately 6 ms to a limit magnitude corresponding to an effective nuclear field of 30 to 40 mT. After the end of pumping, the longitudinal component decays over a similar time scale.

The behavior of the component of dynamic nuclear polarization perpendicular to the applied magnetic field is much more complicated. Observations that defy any straightforward explanation include difference between the buildup and decay times, their opposite variation with applied magnetic field, and increase in magnitude of this component with applied field strength.

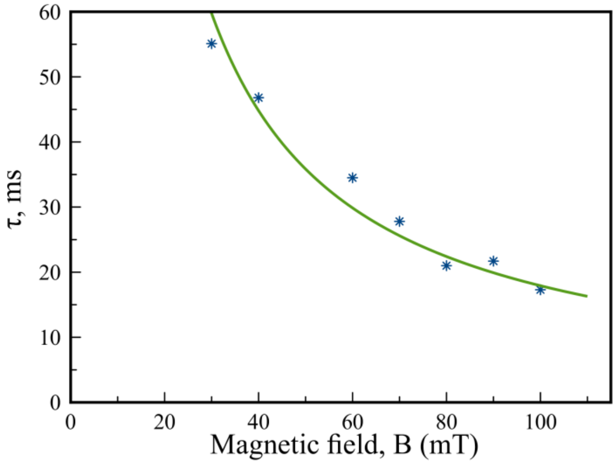

Fig. 9. Field dependence of decay time of the transverse DNP-field component.

In our view, these differences are mainly due to the fact that the dominant contributions to the buildup of longitudinal and transverse polarization components come from states with different spin projections on the viewing direction.

In PII we performed an experimental study of time dependent circular polarization of photoluminescence from quantum dots as a function of magnetic field applied perpendicular to the optical axis (time resolved measurements of the Hanle effect). Experimental data were analyzed using an original approach based on separate treatment of the longitudinal and transverse components of nuclear polarization in quantum dots characterized by strong quadrupole splitting of nuclear spin states. The phenomenological model proposed here takes into account the contribution of nuclear spin fluctuations, which were ignored in previous analyses of experimental data on the Hanle effect. The model is validated both by our finding that nuclear spin fluctuation field is independent of applied field and by good quantitative agreement with results of other studies (Cherbunin, 2009, Petrov, 2009). Using this model to analyze experimental results, we obtained detailed information about the rise and decay times of each component of nuclear polarization in quantum dots in a transverse magnetic field. The rise and decay times of the component parallel to the applied field were found to be almost equal (approximately 5 ms). However, the dynamics of the transverse component is much more complicated: the corresponding rise and decay times differ widely and have opposite dependence on magnetic field strength. Furthermore, the magnitude of the transverse component created by continuous wave pumping significantly increases with applied field strength. We attribute this unexpected behavior of nuclear polarization to nuclear spin relaxation via interaction with photo excited carriers.

2.3 Role of nuclear spin fluctuations

In PIII, we report on detailed measurements of the Hanle effect in (In,Ga)As/GaAs QDs in the weak-field range (0–20 mT field strength), where the effect of the nuclear spin fluctuations (NSF) is expected to be the strongest. We have measured a set of Hanle curves under optical excitation of moderate intensity and at different strengths of an additional magnetic field applied along the optical axis (longitudinal magnetic field). The experimental Hanle curves are compared with the results of calculations using two models, one including NSF and the other one taking into account only mean Overhauser fields. In both theories, the mean Overhauser field has been calculated within the spin temperature approach. Our analysis shows that the mean-field model fails to describe the features of the Hanle curve around zero transversal field, where the so-called W structure appears in a certain range of longitudinal fields B_z. The model including NSF, on the other hand, yields good fits of the experimental data, with a reasonable choice of parameters, for all experimental conditions except for the exact compensation of the Knight field with B_z. In the latter case, nuclear quadruple effects due to strain in the QDs probably play the dominant role.

