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Optically detected magnetic resonance at the quadrupole-split nuclear states in (In,Ga)As/GaAs quantum dots

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Nuclear-magnetic resonances were detected optically in an ensemble of singly charged (In,Ga)As/GaAs quantum dots. The resonances were found in the magnetic field dependence of photoluminescence polarization (Hanle curve) when applying a radio frequency (RF) field. The frequency dependences of the resonances allow us to ascribe them to transitions between Zeeman states of the ^{71}Ga and ^{75}As nuclei perturbed by quadrupole interaction. Applying an RF-field sweep over a wide frequency range results in a notable narrowing of the Hanle curve, which can be explained by suppression of dynamic nuclear polarization that otherwise is stabilized by the quadrupole splitting.

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The nuclear-spin dynamics in quantum dots has attracted considerable attention as it ultimately limits the spin coherence of carriers.[1] Optically detected nuclear-magnetic resonance (ODNMR) is considered as an effective tool to study the dynamics of nuclear polarization in semiconductors.[2,3] Optical detection of nuclear-spin polarization is possible due to the strong coupling of the electron and nuclear spins via the hyperfine interaction. Due to this interaction, optical orientation of electron spins is accompanied by dynamic nuclear polarization (DNP). In turn, the DNP acts on the electron spins as an effective magnetic field, which changes the electron-spin polarization and alters the splitting of electron-spin levels in external magnetic field (Overhauser shift).[4] Application of radio frequency (RF) fields resonant to the transitions between nuclear-spin states can depolarize the nuclei and thereby change the electron-spin polarization. Optical detection of these changes allows one to observe nuclear-magnetic resonance.[2]

The high sensitivity of optical detection makes ODNMR promising for investigation of semiconductor quantum dots (QDs), which contain a relatively small number of nuclei and therefore can hardly be addressed by standard NMR. The efficiency of ODNMR in combination with single QD spectroscopy was demonstrated studying shallow GaAs/(Al,Ga)As QDs.[5,6] The absence of internal strain in these systems, allowed one to observe resonant changes in the Zeeman splitting of optical transitions in external magnetic field. However, there have been no reports of ODNMR on strained QDs containing nuclei that are subject to quadrupole splittings.[7,8]

In this Rapid Communication we report on ODNMR in an ensemble of singly charged self-assembled (In,Ga)As/GaAs QDs. The negative circular polarization (NCP) of the ground-state emission, well established for QDs charged with a single electron,[9–11] was used to measure the spin polarization of the resident electrons. The NCP was measured as function of transverse magnetic field (Hanle effect) with simultaneous application of RF field. Resonances related to the ^{71}Ga and ^{75}As nuclei, modified by the quadrupole splitting, were observed. The obtained data clearly evidence considerable nuclear polarization in relatively strong transverse magnetic fields of tens of millitesla. We attribute the DNP in this field range to stabilization of nuclear spins by quadrupole splitting.

We studied a heterostructure containing 20 layers of self-assembled (In,Ga)As QDs sandwiched between GaAs barriers with modulation *n*-doped sheets close to each dot layer. Donor ionization supplies every dot with on average a single resident electron. The structure was grown by molecular-beam epitaxy on a (001)-oriented GaAs substrate. Rapid thermal post-growth annealing at temperature of 980 °C provides interdiffusion of indium and gallium atoms between QDs and GaAs barriers so that the InAs QDs are partially dissolved, resulting in a blueshift of the lowest QD optical transition to energies around 1.42 eV.[12] Characteristic optical properties of comparable samples were described in Refs. 13–17. Annealing also causes release of internal strain in the QDs, which is an important source of quadrupole splitting of the nuclear-spin states, considerably affecting the nuclear-spin polarization.[2,7,8] The annealing therefore reduces the variation in quadrupole splittings across the dot, which helped to make the nuclear resonances observable.

