Electron and hole spin relaxation in InP-based self-assembled quantum dots emitting at telecom wavelengths

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We investigate the electron and hole spin relaxation in an ensemble of self-assembled InAs/In0.53Al0.24Ga0.23As/InP quantum dots with emission wavelengths around 1.5 µm by pump-probe Faraday rotation spectroscopy. Electron spin dephasing is due to randomly oriented nuclear Overhauser fields with strength of ~ 30 mT is observed. At low temperatures we find sub-microsecond longitudinal electron spin relaxation time $T_1$ which unexpectedly strongly depends on temperature. At high temperatures the electron spin relaxation time is limited by optical phonon scattering through spin-orbit interaction decreasing down to 0.1 ns at 260 K. We show that the hole spin relaxation is activated much more effectively by a temperature increase compared to the electrons.

Semiconductor quantum dots (QDs) offer a promising platform for quantum information technologies [1]. An electron spin in a QD, often considered as a candidate for a quantum bit (Qubit), can be efficiently manipulated by light pulse which gives the possibility of easy integration into existing optical telecommunication networks. In this respect QDs emitting in the telecom spectral range (1.3-1.6 µm) are especially attractive [2-12]. In particular, potential applications as laser active medium [7-8], single-photon emitters [13-21], and polarization-entangled photons emitters [22] are already envisaged.

While for III-V QDs emitting in the technologically and spectroscopically easily accessible wavelength range below 1 µm the spin properties have been intensively studied in recent decades [22-31], information on the spin dynamics of QDs emitting at longer wavelengths, in particular in the telecom spectral range, is limited. So far, the electron and hole g factors [32] with record-high anisotropies [33-35] were measured. The dynamics of the photoluminescence polarization degree related to the exciton spin dynamics was measured as well [36].

In this paper we address the spin lifetimes of carriers in InAs/In0.53Al0.24Ga0.23As/InP QDs emitting at telecom wavelengths which have not been measured so far to the best of our knowledge. At weak magnetic fields, the spin dynamics of the resident electrons in the QDs is governed by the hyperfine interaction with the nuclei and the regime described theoretically in Ref. [40] is observed. In increased longitudinal magnetic fields, at low temperatures we observe a sub-microsecond decay of spin polarization. With increasing temperature we observe dramatic decrease of both $T_1$ and $T_2^*$ which at high temperatures is mediated by the electron interaction with LO phonons.

The QD sample was grown by molecular-beam epitaxy on an (100)-oriented InP substrate. The nominally undoped QDs were formed from 5.5 monolayers of InAs sandwiched between In0.53Al0.24Ga0.23As barriers. The optically active QDs have a diameter of ~ 50 nm and a height of ~ 10 nm; their density is about $10^{10}$ cm$^{-2}$.

The sample is held at temperatures in the range 5-260 K in a helium bath cryostat with a split-coil superconducting magnet. Magnetic fields up to $B = 2$ T are applied either in Faraday (parallel to the light propagation direction, coinciding with the sample growth axis) or in Voigt (perpendicular to the the light propagation direction) geometry. Pump-probe Faraday rotation is employed to study the carriers spin dynamics in the QDs.

Two laser systems are used. The first one consists of a mode-locked Yb:KGW laser pumping an optical parametric amplifier and has the pulse repetition frequency of 40 MHz (repetition period $T_R = 25$ ns). The second laser system is composed of a pulsed Ti:Saphire laser pumping an optical parametric oscillator (OPO) and has pulse repetition rate of 76 MHz ($T_R = 13$ ns). The spectral width of the laser systems shaped below 20 nm and centered at 1520 nm matching the QDs luminescence spectrum [37, 38]. The pulse duration for both systems is less than 2 ps.

The laser beam is split into the pump and the probe. The pump pulses are circularly polarized and create carrier spin polarization in the QDs. The carriers’ spin polarization is analysed by measuring the Faraday ellipticity of the initially linearly polarized probe pulses after transmission through the sample [41]. Varying the time delay between the pump and probe pulses by a mechanical delay line gives the time dependence of the spin polarization. The polarization of the pump beam is modulated between $\sigma^+$ and $\sigma^-$ by a photo-elastic modulator for synchronous detection.