The effect of the longitudinal magnetic fields, ranging from −20 to +20 mT, on the Hanle curve and the dependences calculated in the framework of the nuclear spin cooling model are investigated. For positive B_z, which, for the helicity of excitation used in our experiments, is codirected to the Knight field the experimental and calculated curves are in qualitative agreement with each other. The analysis shows that the effective nuclear field in this case is codirected to the external magnetic field and thus "amplifies" it. This amplification results in a gradual decrease of spin polarization and, correspondingly, of PL polarization beyond the central peak with rising B_z.

When B_z is negative, the effective field is antiparallel to the Knight field. If $B_z = -B_e$, the compensation of the longitudinal component of total field occurs. According to Refs. (Meier and Zakharchenya, 1984, Page, 1977), nuclear spin cooling is not possible in this case. This should result in the disappearance of the W structure, as it is shown in **Fig. 10** for the Hanle curve calculated for $B_z = -1$ mT. At more negative B_z, the W structure appears again, but the additional maxima run away from the central peak with increasing $|B_z|$, maintaining the same amplitude as the central peak. This behavior of the calculated Hanle curves is explained by the fact that in this case the nuclear field is directed against the total effective magnetic field affecting the nuclei. The x component of the nuclear field, B_{Nx}, is compensated by the transverse magnetic field B_x at some magnitude of B_x, giving rise to the additional maxima.

These numerical results, however, are in strong contradiction to our experimental observations (**Fig. 10**).The central peak of the measured Hanle curves is higher than the other parts of the Hanle curve at any negative B_z. We want to stress that the disagreement between the theory and the experiment cannot be eliminated for any set of values of the adjustable parameters. Therefore this contradiction is of principal importance and indicates that the model of mean nuclear field ignores some mechanism causing depolarization of the electron spin at nonzero transverse magnetic field, including points where it is totally compensated by the nuclear field. In the cooling model, such mechanisms are not provided which does not get a description of the experimental data.

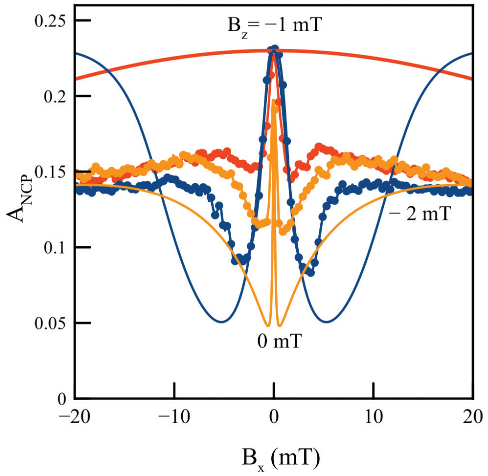

Fig. 10. Comparison of Hanle curves calculated in the framework of standard cooling model (solid lines) with the experimental data (points) for negative longitudinal external fields B_z.

To extend the standard cooling model, we suppose that the effective nuclear field consists of a regular component, B_N, created by the nuclear polarization, and a fluctuating component, B_f, appearing due to the random orientation of the limited number of nuclear spins interacting with the electron spin (Merkulov, 2010). The estimates given in (Ikezawa, 2005, Pal, 2009) for similar QDs show that the average magnitude of the fluctuating nuclear field is of the order of tens of milliteslas.

The dependence of the average electron spin polarization on the transverse external magnetic field within this approximation is a bell-like curve, which can be well fitted by a Lorentzian:

$$\rho(B) \approx \frac{\langle B_{fz}^2 \rangle}{B^2 + \langle B_f^2 \rangle}. \tag{8}$$

