Spin coherence generation and detection in spherical nanocrystals

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Abstract

A theoretical description of electron spin orientation and detection by short optical pulses is proposed for ensembles of singly charged semiconductor nanocrystals. The complex structure of the valence band in spherical nanocrystals is taken into account. We demonstrate that the direction of electron spin injected by the pump pulse depends on both the pump pulse helicity and the pump pulse power. It is shown that a train of optical pulses can lead to the complete orientation of the resident electron spin. The microscopic theory of the spin Faraday, Kerr and ellipticity effects is developed and the spectral sensitivity of these signals is discussed. We show that under periodic pumping pronounced mode-locking of electron spins takes place and manifests itself as significant spin signals at negative delays between pump and probe pulses.

(Some figures may appear in colour only in the online journal)

1. Introduction

One of the most important tasks of semiconductor spintronics, a novel branch of condensed matter physics aimed at the fundamental and applied research of charge carrier spin dynamics, is the study of electron and hole spin control by nonmagnetic means [1–4]. In this regard, the manipulation of electron spins by short optical pulses attracts a lot of research attention nowadays [5–8], see [9] for review.

The spin control is usually realized using the pump–probe technique, where a strong circularly polarized pump pulse orients spins of electrons, holes and their complexes and a weak linearly polarized probe pulse monitors their spin polarization via spin (magneto-optical) Faraday, Kerr and ellipticity effects as shown schematically in figure 1(a) [10, 11]. Various aspects of the pump–probe technique and features of electron spin dynamics manifested in the pump–probe experiments are reviewed in [1, 2, 12, 13].

Among a rich variety of solid-state systems where the pump–probe technique has been successfully applied, the structures with singly charged quantum dots are of special importance [1, 13–15]. Ultra-long spin relaxation times in these systems allow one to observe various interesting phenomena including spin precession mode-locking [16] and nuclei-induced spin precession frequency focusing phenomena [17]. Due to these effects about a million electron spins localized in different dots are coherent and precess synchronously. In most pump–probe spin dynamics studies self-assembled quantum dots (quantum disks), where due to the size quantization and strain the ground valence band state is twofold degenerate and corresponds to the hole spin projections ±3/2 onto the growth axis, were used. For such structures a microscopic theory of spin Kerr, Faraday and ellipticity effects was developed in [18]. This theory is in good agreement with experiments, see [13, 19].

The aim of the present paper is to address theoretically the processes of spin coherence generation, control and detection in spherical nanocrystals (NCs) where the valence band is fourfold degenerate. The specifics of the optical selection rules in this case result in novel qualitative features of spin generation and detection processes absent in the quantum disks. In particular, as shown below, the direction of electron spin initiated by a single pump pulse or by the pump pulse train depends on the pump pulse power: by changing the power of the circularly polarized pump pulse one can generate electron spin oriented either along or in the opposite direction with respect to the light propagation axis. We demonstrate also that the excitation of electron spins by a periodic train of pump pulses can result in the complete orientation of electron spin and in spin coherence mode-locking if a transverse magnetic field is applied.
The paper is organized as follows. In section 2 we provide a theoretical description of spin coherence generation in a single NC under a circularly polarized pump pulse. Then in section 3 we analyze spin dynamics in the external magnetic field and discuss the spin accumulation processes, among them the spin coherence mode-locking. Section 4 is divided into two parts: the first one aims to provide a description of spin Faraday, Kerr and ellipticity signal formation, and the second one presents temporal dependences of spin signals in the NC arrays. The results are summarized in section 5.

2. Spin coherence generation

An array of spherical NCs grown of III–V compounds is considered. The NCs are assumed to be singly charged with electrons. The ground state of the dot corresponds to the electron at the lowest single size-quantization level; this state is twofold degenerate with respect to the electron spin projection on a given axis. The excited state we consider is the triplet state, which consists of a pair of electrons with anti-parallel spins and a hole. Hereafter, we assume that the optical (carrier) frequencies of the pump, \( \omega_p \), and probe, \( \omega_p \), pulses are close to the singlet trion resonance frequency, \( \omega_0 \), and we neglect therefore all other excited states in the system, e.g. the triplet trion.