Figure 1(a) shows the dynamics of the Faraday ellipticity signal for the magnetic field applied in Voigt ($B_V$)
and the hole spin precession with final dynamics in Fig. 1(a), corresponding to the electron
Two precession frequencies show up in the ellipticity signal
and Faraday ($B_\parallel$) geometries. In the Voigt geometry, the electron and hole spins precess around the magnetic field direction and the ellipticity signal oscillates accordingly. Two precession frequencies show up in the ellipticity signal dynamics in Fig. 1(a), corresponding to the electron and hole spin precession with $g$ factors $|g_e| = 1.7$ and $|g_h| = 0.7$, respectively. The dephasing time $T_2^*$ of each of the carrier spin polarizations is determined by the random nuclear Overhauser fields $\bm{B}_N$ (which act on the carrier spins via the hyperfine interaction) if the external magnetic field $B_V$ is smaller than $B_N$. At higher $B_V$, on the other hand, the time $T_2^*$ is determined by the spread of the $g$-factors $\delta g$ in the QD ensemble [23]:

$$1/T_2^* \approx |g| \mu_B B_N / \hbar, \quad B_V \lesssim B_N. \quad (1a)$$

$$1/T_2^* \approx \delta g \mu_B B_V / \hbar, \quad B_V \gg B_N. \quad (1b)$$

Therefore, as it was shown for the same QDs in Refs. [35, 36], an increase in $B_V$ causes a decrease of the signal decay time. Note, that the signal is mostly contributed by carriers forming excitons in the QDs rather than from resident carriers since the majority of QDs are charge-neutral. For this reason, $T_2^*$ is contributed (and limited) by the exciton lifetime of $\sim 0.8$ ns [35].

In a longitudinal magnetic field $B_F$, the spin polarization decays monotonically without oscillations [36, 42]. In high enough $B_F$, this decay is characterized by a fast component with decay time 0.5 ns, close to the exciton lifetime, and a slow component of somewhat smaller amplitude. The existence of the long-living component indicates the presence of resident charge carriers in a fraction of the QD ensemble. The decay of the slow component is determined by the longitudinal spin relaxation time $T_1$, which at low temperatures exceeds the period between the laser pulses $T_R$. This leads to an accumulation of spin polarization, and a signal offset appears at negative pump-probe delays, which can be identified in the Faraday geometry data in Fig. 1(a). Note that this offset is absent in Voigt geometry.

The effects of longitudinal and transverse magnetic fields on the carrier spin polarization at a small negative delay $\Delta t = -0.15$ ns (which is equivalent to a large delay $t \approx T_R$ after the previous laser pulse) are presented in Fig. 1(b), see the black and red lines, respectively. When the longitudinal magnetic field is scanned, the signal has a minimum at $B_F = 0$ and this type of measurement gives a polarization recovery curve (PRC). At high enough longitudinal magnetic field, the PRC signal is mainly determined by the longitudinal spin relaxation time $T_1$.

At zero field the decay of the total spin polarization is governed by the nuclear fields. For an arbitrary QD the direction of the total nuclear Overhauser field $\bm{B}_N$ acting on an electron (a hole) spin is random. The electron spin component perpendicular to $\bm{B}_N$ precesses around $\bm{B}_N$. When precessing, this spin component is averaged over all QDs, and it decays on a short timescale given by Eq. (1a). On the other hand, the electron spin component along $\bm{B}_N$ decays during the much longer time $T_1$. Averaging over all QDs, and, thus, over all directions of $\bm{B}_N$, shows that the nonprecessing component amounts to 1/3 of the initial electron spin polarization [32, 33, 40]. This explains the drop at $B_F = 0$ in the PRC curve [Fig. 1(b)]. The fact that the spin polarization at $B_F = 0$ is smaller than 1/3 of its value at high longitudinal magnetic fields (it amounts to $\sim 1/8$ only) may be related to the anisotropy of $\bm{B}_N$, to the increase of $T_1$ with $B_F$, etc. The half-width of the dip gives the value of the nuclear fields, $B_N \approx 30$ mT.

When the transverse magnetic field $B_V$ is increased, it contributes to the nuclear field and the nonprecessing spin polarization (directed along $\bm{B}_V \times \bm{B}_N$) have vanishing projection on the probe beam direction. This leads to the decrease of the ellipticity signal at negative pump-probe delays with increasing $B_V$ [Fig. 1(b), red line]. The half-width of the zero-field peak in this curve gives the value $B_N \approx 30$ mT, the same as in PRC. Note that there are no resonant spin amplification peaks at nonzero $B_V$ due to the short transverse dephasing time $T_2^*$ [see Eqs. (1a, 1b)]. Another feature characteristic for an electron spin subject to nuclear fields is the non-monotonict
decay of the spin polarization in zero external magnetic field, showing a local minimum at the time which can be estimated from Eq. (1). This agrees with our observations at $t \approx 1.8$ ns [see the inset in Fig. 1(a)], despite the smaller amplitude of the minimum.