Some generalization of Eq. (8) is required to describe electron spin polarization under our experimental conditions. We need to take into account the regular nuclear field B_N with nonzero components B_{Nx} and B_{Nz} created by the dynamic polarization of nuclei. In addition, in our experiments, the external magnetic field has not only the transverse component but also some longitudinal one. For this case we can write down the following expressions for the z and x components of the averaged electron spin $\boldsymbol{S}_{\|}$:

$$S_z = S_0 \frac{(B_z + B_{Nz})^2 + \langle B_{fz}^2 \rangle}{(B_x + B_{Nx})^2 + (B_z + B_{Nz})^2 + \langle B_f^2 \rangle}, \tag{9}$$

$$S_x = S_0 \frac{\left(B_z + B_{Nz}\right)\left(B_x + B_{Nx}\right)}{\left(B_x + B_{Nx}\right)^2 + \left(B_z + B_{Nz}\right)^2 + \left\langle B_f^2 \right\rangle}. \tag{9′}$$

Here we assume that the regular nuclear field B_N is directed along the total effective field B_{Ntot} acting on the nuclei, which consists of the external magnetic field $B_x + B_z$, and the Knight field $B_e = b_e S_{||}$, created by hyperfine interaction with the electron spin. According to standard cooling model we can write the nuclear field the following way

$$\boldsymbol{B}_N = \boldsymbol{B}_{tot}^{(N)} \frac{b_N \left(\boldsymbol{B}_{tot}^{(N)} \cdot \boldsymbol{S}_{||}\right)}{B_{tot}^{(N)2} + \xi B_L^2} \cdot 4I(I+1)/3\,. \tag{10}$$

The above equation allows one to obtain the following expressions for the x and z components of the nuclear field:

$$B_{Nx} = \left(B_x + b_e S_x\right) \frac{b_N \left(B_z S_z + B_x S_x + b_e S_x^2 + b_e S_z^2\right)}{\left(B_x + b_e S_x\right)^2 + \left(B_z + b_e S_z\right)^2 + \xi B_L^2}, \tag{10′}$$

$$B_{Nz} = \left(B_z + b_e S_z\right) \frac{b_N \left(B_z S_z + B_x S_x + b_e S_x^2 + b_e S_z^2\right)}{\left(B_x + b_e S_x\right)^2 + \left(B_z + b_e S_z\right)^2 + \xi B_L^2}. \tag{10″}$$

The coefficient b_e is given, in principle, by be $= -(16\pi/3)\mu_B \xi^2$, where μ_B is the Bohr magneton and ζ is the electron density on a nuclear site. The negative sign means that the direction of the Knight field is opposite to that of the electron spin. Because the electron density is dependent on the QD size, which can sufficiently vary from dot to dot, the value of ζ is unknown a priori.

We solved the whole system of Eqs. (9), (9′), (10′), and (10″) and used their real roots for modeling the Hanle curves, slightly varying the fitting parameters. Examples of the calculated Hanle curves are shown in **Fig. 11.** As seen, reasonable agreement between calculated and measured curves is observed for negative B_z. Some deviations from the experiment occur for magnetic fields B_z in the range from −0.5 to −1 mT, where the theoretically calculated amplitude of the central peak is considerably smaller than the one observed experimentally (see inset in **Fig. 11**). The strong decrease of the peak amplitude obtained in the calculations is due to the depolarization of the electron spin by the nuclear spin fluctuations, when the longitudinal component of total field disappears and the nuclear field does not build up. Experiments also show a decrease of the central peak of about 20%, which is, however, significantly smaller than the one predicted theoretically. A possible reason for this discrepancy between the theory and the experiment could be related to the spread of Knight fields in the QD ensemble, which is ignored in