The singlet trion state degeneracy is determined by the hole spin states. In spherical NCs under study the ground state of the hole transforms according to the \( T_2 \) representation of the \( T_d \) point symmetry group. We assume that the radius of the NC \( R \) is small enough, \( R \ll a_B \), where \( a_B \) is the exciton Bohr radius, and neglect the Coulomb interaction. Therefore, the trion state is fourfold degenerate, and its states can be labeled as \( F_z = \pm 3/2, \pm 1/2 \). In the spherical (isotropic) approximation \( F_z \) is the component of the hole total angular momentum, which includes the orbital momentum of size-quantized state, \( L \), and the angular momentum of the Bloch function, \( J \) [20–23]. The singlet trion wave function can be written as a product of two-electron function

\[
\psi_{sc}(r_1, r_2) = f_e(r_1)f_e(r_2) \left( \frac{1}{2} \right) \left( \frac{1}{2} \right) \left( \frac{1}{2} \right) \left( \frac{1}{2} \right) ,
\]

where \( f_e(r) = (2\pi r^2 R)^{-1/2} \sin(\pi r/R) \) is the size-quantization function of the electron in the limit of infinite barriers, \( |s_z| \), are the corresponding spinors, and the hole wave function

\[
\psi_{h,F_z}(r_h) = f_0(r_h)|L = 0, J = 3/2, F = 3/2, F_z\rangle + f_2(r_h)|L = 2, J = 3/2, F = 3/2, F_z\rangle ,
\]

where \( L \) is the orbital angular momentum of the hole envelope function, \( J \) is the hole spin, and the functions \( |F, J, F_z\rangle \) are eigenfunctions of the total angular momentum \( F = L + J \). Radial functions \( f_l(r) (l = 0, 2) \) can be expressed in the spherical approximation as \[23\]

\[
f_l(r) = C \left[ j_l(\xi r/R) + (-1)^{l/2} \frac{j_2(\sqrt{\xi} r)}{j_2(\sqrt{\xi} R)} j_l(\sqrt{\xi} r/R) \right] ,
\]

where \( j_l(x) \) are the spherical Bessel functions, \( \beta \) is the light to heavy mass ratio in bulk material, \( \xi \) is the first root of \( j_0(\xi)j_2(\sqrt{\xi} R) + j_2(\xi)j_0(\sqrt{\xi} R) = 0 \) and \( C \) is the normalization constant.

The geometry of the system under study is illustrated in figure 1(a). In what follows we choose the light propagation axis to be \( z \parallel [001] \). The optical selection rules at the trion resonant excitation are similar to those for the interband absorption in bulk material [24]: under \( \sigma^+ \) polarized pulse action trions with \( F_z = 3/2 \) and 1/2 are formed, while for \( \sigma^- \) pulse trions with \( F_z = -3/2 \) and -1/2 are formed [25]. Namely, under a \( \sigma^+ \) light pulse optical transitions from the electron state with spin projection \( S_z = +1/2 \) state to the \( F_z = +3/2 \) trion state and from the \( S_z = -1/2 \) electron state to the \( F_z = +1/2 \) trion state take place as schematically shown in figure 1(b). Similar selection rules with inversion of \( F_z \) and \( S_z \) signs are relevant for the \( \sigma^- \) pump pulse. Hence, for given pump pulse helicity two optical transitions are involved, in contrast to the quantum disk case with a simple valence band, considered in [13, 18].

To describe qualitatively the resident electron spin polarization induced by the pump pulse we note that the probabilities to form a trion from \( S_z = +1/2 \) and \( S_z = -1/2 \) initial states are different. Indeed, as follows from the symmetry of the system, the ratio of the matrix element absolute values describing transitions to the states with \( |F_z| = 3/2 \) and \( |F_z| = 1/2 \) is equal to \( \sqrt{3} \) [24]. Therefore, for example, for the \( \sigma^+ \) pump the trions are formed more efficiently from \( S_z = +1/2 \) electrons than from \( S_z = -1/2 \) ones. Under the assumption that the hole-in-trion spin relaxation time, \( \tau_{DP} \), is much smaller than the trion lifetime, \( \tau_{DP} \), the electron returning after the trion recombination is unpolarized. As a result, an imbalance of \( S_z = +1/2 \) and
−1/2 electrons occurs. This model explains the principle of spin orientation of resident electrons in NCs and it is valid only for relatively weak pump pulses. The microscopic theory for arbitrary pump pulse powers is put forward below.