$$S(t) = P_0 T_1 + A \cos(2\pi ft - \phi),$$  \hspace{1cm} (2)

$$A = \frac{P_0 T_1}{\sqrt{1 + (2\pi T_1 f)^2}},$$  \hspace{1cm} (3)

$$\tan(\phi) = 2\pi T_1 f.$$  \hspace{1cm} (4)

Thus, with increasing $f$, the modulation of the spin polarization decreases in amplitude $A$ [Eq. (3)] and becomes retarded relative to the pump modulation by the phase $\phi$ [Eq. (4)]. By performing synchronous detection on the pumping frequency $f$, we are able to measure both $A$ and $\phi$. Figure 2(a) shows PRCs measured at different pump modulation frequencies. The ellipticity, reflecting the amplitude $A$ is the retardation phase of the spin polarization modulation with respect to the pump modulation at $B_F = 150$ mT. The line shows linear fit. $T = 5$ K, $T_R = 13$ ns.

Next, we concentrate on evaluating the longitudinal spin relaxation time $T_1$ and studying its temperature dependence. At low temperatures we use the spin inertia method [44] for that purpose. In order to perform synchronous detection, the intensity of the circularly-polarized pump is modulated at frequency $f$: $P = P_0 [1 + \cos(2\pi ft)]/2$. When the modulation period $1/f$ exceeds the $T_1$ time, the accumulated spin polarization is modulated from 0 up to the maximal value determined by the pumping rate $P_0$ and $T_1$. When the modulation frequency is increased, so that $1/f$ becomes comparable to $T_1$, the spin polarization modulation decreases. One can show that in the case $T_1, 1/f \gg T_R$ the accumulated spin polarization is given by:

$$S(t) = P_0 T_1 + A \cos(2\pi ft - \phi),$$  \hspace{1cm} (2)

$$A = \frac{P_0 T_1}{\sqrt{1 + (2\pi T_1 f)^2}},$$  \hspace{1cm} (3)

$$\tan(\phi) = 2\pi T_1 f.$$  \hspace{1cm} (4)

Let us make two remarks about the spin inertia method and the validity of Eqs. (2-4). First, we do not take into account the saturation effect in QDs: at high-enough pump powers and long-enough $T_1$, the majority of QDs becomes spin-polarized and is no longer affected by further pumping. A more detailed analysis shows that the saturation leads to an effective shortening of $T_1$ entering into Eqs. (2-4). Second, the above analysis assumes a monoeponential dynamics of the spin polarization, characterized by a single time $T_1$. One can show that in the case of a more complex dynamics the dependence of the spin polarization modulation amplitude on $f$ [Eq. (3)] is dominated by the slow component, while the frequency dependence of the retardation phase $\phi$ [Eq. (4)] is dominated by the fast component. Thus, the estimated value $T_1 = 190$ ns is the lower limit for the decay time of the fast component in the longitudinal spin polarization dynamics.

![FIG. 2. Spin inertia effect. (a) Polarization recovery curves taken at a small negative delay for different pump modulation frequencies. The curves are vertically shifted to the same value at $B_F = 0$. (b) Frequency dependence of $\tan(\phi)$, where $\phi$ is the retardation phase of the spin polarization modulation with respect to the pump modulation at $B_F = 150$ mT. The line shows linear fit. $T = 5$ K, $T_R = 13$ ns.](image)

![FIG. 3. Temperature dependencies of longitudinal electron spin relaxation time $T_1$ (stars), inhomogeneous transverse spin relaxation times $T_2^*$ for electrons and holes (solid squares and circles, respectively), and homogeneous transverse spin relaxation times $T_2$ for electrons and holes (open squares and circles, respectively). Solid lines show zero-temperature value of $T_1$ and $T_2$ determined by relaxation with LO phonons according to Eq. (6), dashed lines are guides to the eye. Inset shows polarization recovery curves measured at different time delays (symbols) fitted with Lorentzians (solid lines) at $T = 30$ K.](image)
component of the ellipticity signal at \( B_P = 150 \, \text{mT} \).