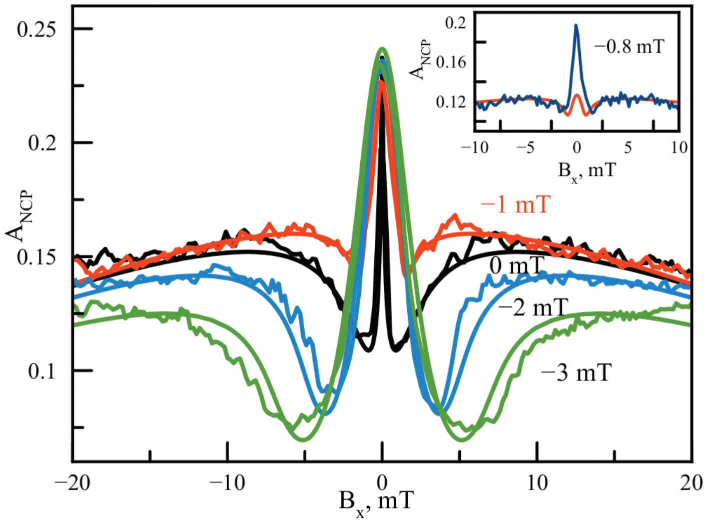

Fig. 11. Experimentally measured curves (noisy lines) and results of calculations taking into account the NSF (smooth solid lines) for negative longitudinal external fields B_z.

the theory. Another possible reason is the polarization of quadrupole-split nuclear spin states, which can stabilize the electron spin polarization (Dzhioev, 2007). Further study is needed to clarify this problem.

The results of the calculations allow us to conclude that the effect of nuclear spin fluctuations is indeed important for the QDs under study. The good agreement between theory and experiment for the whole range of B_z (with the only exception mentioned above) allows us to consider in more detail the physical meaning of the parameters obtained from the fitting and their dependence on the longitudinal magnetic field

We find that the NSF amplitude, $\sqrt{\left\langle B_{fz}^2 \right\rangle}$, can be chosen close to 25 mT for all the Hanle curves measured at various longitudinal magnetic fields. The good overall correspondence of the simulated and measured Hanle curves confirms the validity of the model developed.

The analysis of experimental data has confirmed the prediction of (Merkulov, 2002) about the significant influence of nuclear spin fluctuations on the electron spin orientation due to strong localization of the electron in QDs. The observed behavior is considerably different from that in bulk semiconductor alloys studied in many works (Meier and Zakharchenya, 1984), in which the electron density is spread out over a huge number of nuclei and the effect of the NSF, as a rule, is negligibly small. The analysis allows us to evaluate the maximal value of the effective field of nuclear polarization created in studied quantum dots by optical pumping to be about 200 mT. We have also found that the effective field acting on the nuclei from the electron spin (Knight field) in the sample under study is near 1 mT when the electron spin is almost fully oriented.

2.4. Resonant nuclear spin pumping

In PIV it is discussed the observation of resonant optical pumping of nuclear spin polarization in an ensemble of singly charged (In,Ga)As/GaAs quantum dots subject to a transverse magnetic field. Electron spin orientation by circularly polarized light with the polarization modulated at the nuclear spin transition frequency is found to create a significant nuclear spin polarization, precessing about the magnetic field.

An efficient technique to reach significant nuclear spin polarization (NSP) is optical pumping (Meier and Zakharchenya, 1984). A relatively strong nuclear polarization (tens of percent) can be created by optical pumping of QDs in a magnetic field parallel to the optical axis (longitudinal field) (Gammon, 2001, Braun, 2006, Tartakovskii, 2007). Optical pumping can create dynamic nuclear polarization also in a transverse magnetic field (Paget, 1977, Krebs, 2010). It is commonly accepted (Dyakonov, 2008) that the nuclear polarization is directed along the external magnetic field. The appearance of this longitudinal component corresponds to a difference in population of nuclear Zeeman sublevels and, therefore, is usually treated in terms of "nuclear spin cooling."

In PIV we experimentally demonstrate that not only longitudinal but also transverse NSP components of remarkable magnitude can be created in QDs. We find that polarization-modulated optical excitation of singly charged (In,Ga)As/GaAs QDs results in a strong change of the dependence of photoluminescence (PL) polarization on a transverse magnetic field (Hanle curve). We identify resonances related to spin transitions of the gallium, indium, and arsenic nuclei which are influenced by magnetic field and quadrupole interaction. We suggest that the observed effect is a clear indication of a phasing of the nuclear spin states that corresponds to the creation of transverse NSP components precessing about the magnetic field.