In what follows, we assume that the duration of the pump pulse, \( \tau_p \), is small enough to neglect the spin precession in an external magnetic field and the recombination and relaxation processes during the pump pulse action: \( \tau_p \ll \tau_{QD}, \tau_{T_L}, \tau_{S, e} \), where \( \tau_{S, e} \) is the electron spin relaxation time in the quantum dot and \( \tau_{T_L} \) is the Larmor spin precession period. Hence, it is enough to determine the transformation of electron spin by the pump pulse, and solve afterwards kinetic equations for electron spin dynamics in the interval between the pump pulses [18]. Accordingly, we introduce the six-component wave function \( \Psi \) of the quantum dot:

\[
\Psi = [\psi_{+1/2}, \psi_{-1/2}, \psi_{+3/2}, \psi_{-3/2}, \psi_{+1/2}, \psi_{-1/2}, \psi_{-3/2}].
\] (1)

where \( \psi_{\pm 1/2} \) refer to the resident electron spin states, and four components \( \chi_{F_i} \) \( (F_z = \pm 3/2, \pm 1/2) \) refer to the corresponding trion states. It is assumed that the trion states are chosen in the canonical basis for the electron spin states, and two corresponding trion ones, will be modified by the pump pulse. The non-stationary Schrödinger equation can be written for the \( \sigma^+ \) polarized pump pulse as (cf [13, 18])

\[
\begin{align*}
    \dot{i} \psi_{+1/2} &= \omega_0 \chi_{+3/2} + \sqrt{3} f(t) e^{-i \omega_p t} \psi_{+1/2}, \\
    \dot{\psi}_{+1/2} &= \sqrt{3} f(t)e^{i \omega_p t} \chi_{+3/2}, \\
    \dot{i} \psi_{-1/2} &= \omega_0 \chi_{-3/2} + f(t) e^{-i \omega_p t} \psi_{-1/2}, \\
    \dot{\psi}_{-1/2} &= f(t)e^{i \omega_p t} \chi_{-3/2}.
\end{align*}
\] (2a-d)

Here \( f(t) \) is a smooth envelope of pump pulse defined as

\[
f(t) = -\frac{e^{i \omega_p t}}{\hbar} D \sigma_{+1}, (t),
\] (3)

where \( D \sigma_{+1} \propto e^{-i \omega_p t} \) is the right circularly polarized component of the electric field of the pump and

\[
D = -i \frac{e p_{cv}}{\omega_0 m_0} \frac{1}{4 \pi} \int f_\sigma(r) \delta_\sigma(r) \, dr
\] (4)

is the dipole matrix element [25] of interband transition between the states \( \psi_{-1/2} \) and \( \psi_{+1/2} \). In the last expression \( e = -|e| \) is the electron charge, \( m_0 \) is the free electron mass, and \( p_{cv} \) is the interband matrix element of the momentum operator taken between the conduction and valence band Bloch functions at the \( \Gamma \) point of the Brillouin zone. The dependence of electromagnetic field on coordinates is neglected in equation (3) since the NC radius is much smaller than the radiation wavelength. A set of equations similar to equations (2) holds for the \( \sigma^- \) pump pulse, in which case spins of hole \( F_i \) and electron \( S_i \) should be inverted.