The sample was placed in a cryostat with a superconducting magnet such that the field could be applied perpendicular to the structure growth axis (Voigt geometry) along the [110] crystallographic axis. This configuration allows a detailed study of nuclear-spin effects.[2,7,16,18] The experiments were done at sample temperatures $T=1.6$ or $T=4.2$ K. Photoluminescence (PL) was excited by a continuous-wave Ti:Sapphire laser tuned to energy 1.481 eV corresponding to the wetting-layer optical transition, which provides the largest NCP. The PL was dispersed by a 0.5 m spectrometer and detected with a silicon avalanche photodiode. The PL circular polarization degree, $\rho=(I^{++}-I^{+-})/(I^{++}+I^{+-})$, was measured using a photoelastic modulator operated at frequency of 50 kHz and a photon counting system. Here, $I^{++}(I^{+-})$ is the PL intensity for

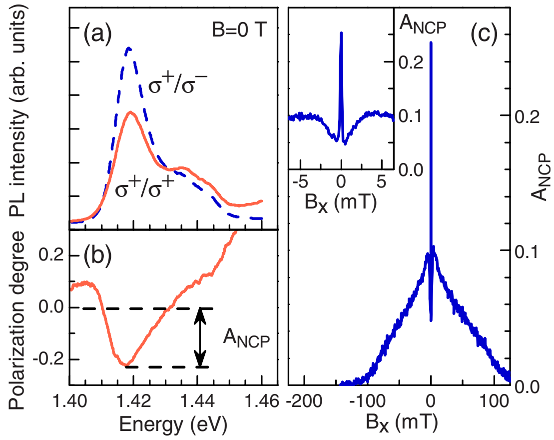

FIG. 1. (Color online) (a) PL spectra of circularly copolarized (solid line) and crosspolarized (dashed line) emission measured for σ^+ polarized excitation. (b) Circular polarization degree extracted from the spectra in panel (a). (c) Hanle curve measured at detection energy 1.418 eV and for excitation density $P_{\mathrm{exc}}=200\ \mathrm{W/cm^2}$. Inset is closeup of central peak. $T=1.6$ K.

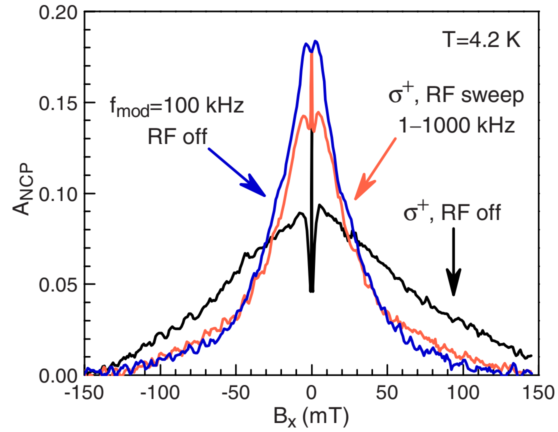

FIG. 2. (Color online) Hanle curves measured for σ^+-polarized excitation in absence (RF off) and presence of RF field applied along z axis with the frequency sweeped in the indicated range. Similar effect is obtained for RF applied along x and y axes. Also, the Hanle curve detected for excitation with polarization modulation at frequency $f_{\mathrm{mod}}=100$ kHz is shown.

copolarization (cross polarization) of excitation and detection for σ^+-polarized excitation. For some experiments, in order to suppress the DNP, the helicity of optical excitation was periodically modulated between σ^+ and σ^- at a frequency of 100 kHz by an electro-optical modulator followed by a quarter-wave plate.

RF fields were applied through a small coil oriented either along the magnetic field axis (x) or along the optical axis (z), or perpendicular to both these directions. Two ODNMR schemes were implemented. In the more common first one the RF-field frequency was fixed and the Hanle curves were measured by scanning the magnetic field. For the second one, the RF-field frequency was swept in the range 1–1000 kHz at a rate of 50 MHz/s, corresponding to hundreds of scans during a several seconds long signal accumulation time at fixed magnetic field.