Figure 12 shows the magnetic field dependence of the NCP amplitude measured for different excitation protocols. All curves show a decrease of NCP (the Hanle effect) with increasing magnetic field. For continuous-wave (cw) excitation with fixed polarization helicity the Hanle curve consists of a narrow central peak and broad shoulders, together forming the so-called W structure (Paget, 1977). The W structure clearly indicates NSP that has built up for these excitation conditions. When amplitude modulation (AM) with a low on-off time ratio is used, the Hanle curve has a Lorentzian shape. Switching on polarization modulation (PM) in addition to the amplitude modulation ("AM + PM" protocol in the inset of **Fig. 12**) does not change notably the Hanle curves, meaning that nuclear polarization does not develop under such excitation conditions and the Hanle curve is determined solely by electron spin dynamics. Therefore we call the curve measured in that way the electronic peak ("*e* peak").

The Hanle curve measured for polarization modulation only [curve (3) in Fig. 12] shows two additional maxima at approximately $B = \pm 10$ mT. The appearance of such additional maxima is the main topic of PIV. The position of these additional maxima, resonances, strongly depends on the polarization modulation frequencies (**Fig. 13**). The positions are shifted to higher fields with increasing modulation frequencies. The frequency shift of the shoulders indicates their resonant character.

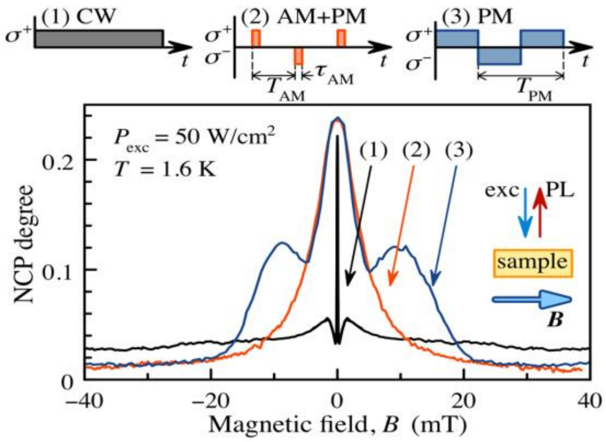

Fig. 12. Hanle curves measured for cw optical excitation [curve (1)], for modulated polarization of the excitation [$T_{PM} = 40\ \mu$s, curve (3)], and for polarization and amplitude modulation of the excitation [$T_{AM} = 20\ \mu$s, $\tau_{AM} = 5\ \mu$s, curve (2)]. The top panels sketch these different timing protocols.

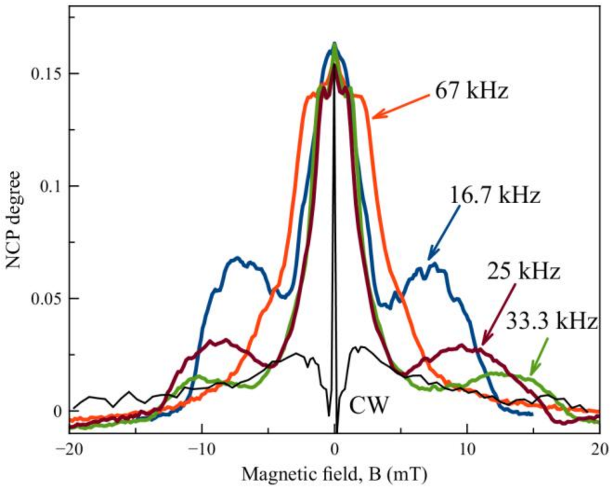

Fig. 13. Hanle curves measured for excitation polarization modulated at different frequencies.

Even stronger modification of the Hanle curves is observed when a radio-frequency (RF) field that is synchronous with the modulation of polarization is applied. The RF field generated with a magnetic component directed along the optical axis (**Fig. 14**).