In pump–probe experiments the generation and detection of electron spin coherence is usually carried out by the train of pump (and probe) pulses following with the repetition period \( T_R \). It exceeds by far the trion lifetime, \( T_R \gg \tau_{QD} \). Therefore, by the next pump pulse, the trion state in the NC is empty, \( \chi_{F_i}(t \rightarrow -\infty) \equiv 0 \). However, the electron can be, in general, spin polarized. Following [18] we represent the solution of the system equations (2) after the pulse is over \( (t \gg \tau_p) \), as

\[
\begin{align*}
    \psi_{+1/2}(t \rightarrow +\infty) &= Q_+ e^{i \omega_p t} \psi_{+1/2}(t \rightarrow -\infty), \\
    \psi_{-1/2}(t \rightarrow +\infty) &= Q_- e^{-i \omega_p t} \psi_{-1/2}(t \rightarrow -\infty).
\end{align*}
\] (5a-b)

Here \( Q_\pm \in [0; 1] \) and \( \Phi_\pm \in (-\pi; \pi) \) are the parameters, which depend on the shape, power and carrier frequency of the pump pulse. The key difference of the present result, equations (2), from the case of a simple valence band, considered in [13, 18], is the fact that both electron spin components are transformed under the pump pulse action. Equations (2) allow one to relate the electron spin components before the pulse, \( S^- = (S^x_-, \ S^y_-, \ S^z_-) \), and after the pulse, \( S^+ = (S^x_+, \ S^y_+, \ S^z_+) \), as follows:

\[
\begin{align*}
    S^x_+ &= \frac{Q^2_+ - Q^2_-}{4} + \frac{Q^2_+ + Q^2_-}{2} S^z_- - S^x_-, \\
    S^y_+ &= Q \cos \Phi S^x_- + Q \sin \Phi S^y_-, \\
    S^z_+ &= Q \cos \Phi S^z_- - Q \sin \Phi S^x_+.
\end{align*}
\] (6a-c)

where \( Q = Q_+ Q_- \) and \( \Phi = \Phi_+ - \Phi_- \). It follows from equations (2) that the electron spin pseudovectors before the pump pulse and after it are connected linearly. In accordance with equation (6a) the pump pulse generates a \( z \)-spin component (first term) and transforms the already present one (second term). The spin components in the plane perpendicular to the light propagation axis are reduced due to the factor \( Q \) in equations (6b) and (6c) and are rotated around the \( z \)-axis by the angle \( \Phi \). Relations similar to equations (2) hold also for the \( \sigma^- \) pump pulse, in which case \( Q_+ (\Phi_+) \) and \( Q_- (\Phi_-) \) should be swapped.

To simplify the following discussion, we assume that the hole-in-trion spin relaxation is fast as compared with the radiative lifetime of the trion. In this case, the carrier returning after the trion recombination is completely depolarized and the long-living spin coherence generation is governed by equations (2). Otherwise, to determine the resident electron spin induced by the pump pulse one has to solve the full system of spin dynamics equations taking into account both electron and trion spins, cf [26–28].

Let us consider first an important limiting case of a resonant pump pulse, when \( \omega_0 = \omega_p \). It can be shown that \( \Phi_\pm = 0 \) and \( Q_\pm \) are expressed via the effective pump area

\[
\Theta = 2 \int_{-\infty}^{\infty} f(t) \, dr \tag{13}
\]

as

\[
Q_+ = \cos \left( \frac{\sqrt{3} \Theta}{2} \right), \quad Q_- = \cos \left( \frac{\Theta}{2} \right).
\] (7)

The periodic dependence of \( Q_+ \) and \( Q_- \) on \( \Theta \) is related to the Rabi oscillations taking place in two-level systems corresponding to optical transitions between the electron state with \( S_z = +1/2 \) and the trion state with \( F_z = +3/2 \), and between the electron state \( S_z = -1/2 \) and the \( F_z = +1/2 \).
The pump pulse with given helicity can be directed parallel or the Rosen and Zener shape of the pump pulse: pumping. In order to obtain an analytic solution we consider with Following [18], we obtain for \( Q \) considered here, depending on the pump pulse area \( S_z \) in the ground state [18]. Interestingly, for the spherical NC dimensionless detuning. It follows from equations (6 | −valence band structure, and strength one can orient the spin in arbitrary direction for NC systems like those studied in [30, 31].

3. Spin accumulation caused by the train of pump pulses

In the pump–probe Faraday and Kerr rotation experiments the sample is usually subjected to a train of pump pulses that follow with a certain repetition period \( T_R \). We assume that \( T_R \) exceeds by far the radiative lifetime of a trion in a quantum dot, \( \tau_{\text{QD}} \), but it may be comparable to or smaller than the single electron spin relaxation time in a QD, \( T_R \leq \tau_{\text{e-e}} \).

