Anomalous Hanle curves induced by in-plane nuclear field in single self-assembled InAlAs and InAs nanostructures

S. Yamamoto¹, R. Matsusaki¹, R. Kaji¹, and S. Adachi¹
¹ Department of Applied Physics, Hokkaido University, Sapporo 060-8628, Japan
E-mail: yama-st@eis.hokudai.ac.jp

Abstract. We studied the formation mechanism of in-plane nuclear field by anomalous Hanle effect measurements in single self-assembled In₀.₇₅Al₀.₂₅As/Al₀.₃Ga₀.₇As quantum dots and InAs/GaAs quantum rings. The observed anomalous Hanle curves indicated quite large width and hysteretic behavior to the externally applied magnetic field. These anomalies appears in both materials whose electron g factors have opposite sign each other, and cannot be reproduced by a traditional spin dynamics model. In this study, we show that a proposed spin dynamics model can explain the formation of a large in-plane nuclear field regardless of the sign of electron g factor. The model is based on the dynamic nuclear spin polarization mechanism including the effects of nuclear quadrupole interaction.

1. Introduction

In the semiconductor nanostructures, the photon angular momentum can be transferred onto the lattice nuclei via the localized electron spin by hyperfine interaction (HFI), and thus a large nuclear spin polarization (NSP) can be generated [1, 2]. The NSP affects an electron spin as a nuclear field (Overhauser field) $B_n$, and conversely, the electron spin acts on the lattice nuclei as a Knight field $B_e$. This reciprocal interplay complicates the spin physics in semiconductor nanostructures. Therefore, the engineering of NSP is required for the realization of potential applications such as spin storage and optical manipulation of a confined electron spin.

Because HFI induces simultaneous flip-flop of the electron and nuclear spins generally [1], it has been believed that the direction of NSP, hence the direction of $B_n$, is only parallel to the injected electron spin. However, recent experiments revealed that the large in-plane nuclear field $B_{n,x}$ which is perpendicular to the photo-injected electron spin can be generated under the external in-plane magnetic field $B_x$ [3, 4]. Further, an independent experiment with single strain-free quantum dot (QD) suggested that the formation of $B_{n,x}$ is closely associated with quadrupole interaction (QI) [5], which is the coupling between an electric quadrupole moment of nucleus with spin $I>1/2$ and the electric field gradient caused by residual strain and/or random alloying in the self-assembled (SA) nanostructures.

Although the model of $B_{n,x}$ formation was proposed recently and the qualitative agreement has been achieved [6], further experimental and theoretical investigations should be carried out for complete understanding of the QI effects. In this paper, we compare the $B_{n,x}$ generated in two different materials where the sign of electron g factor $g_e$, one of the most crucial parameters for the direction of $B_n$, is opposite each other.
2. Sample and Experimental Setup

Single SA In$_{0.75}$Al$_{0.25}$As/Al$_{0.3}$Ga$_{0.7}$As QDs and SA InAs/GaAs quantum rings (QRs) grown by molecular beam epitaxy on (100)-GaAs substrate were used in this study. From the independent experiments, it was confirmed that the sign of $g_e$ of the conduction band in InAlAs QDs and InAs QRs are positive and negative respectively [7]. After fabrication of small mesa structures, time-integrated micro-photoluminescence ($\mu$-PL) measurements under $B_x$ up to 1 T were carried out at 6 K. Typical PL spectra of both single nanostructures are shown in Fig. 1. In this paper, we focus on the positively charged exciton $X^+$ which showed no fine-structure splitting. This is because the degree of circular polarization (DCP) $c$ of $X^+$ PL is determined essentially by $z$ component of the electron spin polarization $\langle S_z \rangle$ through the relation

$$ c = 2 \langle S_z \rangle.$$

Here, the DCP is defined as $c = (I^+ - I^-)/(I^+ + I^-)$ where $I^{+(-)}$ is the $\sigma^+(\sigma^-)$ polarized PL intensity. A continuous wave Ti:Sapphire laser was used for excitation to the tail of the wetting layer. The corresponding wavelengths were $\sim$730 nm for InAlAs QDs and $\sim$865 nm for InAs QRs. The excitation light was adjusted to the circularly polarized one by combination of a linear polarizer, a half waveplate, and a quarter waveplate. The simultaneous acquisition of the $I^+$ and $I^-$ PL components was achieved with a beam displacer. Each displaced PL component is focused on a different detection area of the CCD, and then, DCP can be acquired by a single exposure process in highly accurate manner. Details of the apparatus are found in Ref. [8].