Polarized PL spectra measured for σ^+ polarized excitation at zero magnetic field are shown in Fig. 1(a). The strongest peak corresponds to trion emission from the QD ground state. Its intensity in cross polarization is larger than in copolarization, resulting in *negative* polarization, as shown in Fig. 1(b). The NCP amplitude, A_{NCP}, is proportional to the mean electron-spin polarization along the z axis.[11,19–21] As the resident electrons are interacting with the QD nuclei, the NCP can be used as indicator of nuclear-spin polarization.

The magnetic field dependence of NCP (Hanle curve) is shown in Fig. 1(c). It consists of a narrow central peak with half-width at half maximum (HWHM) of about 0.07 mT and a wide Lorentz-type pedestal with HWHM of about 50 mT. Narrow dips near the central peak (referred to as W structure[2]) are also observed. This W structure is typically explained by the presence of DNP. It is assigned to the appearance of a DNP component, antiparallel to the external magnetic field, B_x.[2,22] At small, but nonzero, B_x it efficiently depolarizes the electron spin thus leading to the dips. At higher external fields, the electron-spin polarization is partially restored due to vanishing of this component of DNP field. The wide pedestal can be explained by the presence of z component of DNP, which is stabilized by the nuclear quadrupole splitting.[7,23] To verify the presence of DNP in our sample, we applied RF excitation, which directly affects the nuclear-spin system.

The RF-field effect as a whole obtained using the rapid frequency sweep at range 1–1000 kHz is shown in Fig. 2. As seen from the figure, the RF-field application is accompanied by a pronounced change of Hanle curve. The sign of this change depends on the magnetic field strength: it is positive for $|B_x|<20$ mT and negative at higher magnetic fields. Interestingly a similar effect appears when the excitation polarization is modulated while the RF is switched off (see Fig. 2). Such modulation suppresses the DNP.[2] Therefore, both the positive and negative changes in NCP amplitude for sweeping the RF or modulating the polarization are related to DNP.

We suggest that, for magnetic fields exceeding 20 mT, the orientation of electron spin is stabilized by the longitudinal DNP component parallel to the z axis. The suppression of nuclear polarization by the RF field results in a decrease in electron-spin polarization as observed experimentally. The reason for presence of this DNP component is the nuclear quadrupole splitting[7] for nuclear spins $I \geq 3/2$. The origin of this splitting is an electric field gradient, $\nabla \boldsymbol{F}$, which splits the nuclear-spin states into Kramers doublets $|\pm m/2\rangle$ with different moduli of the spin projection m onto $\nabla \boldsymbol{F}$.[24]

In presence of quadrupole interaction, the Zeeman splitting of the states $|\pm m/2\rangle$, $m \geq 3$ in a magnetic field, $\boldsymbol{B}$, normal to the principal axis of $\nabla \boldsymbol{F}$ is suppressed at small B.[24] Simultaneously, the spin depolarization is suppressed.[7] As a result, the nuclear field created by polarization of these states is directed along the electric field gradient rather than the external magnetic field. A RF-field resonant to the quadrupole splitting destroys the nuclear polarization and, in that way, suppresses the electron-spin polarization. This is the reason for the negative RF-field effect in large magnetic fields in Fig. 2.

The main origin of the electric field gradient in the studied

TABLE I. Nuclear parameters. The values of S_{11}, Q, and γ are taken from Refs. 27, 29, and 30, respectively. The values of ν_Q are calculated for strain ε_{zz}=0.01.