It is instructive to compare the temperature dependence of \( T_1 \) to that of \( T_2^* \) for electrons and holes (Fig. 3) solid squares and circles, respectively). The times \( T_2^* \) are determined from the decay of the oscillations in the transverse magnetic field \( B_V = 250 \, \text{mT} \). At low temperatures \( T_2^* \) is determined by the nuclear field and the spread of \( g \) factors [Eqs. (1a,1b)] and is much shorter than \( T_1 \). At higher temperatures the homogeneous dephasing mechanisms related to phonons become important and \( T_2^* \) decreases with \( T \). One can separate the inhomogeneous \( (T_2^\text{inh}) \) and homogeneous \( (T_2) \) contributions to \( T_2^* \):

\[
1/T_2^* = 1/T_2^\text{inh} + 1/T_2
\]  

(5)

Note that \( T_2^\text{inh} \) is almost temperature independent as evidenced from the temperature independent width of the PRC minimum (for \( T < 50 \, \text{K} \) where it can be measured) which is determined by \( B_N \) and the temperature independent spread of \( g \) factors, depending on the QDs’ shape and composition spread. Taking into account that \( T_2^\text{inh} \approx T_2^* (T = 0) \), we can estimate the homogeneous transverse spin relaxation times \( T_2 \) at elevated temperatures using Eq. 5 (at least the part of the homogeneous spin relaxation rate that is temperature dependent). They are shown by the open squares and circles in Fig. 3 for electrons and holes, respectively. The decrease of \( T_2 \) with temperature is especially pronounced for holes. For electrons, \( T_2 \) is close to \( T_1 \) in the whole temperature range, as it was predicted theoretically [45] and which allows us to attribute the \( T_1 \) dependence and, in general, the long-living spin polarization component to electrons that are residently present in a fraction of QDs.

Now we discuss the origin of the \( T_1 \) temperature dependence for electrons. In the limit of zero temperature \( T_1 \) is determined by the hyperfine interaction with nuclear spins as already discussed (horizontal solid line in Fig. 3). For high temperatures, \( T \gtrsim 50 \, \text{K} \), the spin relaxation is governed by the interaction with LO phonons [30, 40, 47]. In particular, the two-phonon mechanism with absorption and emission of an optical phonon leads to spin relaxation. The relaxation rate due to this process can be described by the following equation [30, 46]:

\[
1/T_{1,LO} = \beta N_{LO} (N_{LO} + 1),
\]

\[N_{LO} = \left( \exp(\epsilon_{LO}/k_B T) - 1 \right)^{-1}, \]  

(6)

where \( N_{LO} \) is the number of phonons, \( \epsilon_{LO} \) is LO phonon energy, and \( \beta \) defines the strength of electron-phonon interaction. The corresponding dependence shown in Fig. 3 by the solid line with \( \epsilon_{LO} = 30 \, \text{meV} \) (LO phonon energy in InAs) and \( \beta = 20 \, \text{ns}^{-1} \) fits the experimental data at high temperatures.

The relatively strong temperature dependence of \( T_1 \) for low temperatures, \( T \lesssim 50 \, \text{K} \), is unclear. The usual temperature-dependent QD spin relaxation mechanisms, spin-orbit interaction involving phonons [48] and phonon-activated electron-nuclear flip-flop processes [48–50], give rates several orders smaller than in the experiment. We note that for the QDs emitting around 900 nm similar temperature dependence of \( T_2 \) was reported [28]. However, in that case \( T_2 \) variation starts from \( T = 15 \, \text{K} \), while in our case \( T_1 \) strongly depends on \( T \) already from 5 K.

One possible source of temperature-dependent spin relaxation at low temperatures might be exchange interaction with carriers in the wetting layer which are localized by shallow inhomogeneities. With moderate increase of temperature these carriers become delocalized activating exchange interaction.

To summarize, we have studied the longitudinal and transverse electron spin relaxation in an ensemble of InAs/In$_{0.53}$Al$_{0.47}$As/In$_P$ quantum dots emitting in the telecom wavelength range. At weak magnetic fields, the major fraction of the total spin polarization decays on the nanosecond time scale due to precession of the individual spins in random nuclear fields. At increased longitudinal magnetic field the spin polarization decays during the sub-microsecond time \( T_1 \) at low temperatures, which decreases by three orders of magnitude when approaching room temperature. At low temperatures (\( T \lesssim 50 \, \text{K} \)) we found relatively strong variation of \( T_1 \) with \( T \) which is so far not understood, while at elevated temperatures (\( T \gtrsim 50 \, \text{K} \)) \( T_1 \) is dominated by spin-orbit relaxation with emission and absorption of an optical phonon. The transverse spin relaxation time \( T_2 \) at elevated temperatures is limited by \( T_1 \) time.

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