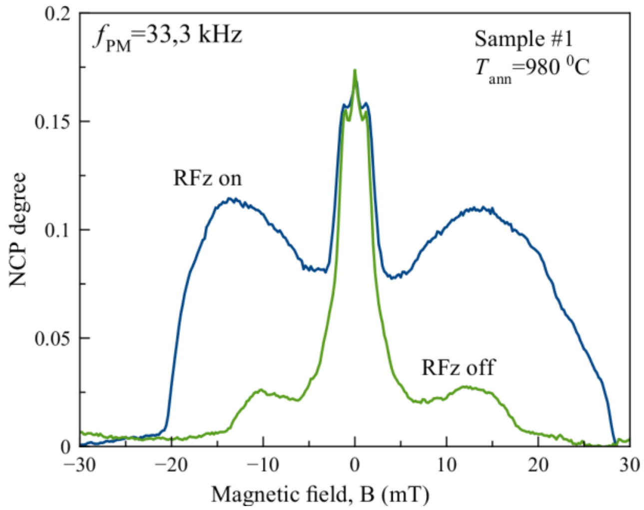

Fig. 14. The blue curve is measured with applying a Rfz-field. The green curve is measured with no RF-field.

To identify the resonances which contribute to the Hanle curves, we analyze the nuclear spin splitting in a transverse magnetic field (**Fig. 15**). The calculations are made taking into account the quadrupole splitting of nuclear spin states caused by the strain-induced gradient of the crystal field as well as by the statistical population of crystal sites by Ga and In atoms. The QDs under study contain several types of nuclei (including isotopes): ^{69}Ga, ^{71}Ga, ^{75}As, ^{113}In, and ^{115}In. The principle axis of the strain-induced gradient is directed along the growth axis (*z* axis).

We calculate the splitting of nuclear spin states by the magnetic field and the quadrupole interaction assuming a strain magnitude $\varepsilon_b = 0.01$ as estimated in Flisinskii, 2010 for sample annealed at T = 980 °C. Each resonance at a calculated energy is modeled by a Gaussian with amplitude and width as fit parameters. Our analysis shows that the Hanle curves measured at different modulation frequencies can be described well using the calculated resonance energies.

The analysis of a Hanle curve measured with RF-field application is shown in **Fig. 16**. To work out the resonances more clearly, we subtracted the *e* peak from the experimentally measured Hanle curve. The central part of the curve is given by the resonances $\langle +1/2| \leftrightarrow |-1/2\rangle$. The wide part of the Hanle curve can be well described by the resonances $\langle +3/2| \leftrightarrow |-3/2\rangle$ or the In and Ga nuclei, as well by the resonances $\langle +5/2| \leftrightarrow |-5/2\rangle$ for the In nuclei.

At higher modulation frequencies, additional maxima appear at the Hanle curves as it is shown in **Fig. 17**. According to the calculations of energy splitting of the nuclear spin states (see **Fig. 15**), these maxima can be attributed to “quadrupole” resonances

