Nuclear spin effects in negatively charged InP quantum dots

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Abstract. Effects of both the dynamic nuclear polarization (DNP) created by circularly polarized light and the fluctuations of average nuclear spin in a quantum dot (QD) on the electron spin orientation are studied for singly negatively charged InP QDs. From the dependence of the negative circular polarization of photoluminescence on the applied longitudinal magnetic field, the hyperfine field $B_N$ of a few mT appearing due to DNP and the effective magnetic field $B_{\text{eff}}$ of a few tens of mT arising from nuclear spin fluctuations (NSF) are estimated. A lifetime of about 1 µs is estimated for NSF.

Strong localization of electrons in quantum dots (QDs) may enhance hyperfine interaction of electron spins with those of nuclei [1]. Various aspects of the hyperfine interaction of electron and nuclear spins have been studied for last three decades in different materials [2], including InP QDs [3]. Charge-tunable InP QDs with one resident electron per QD, on an average, have recently attracted considerable research interests due to the observation of millisecond range spin lifetime of resident electrons in these QDs [4, 5]. This observation makes it a promising candidate for quantum memory element in the emerging fields of quantum information technology and spintronics [6]. However, the influence of the hyperfine interaction between electron and nuclear spins on the long-lived electron spin orientation needs to be clarified.

Two effects of the electron-nuclear spin-spin interactions are possible. One of them is the so-called dynamic nuclear polarization (DNP). In the optical orientation of electron spins, the spin-polarized electrons dynamically polarize the nuclear spins due to the hyperfine coupling of the electron and nuclear spin subsystems [2]. In turn, the spin polarized nuclei produce an internal magnetic field $B_N$, which may influence electron spin dynamics. In presence of an externally applied magnetic field $B_{\text{ext}}$, electron spin subsystem should feel an effective magnetic field $B_{\text{eff}} = B_{\text{ext}} + B_N$.

Another effect arises from the nuclear spin fluctuations (NSF). Due to limited number of nuclear spins, typically $n \sim 10^5$, interacting with the electron spin in a QD, random correlation of nuclear spins may create a fluctuating nuclear polarization, $\Delta S_N \propto S_N / \sqrt{n}$, where $S_N$ is the total spin of the polarized nuclei. Fluctuation $\Delta S_N$ acts on the electron spin subsystem as another internal magnetic field, $B_f$, with random magnitude and orientation [7]. Electron spin precession in this field results in the dephasing of electron spins in the QD ensemble and in the three-fold decrease in magnitude of the total electron spin polarization [7, 8].

In the present paper we describe our experimental study of nuclear spin effects on long-lived spin polarization of resident electrons observed recently [4, 5] in singly negatively charged InP QDs.

The sample consists of a single layer of self-assembled InP QDs embedded between GaInP barriers grown on a $n^+$-GaAs substrate. The average base diameter (height) of the QDs is about 40 (5) nm with an areal density of about $10^{10}$ cm$^{-2}$. Semi-transparent indium-tin-oxide electrode was deposited on top of the sample to control the charge state of the dots by means of applied electric bias [4, 5]. For the present study on singly negatively charged QDs we apply an electric bias of $U_b = -0.1$ V, as it was found from a previous study of trionic quantum beats [9] on the same sample that at this bias the QDs contain one resident electron per dot (on an average).

Electron spins in the QD ensemble were polarized in our experiments by using the well-known optical orientation technique [2, 4, 5]. A negative circular polarization of the trionic (negatively charged exciton) photoluminescence (PL) with absolute value up to a few tens of percentage was observed for such QDs under quasi-resonant excitation and was interpreted as a result of long-lived (hundreds of microseconds) spin memory of resident electrons [4, 5]. We monitor the degree of circular polarization $p_{\text{circ}} = (I^+ - I^-) / (I^+ + I^-)$, where $I^{+(-)}$ is the PL intensity for $\sigma^+$ excitation and detection of $\sigma^{+(-)}$ PL, as a function of an external magnetic field $B_{\text{ext}}$ applied along the optical excitation axis (longitudinal magnetic field, Faraday geometry). The internal mag-
netic field $B_1$ is expected to cause a three-fold reduction in the electron spin polarization (and correspondingly in $\rho_{\text{circ}}$) when $B_{\text{eff}} = B_{\text{ext}} + B_N \approx 0$. Influence of $B_1$ may be suppressed by applying a longitudinal magnetic field exceeding $B_1$ [7, 8]. A clear demonstration of this effect is seen in Fig. 1, where $\rho_{\text{circ}}$ is plotted as a function of $B_{\text{ext}}$ for $\sigma^+$ and $\sigma^-$ excitations. For $B_{\text{ext}} > 50$ mT, $\rho_{\text{circ}}$ approaches a constant value and a decrease from this constant value is seen for $B_{\text{ext}}$ near by, but not exactly zero. The behavior of $\rho_{\text{circ}}$ as a function of $B_{\text{ext}}$ is fitted with a Lorentzian, full width at half maximum of which estimates $B_1 \approx 15$ mT. The shift of the Lorentzian from $B_{\text{ext}} = 0$ is due to $B_N$. The sign of $B_N$ created by light should be opposite for the two counter circular polarizations ($\sigma^+$ and $\sigma^-$) of the excitation beam. As a result, the values of $B_{\text{ext}}$ corresponding to $B_{\text{eff}} = B_{\text{ext}} + B_N \approx 0$ for $\sigma^+$ and $\sigma^-$ excitations differ by $2B_N$ [Fig. 1]. A plot of $B_N$ as a function of $B_{\text{ext}}$ fits a product of $\rho_\ast \cdot P_\ast \approx (1 - 1)$ for $\sigma^+$ ($\sigma^-$) excitation and $P_\ast$ (the excitation power) shows that DNP builds up linearly with the excitation laser power (Fig. 1). At high excitation power of about 50 mW a rather small dynamic nuclear magnetic field of about 6 mT is seen, in agreement with a previous report [3].

Theoretically proposed three-fold decrease of the electron spin polarization [7] due to hyperfine interaction with “frozen” NSF was not observed experimentally in case of very weak optical pumping of the InP QDs. This is related to a variation of the amplitude and orientation of the NSF in time (finite lifetime of NSF) which is able to totally destroy the electron spin polarization if the pump photons rarely come and restore the polarization. To estimate the lifetime of NSF we perform a time-domain measurement by using PL pump-probe technique [4, 5] shown schematically in the left inset of Fig. 2. A circularly ($\sigma^+$ or $\sigma^-$) polarized strong pump pulse creates spin orientation of the resident electrons [5]. The electron spin orientation is then probed by measuring $\Delta \rho$, the difference of $\rho_{\text{circ}}$ of the probe PL for the $\sigma^+$ and $\sigma^-$ pump, as a function of the pump-probe delay ($\tau$). Figure 2 plots $\Delta \rho$ as a function of $\tau$ for different values of $B_{\text{ext}}$. The spin decay time $\tau_d$ is estimated from a fit $\Delta \rho = A \exp(-\tau/\tau_d) + B$ to these data. A plot of $\tau_d$ as a function of $B_{\text{eff}}$ is shown in the right inset of Fig. 2. We consider that near the minimum where $B_{\text{eff}} \approx 0$, electron spin dynamics is ruled by the NSF lifetime, which is estimated to be about 1 $\mu$s from our data.

In conclusion, the effect of nuclear spins on electron spin dynamics in InP QDs is studied. The values of NSF lifetime and of hyperfine fields $B_N$ and $B_1$ appearing due to DNP and due to NSF are estimated.

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