Dynamical nuclear polarization and nuclear magnetic resonance in a (In,Ga)As/GaAs quantum dot ensemble

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Abstract. Peculiarities in the dependence of the photoluminescence polarization of (In,Ga)As/GaAs quantum dots on the transverse magnetic field were observed under radio frequency (RF) field application. We show that these peculiarities result from the suppression of dynamical polarization of the $^{71}$Ga and As nuclei by the RF field. The effect of quadrupole splittings on the nuclear spin dynamics has been evaluated.

Optically detected nuclear magnetic resonance (ODNMR) has been used as an effective tool for studying the dynamics of nuclear polarization in bulk semiconductors [1, 2] and semiconductor quantum wells [2, 3, 4]. The high sensitivity of optical detection makes ODNMR promising for studying semiconductor quantum dots (QDs), which contain a relatively small number of nuclei. At the same time, there are very few experimental data on ODNMR in such structures [5, 6].

In this paper we present experimental studies of nuclear magnetic resonances in (In,Ga)As/GaAs QDs detected in the polarization of their PL in transverse magnetic field (Voigt geometry). Analysis of the experimental results allows us to identify the nuclear species responsible for DNP formation in the magnetic field range 0–10 mT.

We studied two samples, #1 and #2, with self-assembled (In,Ga)As/GaAs QDs annealed at temperatures 900 °C and 980 °C, respectively. The annealing causes relaxation of the internal strain in the QDs, which is one of the important sources of the quadrupole splitting of the nuclear spin states, which considerably affects the nuclear spin polarization. In the structures studied, each QD contains on average one resident electron, which is supplied by delta-doped barrier layers. The optically oriented electron spin interacts with the nuclear spins thereby efficiently polarizing the nuclei. At the same time, the electron spin polarization degree can be used as an indicator of the nuclear spin polarization.

The electron spin polarization was detected via the negative circular polarization (NCP) of the photoluminescence (PL) [7]. The PL was excited by a cw Ti-Sapphire laser, whose wavelength was tuned to the wetting layer optical transition. The PL polarization was detected using a photoelastic modulator operating at a frequency of 50 KHz and a photon counting system. All experiments were performed at a temperature $T = 2$ K. A split-coil superconducting magnet was used for the magnetic field (along the x-axis) to the samples. A radio frequency (RF) field was generated by a coil with its axis (y-axis) oriented perpendicular to both the optical axis and the external magnetic field.

We studied the PL polarization degree as function of the magnetic field applied perpendicular to the optical pumping direction parallel to z-axis (Hanle effect). We found that the Hanle curves have a W-shape, which can be explained by the development of a nuclear field along the total field acting on the
nuclei [1, 8]. This total field is the sum of the external magnetic field and the Knight field of the spin polarized electron. For sufficiently strong optical excitation the nuclear field can be considerably larger than the external field and, therefore, is able to depolarize the electron spin. The efficiency of nuclear spin polarization along the total field gradually goes down with the magnetic field rise thus forming the W-shape of Hanle curve [8].

We studied the modification of the Hanle curves by RF field application. The effect of RF field on sample #1 is shown in Fig. 1(a), revealing an increase of spin polarization. The differences of Hanle curves measured in presence and absence of RF excitation are shown at the bottom of Fig. 1(a) and consists of wide peaks, which shift towards larger external magnetic field with increasing RF frequency. The large width of these peaks does not allow us to uniquely identify, which nuclei are responsible for the observed resonances.

Figure 1(b) shows the Hanle curves measured for sample #2. The stronger annealing of this sample changes the Hanle curves considerably. In this case, application of RF field is accompanied by the appearance of pronounced resonances. The resonance position is found to depend linearly on RF frequency. We suggest that the two observed resonances correspond to the $\left|\frac{1}{2}\right> \leftrightarrow \left|\frac{-1}{2}\right>$ transitions of the $^{71}\text{Ga}$ and As nuclei which are subject of a change of their gyromagnetic ratios due to the quadrupole splitting of the nuclear states. The source of this splitting is the strain-induced electric field gradient, $\nabla F$, which lifts the degeneracy of nuclear spin states with different absolute values of the projections of nuclear spin on the principal axis of the tensor $\nabla F$ [9].