Isotope	I	γ [rad/(Ts)]	Q (mbar)	S_{11} (statC/cm^3)	ν_Q (kHz)
^{69}Ga	3/2	6.439×10^7	171	9.1×10^{15}	564
^{71}Ga	3/2	8.181×10^7	107	9.1×10^{15}	353
^{75}As	3/2	4.596×10^7	314	1.31×10^{16}	1490
^{113}In	9/2	5.885×10^7	759	1.67×10^{16}	388
^{115}In	9/2	5.897×10^7	770	1.67×10^{16}	383

QDs is the strain caused by the lattice mismatch between (In,Ga)As and GaAs.[25] The magnitude of this strain-induced quadrupole splitting is given by[26] $h\nu_Q=3eQV_{zz}/[2I(2I-1)]$, where h, e, Q, and I are Planck constant, electron charge, nuclear quadrupole moment, and nuclear spin, respectively. Here, the nuclear quadrupole frequency, ν_Q, is proportional to the strain-induced electric field gradient at the nuclear site, $V_{zz}=S_{11}\varepsilon_{zz}$, which is determined by the strain ε_{zz}; S_{11} is the principal component of the so-called S tensor, which can be extracted from nuclear acoustic resonance.[27] Using a cylindrically symmetric QD model[12] and the transversal isotropic approximation[28] in continuum elasticity theory, we estimated ε_{zz} to be on the order of a few percent. As an example, the ν_Q values for the Ga, As, and In nuclei calculated for ε_{zz}=0.01 are listed in Table I. So, the quadrupole splittings, $h\nu_Q$, are sufficiently large being comparable to the Zeeman splittings, $\gamma\hbar B$ (here γ is the nuclear gyromagnetic ratio, see Table I), which can be obtained in magnetic fields ranging from tens to hundreds of millitesla. In this range of magnetic field we expect DNP stabilization and its gradual suppression with increasing magnetic field.

The positive effect within the W range of the Hanle curve (Fig. 2) indicates that the nuclear polarization suppressed by the RF field or the modulated polarization of excitation is orthogonal to the photocreated electron-spin polarization.[2,22] To identify the types of nuclei, which create this polarization, we studied Hanle curves at a fixed RF frequency (see Fig. 3). The application of RF field is accompanied by the appearance of clearly observed resonances which are positioned symmetric to the central peak. Although the resonances are relatively wide (HWHM of about 0.5 mT), their magnetic field position can be extracted from experiment.

The applied frequencies of the RF field are much smaller than the quadrupole frequencies ν_Q. This means that the observed resonances can be caused by transitions between the states split by magnetic field. These can be the $|\pm 1/2\rangle$ states only because the Zeeman splitting of the other states ($|\pm m/2\rangle$ with $m\geq 3$) is suppressed by the quadrupole effect. The splitting of the $|\pm 1/2\rangle$ states, on the contrary, is enlarged by the quadrupole interaction.[24] The Hamiltonian describing Zeeman splitting of nuclear states in the presence of quadrupole interaction is[24,26]

$$\hat{\mathcal{H}} = -\gamma\hbar(\hat{\boldsymbol{I}}\cdot\boldsymbol{B}) + h\nu_Q[\hat{I}_z^2 - I(I+1)/3]/2. \qquad (1)$$

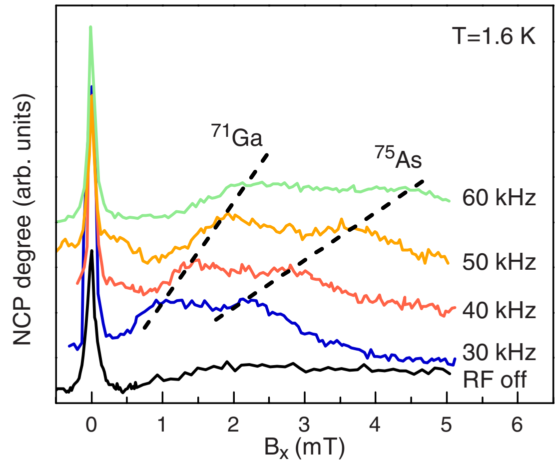

FIG. 3. (Color online) Effect of RF field applied along y axis on Hanle curve in the range of W shape. RF frequencies are given at each curve. The curves are shifted vertically for clarity of presentation. Dashed lines indicate the magnetic field position of NMR.