![Figure 1. PL spectra of single InAlAs QD and single InAs QR at 6 K and 0 T.](image)

3. Hanle effect measurements

As is well known, the precession of the electron spin $\langle S \rangle$ due to a magnetic field in Voigt geometry is described by Bloch equation. The steady-state $\langle S_z \rangle$ decays with increasing $B_x$, and the depolarization curve (Hanle curve) shows a Lorentzian function of $B_x$ if $\langle B_n \rangle = 0$. Further, the shape of Hanle curve is modified reflecting the direction and the magnitude of $B_n$ induced by HFI [1]. In the traditional model, $B_n$ induced by nuclear spin cooling due to $B_e$ is written as

$$ B_n = b_n[(B_T^{(n)} \cdot \langle S \rangle)B_T^{(n)}]/(|B_T^{(n)}|^2 + \xi B_e^2),$$

where $B_T^{(n)}$ is a sum of externally applied field $B_{ext}$ and $B_e$, $B_L$ is a dipolar field among neighboring nuclei, and $\xi=2-3$ is a linkage coefficient. The factor $b_n$ is a material-dependent parameter which is proportional to $1/g_e$. Figure 2(a) shows $\langle S_z \rangle$ calculated with the traditional model. The upper (lower) panel corresponds to the case of negative (positive) $g_e$. In the figure, the dashed lines indicate Lorentzian shaped Hanle curves. The normal Hanle curve is deformed only in the narrow $B_x$ region and the deformation is different depending on the sign of $g_e$. Former is related to the magnitude of $B_e (~a few mT)$, and latter can be explained by considering that the direction of $B_{n,x}$ depends on the sign of $g_e$ through $b_n$ in Eq. (1); namely amplification (compensation) of $B_x$ occurs when $g_e$ is negative (positive).

Figure 2(b) and (c) show the observed anomalous Hanle curves in InAlAs QD and InAs QR, respectively. The dashed curves are normal Lorentzian ones expected from the spin lifetime $T_s=0.5$ ns and $g_e=+0.35 (-0.42)$ for InAlAs QD (InAs QR). It is obvious that the observed
Figure 2. (a) The calculated Hanle curves by traditional theory (upper: $g_e<0$, lower: $g_e>0$). The dashed line shows Lorentzian shaped Hanle curve without $B_n$ and the pairs of arrows show the direction of $B_x$ and $B_{nx}$ schematically. (b), (c) The observed anomalous Hanle curves in InAlAs QD and InAs QR at 6 K. The dashed line shows Lorentzian shaped Hanle curves. (d), (e) The calculated anomalous Hanle curves with $g_e=+0.35$ in (c) for InAlAs and $g_e=-0.42$ in (d) for InAs QR. (f) The calculated anomalous Hanle curves with the tilted $B_x$ (top), with the normal condition (middle), and with the tilted QI axis (bottom). Top insets are the configuration of the principal axis of QI ($q$) and $B_x$.

Curves deviate significantly from the Lorentzian and have quite large width despite the opposite sign of $g_e$. Further, the critical fields $B_c$ at which $\langle S_z \rangle$ changes abruptly are seen in both curves and the value of $B_c$ depends on the sweep direction of $B_x$ (i.e. hysteresis). Highly-preserved $\langle S_z \rangle$ suggests the compensation of $B_x$ by $B_{nx}$, and the hysteresis originates from the bistability of NSP, hence $B_n$. Similar compensation of external field by $B_n$ has been well-studied in $z$-direction [2]. Compared with the traditional theory, the most curious point is that $B_{nx}$ was always anti-parallel to $B_x$ regardless of the sign of $g_e$.

Figure 2(d) and (e) show the computed $\langle S_z \rangle$ according to the model of $B_{nx}$ formation which was proposed in our previous study [6]. Both results reproduce the above-mentioned anomalies qualitatively, and therefore, the model proves to explain well the formation of $B_{nx}$ in spite of the opposite sign of $g_e$. Our model is based on the dynamic nuclear spin polarization where the dynamics of NSP $\langle I_k \rangle$ along $k=x, y, z$ direction can be written as

$$\frac{d\langle I_k \rangle}{dt} = \frac{1}{T_{NF,k}} [Q(S_k - S_{eq,k}) - \langle I_k \rangle] - \frac{1}{T_{ND,k}} \langle I_k \rangle,$$

where $1/T_{NF,k}, 1/T_{ND,k}, S_k$, and $S_{eq,k}$ are $k$ components of formation and relaxation rate of NSP, electron spin polarization, and thermal equilibrium value of $S_k$, respectively. $Q$ is a momentum conversion coefficient from electron to nuclei. Note that, $1/T_{NF,k}$ includes $\langle I_k \rangle$ self-consistently and thus NSP shows nonlinear response such as bistability [2].