Figure 2. (a) The spin of the resident electron in the NC generated after a single \( \sigma^+ \) pump pulse (red/solid line) and after the train of such pulses (blue/dot–dashed line) as functions of the pump pulse area \( \Theta \) at zero magnetic field. Resident electron spin in the quantum disk after a single pulse \( S_z = -\sin^2(\sqrt{3}\Theta/2)/4 \) is presented by a thin magenta/dashed line. (b) Resident electron spin after a single Rosen and Zener pump pulse for different detunings between the pump optical frequency and NC transition frequency \((\omega_0 - \omega_p) \tau_p/(2\pi) = 0, 0.25, 0.5\) for red/solid, blue/dash–dotted and magenta/dashed curves, respectively.

It is worth noting that for a deformed NC of elliptic shape, or in an NC made of the wurtzite semiconductor, the transitions involving \( S_z = +1/2 \) and \( S_z = -1/2 \) electrons are characterized by different detunings, in general. This results in even more complex dependence of \( S_z^{(1)}(\Theta, y) \) on the pump pulse area. In the particular case where the splitting between \( |F_z| = 3/2 \) and \( |F_z| = 1/2 \) trion states exceeds by far the spectral width of the pump pulse \( h/\tau_p \), only one optical transition can be excited and the spin orientation is described by the model of [18]. In the opposite case the model presented here is valid. Hence, by choosing appropriate pulse spectral width and strength one can orient the spin in arbitrary direction for NC systems like those studied in [30, 31].

\[
\begin{align*}
Q_\pm &= \sqrt{1 - \frac{\sin^2(\Theta_\pm/2)}{\sin^2(\pi y)}}, \\
\Phi_\pm &= \arg\left\{\frac{\Gamma(\frac{1}{2} - iy)}{\Gamma(\frac{1}{2} - \tilde{\Theta}_\pm - iy)\Gamma(\frac{1}{2} + \tilde{\Theta}_\pm - iy)}\right\},
\end{align*}
\]

where \( \Theta_+ = \sqrt{3}\Theta_- = \sqrt{3}\Theta \), and \( y = (\omega_p - \omega_0) \tau_p/(2\pi) \) is the dimensionless detuning. It follows from equations (6a) and (8) that the electron spin \( z \) component induced by a single detuned pump pulse has the form

\[
S_z^{(1)}(\Theta, y) = \frac{S_z^{(1)}(\Theta, 0)}{\sin^2(\pi y)} = \frac{\sin^2(\Theta/2) - \sin^2(\sqrt{3}\Theta/2)}{4\sin^2(\pi y)}.
\]

and decreases monotonically with an increase of the detuning, \( |y| \). This dependence is illustrated in figure 2(b), where different curves correspond to different values of \( y = 0, 0.25, 0.5 \). The overall dependence of \( S_z^{(1)}(\Theta, y) \) is the same for all detunings in the case of the Rosen and Zener pump pulse.

Now let us briefly consider the case of non-resonant pumping. In order to obtain an analytic solution we consider the Rosen and Zener shape of the pump pulse:

\[
f(t) = \frac{\mu}{\text{ch}(\pi t/\tau)}.
\]

where the parameter \( \mu \) determines the pulse area \( \Theta = 2\mu \tau_p \). Following [18], we obtain for \( Q_\pm \) and \( \Phi_\pm \):

\[
Q_\pm = \sqrt{1 - \frac{\sin^2(\Theta_\pm/2)}{\sin^2(\pi y)}},
\]

where \( \Theta_+ = \sqrt{3}\Theta_- = \sqrt{3}\Theta \), and \( y = (\omega_p - \omega_0) \tau_p/(2\pi) \) is the dimensionless detuning. It follows from equations (6a) and (8) that the electron spin \( z \) component induced by a single detuned pump pulse has the form