The Hamiltonian of the nuclear spin $I$ interaction with an external magnetic field $B$ in presence of a
quadrupole splitting along the \( z \) axis can be written as [9]

\[
\mathcal{H} = -\gamma \hbar \mathbf{I} \cdot \mathbf{B} + \frac{\hbar Q}{2} \left( I_z^2 - I(I+1)/3 \right),
\]

(1)

where \( \gamma \) is the nuclear gyromagnetic ratio, and \( \hbar \) is the Planck constant. The quadrupole splitting is determined solely by the frequency

\[
\nu_Q = \frac{3eV_{zz}Q}{2I(2I-1)\hbar}.
\]

(2)

Here \( e \) is the elementary charge, \( Q \) is the nuclear quadrupole momentum, and \( V_{zz} = S_{11}e_{zz} \) is the strain-induced electric field gradient at the nuclear site. The values of quadrupole momentum \( Q \) and of \( S \)-tensor for different isotopes are extracted from experimental data on nuclear acoustic resonance [10, 11]. We also estimated the value of strain \( e_{zz} \) to be about 0.01 (one percent) for the QD annealed at 980 °C using a cylindrically symmetric model for the QD [12] and the transversal isotropic approximation in the frame of continuum elasticity theory [13]. The magnitude of \( \nu_Q \) obtained for gallium, arsenic, and indium nuclei are summarized in Table 1.

| Isotope | \( I \) | \( Q \) (mbar) | \( S_{11} \) (statC/cm\(^3\)) | \( \nu_Q \) (kHz) |
|---------|-------|----------------|----------------|----------------|
| \(^{71}\text{Ga}\) | 3/2 | 107 | \( 9.1 \times 10^{12} \) | 353 |
| \(^{69}\text{Ga}\) | 3/2 | 171 | \( 9.1 \times 10^{15} \) | 564 |
| \(^{75}\text{As}\) | 3/2 | 214 | \( 1.31 \times 10^{16} \) | 1490 |
| \(^{113}\text{In}\) | 9/2 | 759 | \( 1.67 \times 10^{16} \) | 388 |
| \(^{115}\text{In}\) | 9/2 | 770 | \( 1.67 \times 10^{16} \) | 383 |

Table 1. Nuclear quadrupole parameters. The values of \( Q \) are taken from Ref. [10] and the values of \( S_{11} \) are taken from Ref. [11]. Quadrupole frequencies \( \nu_Q \) are calculated for the value of strain \( e_{zz} = 0.01 \).

Calculated Zeeman splittings of different levels of As and In nuclei obtained by direct diagonalization of Hamiltonian (1) are shown in Fig. 2. The Zeeman splittings strongly depend on the angle between the principal quadrupole axis (vertical axis) and the magnetic field direction for the case of strong quadrupole splitting (or weak magnetic field). In particular, when the magnetic field is applied along the quadrupole axis, there is a normal Zeeman splitting for which \( \Delta E_{\pm n/2} = n\Delta E_{\pm 1/2} \) with \( n = 3 \) for As and \( n = 3, 5, 7, 9 \) for In. When the magnetic field is weak enough and applied perpendicular to the quadrupole axis, the \( \pm n \) states are not split for \( n \geq 3 \). At the same time, the splitting of the \( | \pm 1/2 \rangle \) states is larger by a factor of two for the As nuclei and a factor of five for the In nuclei as compared to the absence of a quadrupole splitting. A similar behavior of Zeeman splittings is also obtained for the Ga nuclei. Taking into account the two-fold increase of the gyromagnetic ratio for the \( | \pm 1/2 \rangle \) states, the resonances observed in Fig. 1(b) well correspond to the RF transitions \( |1/2 \rangle \leftrightarrow |-1/2 \rangle \) for the \(^{71}\text{Ga}\) and As nuclei.

Figures 2(c) and 2(d) show the dependence of Zeeman splittings of the As and In nuclei on magnetic field applied perpendicular to the principal quadrupole axis. The quadrupole splittings, \( \nu_Q \), of the nuclei were calculated for a strain \( e_{zz} = 0.01 \), as estimated for sample #2. Even such small strain gives rise to quadrupole splittings, which are larger than the Zeeman splitting of the \( | \pm 1/2 \rangle \) states up to magnetic fields of about 10 mT for the In and 100 mT for the As nuclei. A similar analysis for the Ga nuclei gives a comparable Zeeman splitting at about \( B = 20 \) mT for the \(^{69}\text{Ga}\) and \( B = 15 \) mT for the \(^{71}\text{Ga}\) nuclei. In all cases, the approximation of large quadrupole splittings in the analysis of resonances shown in Fig 1(b) is valid.

In conclusion, we have optically detected nuclear magnetic resonances in an ensemble of (In,Ga)As/GaAs quantum dots, which is a direct proof of DNP formation. The resonances are detected as relatively wide peaks in the magnetic field dependence of the PL polarization degree, which appear when a radio-frequency field is applied. The origin of this effect is the suppression of dynamical nuclear polarization by the resonant field.
Figure 2. (a) Angular dependence of Zeeman splittings for As nuclei for large quadrupole splittings. The vertical axis corresponds to the principal axis of the quadrupole tensor. (b) The same for In nuclei. (c) Zeeman splittings for As nuclei in magnetic field perpendicular to quadrupole axis. (d) The same for In nuclei.

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