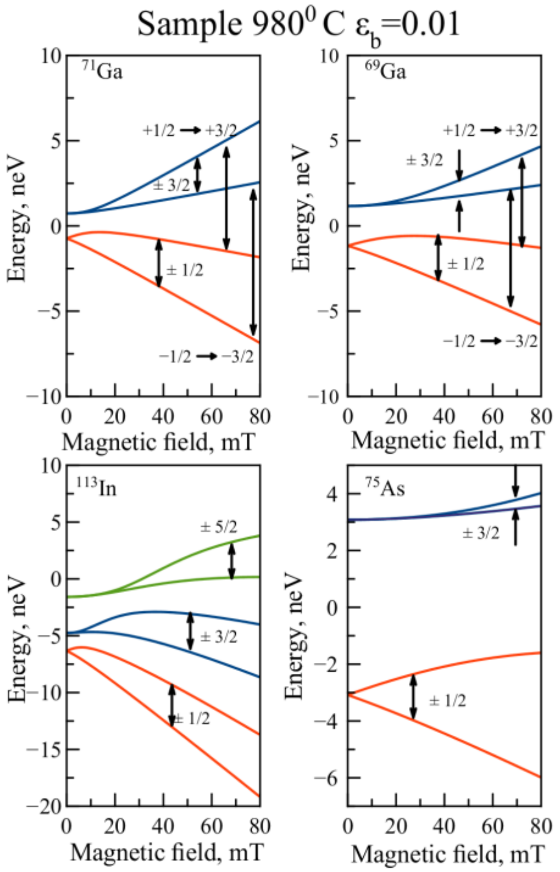

Fig. 15. Calculated energies of nuclear spin sub-levels for isotopes ^{71}Ga, ^{69}Ga, ^{113}In, and ^{75}As as functions of magnetic field.

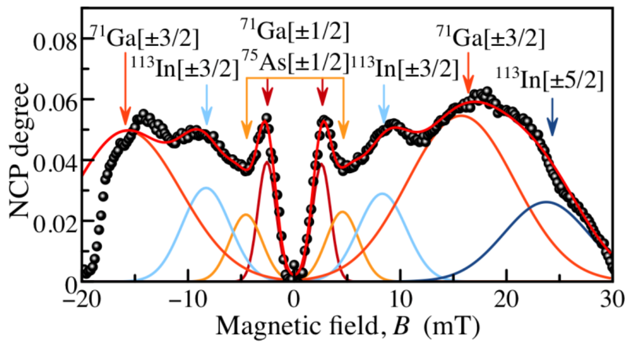

Fig. 16. Gaussian decomposition of Hanle curve measured at fPM = 67 kHz in presence of RFz field for the sample 980°C.

$+1/2 \leftrightarrow +3/2$ and $-1/2 \leftrightarrow -3/2$. As seen in **Fig. 17**, inclusion of these resonances into consideration allows one to adequately describe the experimentally measured Hanle curve.

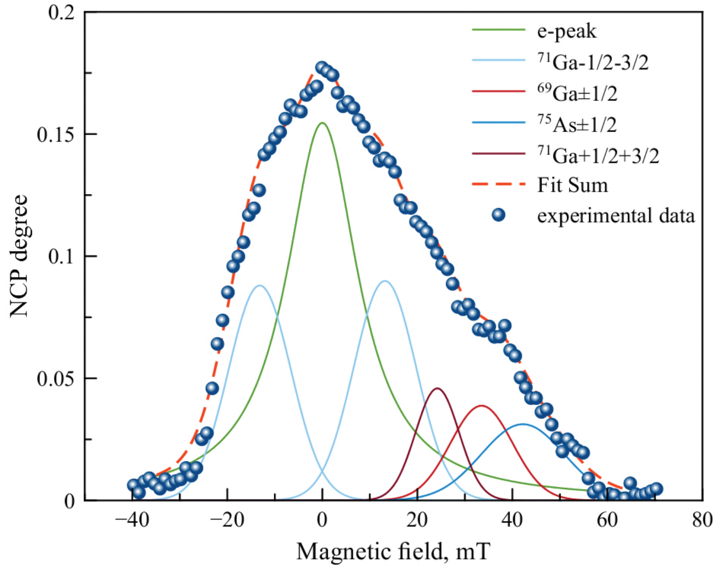

Fig. 17. Gaussian decomposition of Hanle curve measured at $f_{PM} = 600$ kHz in presence of RFz field for the sample 980°C. The colored Gaussians model resonances, whose positions are fit parameters.

The comparizon of the resonance positions obtained from the experiment with those obtained from the calculation of Zeeman splitting of nuclear spin states are shown in **Figs. 18, 19. Fig. 18** demonstrate the data for observed transitions $+1/2 \leftrightarrow -1/2, +3/2 \leftrightarrow -3/2$, and $+5/2 \leftrightarrow -5/2$.