\[
S_z^{(1)}(\Theta, y) = \frac{S_z^{(1)}(\Theta, 0)}{\sin^2(\pi y)} = \frac{\sin^2(\Theta/2) - \sin^2(\sqrt{3}\Theta/2)}{4\sin^2(\pi y)}.
\]
long train of pump pulses we obtain
\[
S^+_x = S^-_x = S^+_y = S^-_y = 0, \quad (11a)
\]
\[
S^+_z = S^-_z = \frac{Q^2_+ - Q^2_-}{4(2Q^2_+ + Q^2_-)}. \quad (11b)
\]
The dependence of electron spin $S_z$ component accumulated by the train of pump pulses and calculated after equation (11b) is shown in figure 2(a) by the dash–dotted line. Interestingly, for the Rosen and Zener pump pulse shape, the dependence of $S_z$ on the pump pulse area is independent of the detuning between the pump pulse frequency and the quantum dot transition frequency, as follows from equations (8) and (11b). We have checked that this dependence is weak for a rectangular-shaped pulse.

It is noteworthy that the electron spin polarization may reach 100% for a spherical NC. This is in contrast to the classical optical orientation regime in the systems with $\Gamma$ symmetry of the valence band, where up to 50% spin polarization can be realized. Such an enhancement of spin polarization and the dependence of the electron spin direction on the pump pulse area is related to the two-level nature of the optical transitions in NCs. For example, if one of the two-level systems (associated with the $F_e = +3/2$ or $+1/2$ trion) is inactive, which happens if $Q_+ = 1$ or $Q_- = 1$, only electrons with the fixed spin component are depolarized ($S_z = 1/2$ or $-1/2$, respectively). Such a situation is similar to the one considered in [18, 33]. As a result, a sufficiently long train of pump pulses completely erases one electron spin component, hence resident carriers become fully polarized.

In the general case, where the magnetic field is applied to the NC, the resident electron spin dynamics is governed by the following equation:
\[
\frac{dS}{dt} + S \times \Omega_L + \frac{S}{\tau_{s,e}} = 0, \quad (12)
\]
The equation takes into account the electron spin relaxation processes and the precession of electron spins in the magnetic field $B$ with the frequency $\Omega_L = g_e \mu_B B$, where $g_e$ is the electron Landé factor and $\mu_B$ is the Bohr magneton. Following [18, 28] we obtain the following steady-state expressions for the electron spin components $S$ right before the pump pulse arrival:
\[
S^+_x = KS^-_y, \quad (13a)
\]
\[
S^-_y = \frac{Q^2_+ - Q^2_-}{4\Delta} e^{-T_R/\tau_{s,e}} \sin(\Omega_L T_R), \quad (13b)
\]
\[
S^-_z = \frac{Q^2_+ - Q^2_-}{4\Delta} e^{-T_R/\tau_{s,e}} \sin(\Omega_L T_R) \times \left[ Q(\cos \Phi - K \sin \Phi) e^{-T_R/\tau_{s,e}} - \cos(\Omega_L T_R) \right], \quad (13c)
\]
where
\[
\Delta = 1 - e^{-T_R/\tau_{s,e}} \left[ \frac{Q^2_+ + Q^2_-}{2} + Q(\cos \Phi - K \sin \Phi) \right] \times \cos(\Omega_L T_R) + (1 - \cos(\Omega_L T_R)(2 - \Theta^2) + (41 - 17 \cos \Omega_L T_R) \Theta^4) \left\{ (1 - \cos \Omega_L T_R) \right\}^{-1}, \quad (14a)
\]
\[
\frac{Q(Q^2_+ + Q^2_-)}{2} e^{-2\pi N/\tau_{s,e}} (\cos \Phi - K \sin \Phi), \quad (14b)
\]
\[
K = \frac{Q e^{-T_R/\tau_{s,e}} \sin \Phi}{1 - Q e^{-T_R/\tau_{s,e}} \cos \Phi}. \quad (14c)
\]
We recall that $Q = Q_+ Q_-$. In the limit of $Q_- = 1$, i.e. where one of the two-level systems is inactive, equations (13) and (14) pass to those obtained in [18] for the simple valence band system.