The dependencies of Zeeman splitting of the spin states for Ga, As, and In nuclei in a magnetic field perpendicular to the quadrupole axis, were calculated by direct diagonalization of Hamiltonian (1). The splitting for ^{71}Ga is shown in Fig. 4(a) as an example. For weak magnetic field of a few millitesla, the splitting of the $|\pm 3/2\rangle$ states is negligibly small whereas the splitting of the $|\pm 1/2\rangle$ states is twice larger than in absence of the quadrupole interaction. A similar behavior of Zeeman splitting is obtained for the other types of nuclei.

The calculations allow us to assign the resonances in Fig. 3 to RF-induced transitions between the $|\pm 1/2\rangle$ states in the ^{71}Ga and ^{75}As nuclei. As seen from Fig. 4(b), the resonance field dependences are well described by the calculations in-

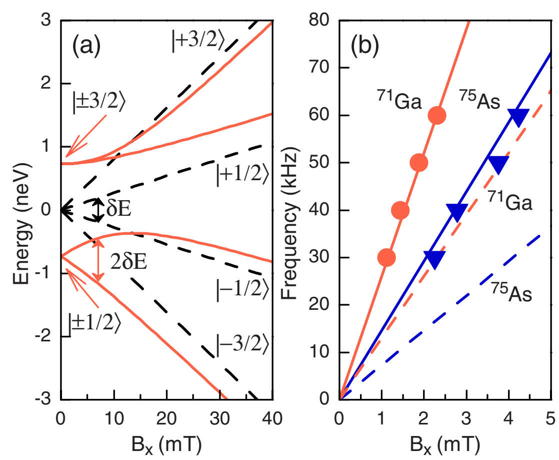

FIG. 4. (Color online) (a) Zeeman splitting of spin states of ^{71}Ga in magnetic field normal to the quadrupole axis (solid lines) calculated for ε_{zz}=0.01. Dashed lines show splitting in absence of quadrupole interaction. Arrows indicate NMRs for both cases. (b) Magnetic field dependences of resonant frequencies extracted from experiment (symbols). The dependences of NMRs for $|\pm 1/2\rangle$ states in ^{71}Ga and ^{75}As nuclei calculated with and without quadrupole interaction are shown by the solid and dashed lines, respectively.

cluding the quadrupole splitting. The calculations also show that the Zeeman splitting depends on the mutual orientation of the quadrupole and magnetic field axes rather than the quadrupole interaction strength as long as it is strong enough. Therefore, the main source of the observed broadening of the resonances is most likely related to the spread of the quadrupole axis directions in the dot ensemble.

For In nuclei with $I=9/2$ the Zeeman splitting of the $|\pm 1/2\rangle$ states is considerably larger than for other QD nuclei having $I=3/2$. As a result, the respective resonances are shifted toward zero magnetic field, and are strongly broadened, which makes them hard to resolve. Moreover, due to the larger number of spin states in In nuclei, the population of states $|\pm 1/2\rangle$ is 2.5 times smaller than for Ga and As nuclei at the same level of spin polarization. We believe that due to these reasons, we were not able to observe the In resonances. The RF resonances for the ^{69}Ga nuclei were also not observed, possibly because the, receptivity, of the ^{69}Ga nuclei to the RF field is twice smaller than that for the ^{71}Ga nuclei.[30]

Summarizing, we have demonstrated that due to the quadrupole interaction, which is relevant for many QD systems, the nuclear-spin states $|\pm 1/2\rangle$ and $|\pm m/2\rangle$ with $m \geq 3$ behave as independent nuclear subsystems, when the quadrupole splitting is considerably larger than the Zeeman splitting. The subsystems are responsible for different components of nuclear field, resulting in distinct features in experiment. In particular, the positive RF-field effect at small magnetic fields is due to suppression of nuclear field created by polarization of the $|\pm 1/2\rangle$ states whereas the negative effect at larger magnetic fields results from RF-induced depolarization of the $|\pm m/2\rangle$ states with $m \geq 3$. The negative effect appears when the RF sweep range includes the quadrupole frequencies of the polarized nuclei.

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