Figure 19 demonstrates similar data for transitions $+1/2 \leftrightarrow +3/2$ and $-1/2 \leftrightarrow -3/2$. Resonant frequencies for these transitions are determined with less accuracy however

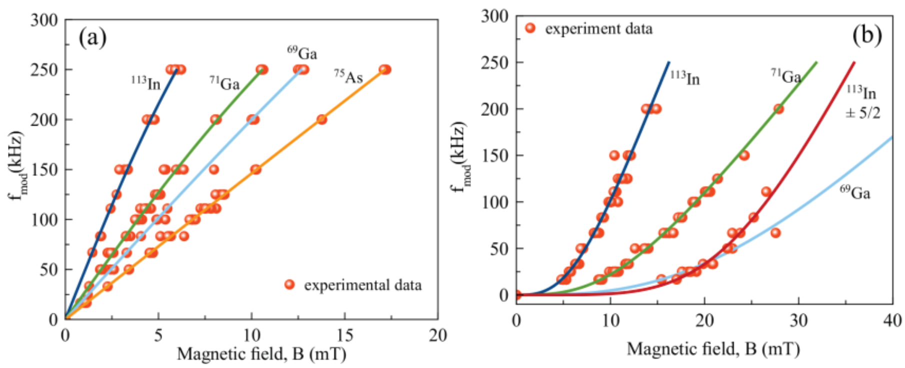

Fig. 18. Theoretically calculations of frequency dependence on magnetic field for nuclear spin states Ga, In, As and compare it with experimental data (a) for transitions and (b) for $+3/2 \leftrightarrow -3/2$, $+5/2 \leftrightarrow -5/2$.

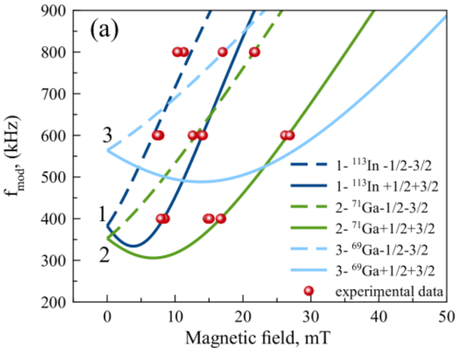

Fig. 19. Theoretically calculations of frequency dependence on magnetic field for nuclear spin states Ga, In and compare it with experimental data.

the Hanle curve cannot be adequately described with no consideration of these resonances.

We want to note here that the explanation of the Hanle curve peculiarities by peaks centered at certain resonance fields is not generally accepted, but rather the object of discussion. Further, the authors of Refs. Tartakovskii, 2007, Kalevich, 1980, Eickhoff, 2002 observed experimentally dispersion like peculiarities superimposed on a smooth Hanle curve.

We assume that the key point for understanding the origin of the resonances observed in our work is the appearance of a significant component of nuclear polarization in the plane perpendicular to the external magnetic field (transverse component).

The creation of a transverse component is a resonant process, as our experiments show. For excitation with constant polarization of light, polarized nuclear spins are created with arbitrary phases so that a transverse component is not created. Only resonant modulation of the optical polarization results in cophase pumping of a large number of nuclear spins, giving rise to resonant amplification of the transverse component of nuclear polarization.

In conclusion, we have observed a significant nuclear polarization in the plane perpendicular to the external magnetic field in semiconductor QDs. The polarization is created by circularly polarized optical pumping modulated at a frequency that is resonant to one of the nuclear spin transitions. The effect, which may be termed resonant optical pumping of nuclear spin polarization, is evidenced by several intense peaks in the Hanle curve. The number and amplitude of peaks increase for joint action of polarization modulation of optical excitation and synchronous RF-field application. In particular, the RF-field enhances resonances related to transitions between $|\pm 3/2\rangle$ nuclear states split off from the $|\pm 1/2\rangle$ states by a quadrupole interaction.

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