In order to analyze equations (13) we consider an important limiting case of resonant pumping, where $\Phi = 0$, and neglect completely the resident electron spin relaxation ($\tau_{s,e} \rightarrow \infty$). This results in $K = 0$,
\[
\Delta = 1 - \left[ \frac{Q^2_+ + Q^2_-}{2} + Q \right] \cos(\Omega_L T_R) + \frac{Q(Q^2_+ + Q^2_-)}{2}, \quad (15)
\]
see equations (14), and leads to the following simplified expressions for the electron spin components accumulated by the train of pump pulses:
\[
S^+_x = 0, \quad (16a)
\]
\[
S^-_y = \frac{Q^2_+ - Q^2_-}{4\Delta} \sin(\Omega_L T_R), \quad (16b)
\]
\[
S^-_z = \frac{Q^2_+ - Q^2_-}{4\Delta} [Q - \cos(\Omega_L T_R)]. \quad (16c)
\]
Clearly, $S_x \equiv 0$ for the considered geometry, since the magnetic field is parallel to the $x$ axis and optical pulses do not lead to spin rotation in the $(xy)$ plane in the resonant case.

Under the phase synchronization condition [16, 17, 28, 32, 34–36],
\[
\Omega_L T_R = 2\pi N, \quad N = 0, 1, 2, \ldots, \quad (17)
\]
the spin of the electron makes an integer number of turns between the consecutive pump pulses and the electron spin is given by the same expressions as in the absence of the magnetic field (see equations (11)), and may reach $\pm 1/2$ depending on pump pulse area.

For weak pump pulses, where the pulse area $\Theta \ll 1$, one obtains
\[
S^+_x \approx \frac{Q^2}{16} \left( 1 - (2 + \cos \Omega_L T_R) \Theta^2 \right) \left\{ 3(1 - \cos \Omega_L T_R) \right\}^{-1}, \quad (18a)
\]
\[
S^-_y \approx \frac{Q^2}{8} \times \sin(\Omega_L T_R) \left( \cos(\Omega_L T_R)((2 - \Theta^2) + (41 - 17 \cos \Omega_L T_R) \Theta^4) \right) \left\{ (1 - \cos \Omega_L T_R)(2 - \Theta^2) + (41 - 17 \cos \Omega_L T_R) \Theta^4 \right\}^{-1}, \quad (18b)
\]
Note that the peaks of the dependence of $S_z$ on magnetic field corresponding to the phase synchronization condition (17) are very sharp. If the spin relaxation and the detuning are completely neglected, their width is determined by the pump pulse area ($\sim \Theta^2$), otherwise it is determined by the ratio $T_R/\tau_{s,e} < 1$ or $|\Phi|$, whichever is larger; see [18] for details.

In the resonant case the electron spin $z$ component is equal to

\[
\frac{Q(Q^2_+ + Q^2_-)}{2} e^{-2\pi N/\tau_{s,e}} (\cos \Phi - K \sin \Phi), \quad (14b)
\]
\[
K = \frac{Q e^{-T_R/\tau_{s,e}} \sin \Phi}{1 - Q e^{-T_R/\tau_{s,e}} \cos \Phi}. \quad (14c)
\]
Here we assume that $x$ probe pulse oscillates along the Faraday and Kerr rotation experiments is carried out by a
The detection of the QD spin polarization in pump–probe signals at long enough delays are determined by
radiative lifetime of an electron–hole pair in the NC; therefore, the electron spin
component only. It is assumed below that $Q_0 = 0$ for the sake of definiteness. In this situation under the synchronization condition (17) the electron spin becomes fully polarized.