In addition, the effects of QI have to be considered. The introduced effects into our model are (i) suppression of NSP precession and (ii) sign inversion between out-of- and in-plane nuclear g factor $g_n$. The first QI effect (i) originates from unequally-spaced splitting of nuclear energy levels [quadrupolar splitting (left inset of Fig. 3)]. As seen in Fig. 3, the spin pairs with $|I_z| > 1/2$ indicate zero or quite small splittings. Namely, the effective $g_n$ decreases, and thus the precession of NSP is suppressed even under a large $B_x$. The second QI effect (ii) is supposed to arise from nuclear spin state-mixing induced by the combination of QI and Zeeman splitting due to $B_x$. Since the direction of $B_{nx}$ depends on $g_{nx,k}$ as well as $g_e$, the sign inversion between $g_{nx,x}(y)$ and $g_{nx,z}$ could realize the $B_{nx}$ formation regardless of the sign of $g_e$. Note that $g_e$ is nearly isotropic because of the symmetrical nature of conduction band.
Finally, we should comment about the asymmetry of the observed curve in Fig. 2(c) while the corresponding calculation [Fig. 2(e)] is symmetrical. The possible origin of the asymmetry is inclination of the principal axis $q$ of QI from the crystal growth axis ($z$ axis) and/or deviation of external magnetic field from the sample plane ($x$-$y$ plane). In general, $q$ is determined by strain axis and lies along almost parallel to $z$ axis in SA nanostructures. Also, misalignment of the apparatus could make the external field deviated from $x$-$y$ plane. Figure 2(f) shows the computed $\langle S_z \rangle$ with a tilted $q$ by 10 degrees (red), with a tilted $B_x$ by 5 degrees (blue) and with the normal condition (middle) where $B_x$ and $q$ point strictly $x$ and $z$ axes respectively as with Fig. 2(e). The respective configurations are schematically depicted in the insets. Both cases induce the asymmetry in Hanle curve, but there are some differences. The inclination of $q$ induces a horizontal shift of whole curves, while the tilting of $B_x$ induces horizontal shifts of the inner and outer $B_c$ in the opposite direction each other, and thus the hysteresis width becomes different depending on the sign of $B_x$. Therefore, it is likely that the observed asymmetry in Fig. 2(c) comes from the inclination of $B_x$ due to the misalignment of the sample mounting.

4. Summary
We observed the anomalous Hanle curves in InAlAs QDs and InAs QRs, which indicated the formation of considerable $B_{n,x}$ regardless of the sign of $g_e$. The observed anomalies in the curves could not be explained by the traditional theory, and we indicated that our model of $B_{n,x}$ formation could reproduce experimental results qualitatively. In the model, the strain-induced QI plays a key role to generate $B_{n,x}$. Moreover, the tilting effect of $B_x$ and QI axis which induced the asymmetry to the curves was discussed.

Acknowledgments
This work is supported in part by JSPS KAKENHI (Grant No. JP26800162) and the Asahi Glass Foundation (Japan).

References
[1] Fleisher V G and Merkulov I A 1984 Optical Orientation (Modern Problems in Condensed Matter Sciences) vol 8 ed F P Meier and B P Zakharchenya (North-Holland: Elsevier Science Publishers B.V.) chaps. 2 and 5
[2] Recent optical investigation of nuclear spin physics in QDs are reviewed comprehensively: Urbaszek B, Marie X, Amand T, Krebs O, Voisin P, Maletinsky P, Hogaé A and Imamoglu A 2013 Rev. Mod. Phys. 85 79
[3] Krebs O, Maletinsky P, Amand T, Urbaszek B, Lemaitre A, Voisin P, Marie X and Imamoglu A 2010 Phys. Rev. Lett. 104 056603
[4] Nilsson J, Bouet L, Bennett A J, Amand T, Stevenson R M, Farrer I, Ritchie D A, Kunz S, Marie X, Shields A J and Urbaszek B 2013 Phys. Rev. B 88 085306
[5] Sallen G, Kunz S, Amand T, Bouet L, Kuroda T, Mano T, Paget D, Krebs O, Marie X, Sakoda K and Urbaszek B 2014 Nat. Commun. 5 3268
[6] Yamamoto S, Matsusaki R, Kaji R and Adachi S 2018 Phys. Rev. B 97 075309
[7] Matsusaki R, Kaji R, Yamamoto S and Adachi S 2018 Appl. Phys. Lett. (submitted)
[8] Matsusaki R, Kaji R, Yamamoto S, Sasakura H and Adachi S 2018 Appl. Phys. Express (in press)