4. Probing the electron spin in quantum dots

4.1. Formation of spin Faraday and ellipticity signals in NC

The detection of the QD spin polarization in pump–probe Faraday and Kerr rotation experiments is carried out by a weak linearly polarized probe pulse. The electric field of the probe pulse oscillates along the $x$ axis and can be written as

$$\mathbf{E}^p(\mathbf{r}, t) = E^p_t(\mathbf{r}, t) \mathbf{a}_x + \text{c.c.}$$

where $E^p_t(\mathbf{r}, t) = E_0 s(t) e^{-i\omega_{pr} t}$, where $\omega_{pr}$ is the carrier frequency of the probe beam, $E_0$ is the probe pulse amplitude, $s(t)$ is its smooth envelope, and $\mathbf{a}_x$ is the unit vector along the $x$ axis. The Faraday signal $\mathcal{F}$ detected in the transmission geometry can be written as [13, 18]

$$\mathcal{F} = \lim_{z \to +\infty} \int [\mathcal{E}^{(0)}_s(z, t)]^2 - [\mathcal{E}^{(0)}_y(z, t)]^2 \, dt,$$

where $x'$, $y'$ axes are oriented at $45^\circ$ with respect to the initial reference frame $x$, $y$; $E^{(0)}_{x'}(z, t)$ and $E^{(0)}_{y'}(z, t)$ are the components of the transmitted field. The Kerr rotation signal can be presented similarly to equation (20) with the replacement of the transmitted fields by the reflected ones. Similarly, the ellipticity of the transmitted beam reads

$$\mathcal{E} = \lim_{z \to +\infty} \int [\mathcal{E}^{(0)}_{\sigma^+}(z, t)]^2 - [\mathcal{E}^{(0)}_{\sigma^-}(z, t)]^2 \, dt.$$  

Here the subscripts $\sigma^+$ and $\sigma^-$ correspond to the circular components of the transmitted light.

In order to find the transmitted and reflected electromagnetic field we follow the procedure developed in [13, 18] and calculate the response of the NC to the linearly polarized probe field. To that end, the electric field of the probe pulse is decomposed in a superposition of circularly polarized waves, and system (2) and its counterpart for the other circular polarization are used to determine the change of the quantum dot wavefunction. Corresponding equations are solved in the first order in electric field $\mathbf{E}^{pr}$.\footnote{The linearly polarized pulse affects the electron spin polarization in the NC [8]. One can show that electron spin components before ($S^\pm$) and after the dielectric polarization of the NC is calculated and the re-emitted field is determined from the Maxwell equations. Under the assumption that the distance between the NCs is smaller than the wavelength of the probe, the resulting expressions for spin Faraday and ellipticity signals for the three-dimensional (bulk) array of NCs read:

$$\mathcal{E} + i\mathcal{F} = \frac{\pi N_{NC} L}{4\pi^{2} T_{NC}}[|J_z| + i(|J_x|, J_y)|s| - 2S_z] 
\times G(\omega_{pr} - \omega_0),$$

where $\omega = \omega_{pr} \sqrt{\varepsilon_0 / \varepsilon_b}$ is the radiation wavevector in the medium, $\varepsilon_b$ is the dielectric constant of the matrix, which is assumed to coincide with the background dielectric constant of the NCs,

$$\tau_{NC} = \frac{3\hbar c^3}{4|d|^2 \varepsilon_0 \varepsilon_b}$$

is the radiative lifetime of an electron–hole pair confined in the NC [25], $N_{NC}$ is the (volume) density of the NCs and $L$ is the thickness of the array. The complex-valued function $G(\omega_{pr} - \omega_0)$ in equation (22) determines the spectral sensitivity of the spin Faraday and ellipticity signals [13, 18].}

The explicit expressions for $G(\Lambda)$ are given by equation (61) of [18] for different probe pulse shapes. Its real and imaginary parts for the case of a Rosen and Zener pulse are shown by red dashed and blue dotted lines in figure 3(a), respectively. It is noteworthy that the spin Kerr signal is a superposition of spin Faraday and ellipticity signals with the coefficients determined by phase acquired by the probe pulse in the cap layer [13, 18].

Quantities $S_z$ and $J_z$ in equation (22) are the electron and the hole-in-trion $z$-spin components at the moment of the probe pulse arrival. The probe pulse duration is assumed to be short as compared with all other time scales in the system, and therefore the electron spin can be considered as frozen during the probe pulse action. Interestingly, the Faraday rotation and ellipticity signals contain contributions from the quantity $\langle |J_x|, J_y|s| \rangle$, which is the quantum mechanical average of the symmetrized product of $J_x$, $J_y$ hole spin operators, $\langle J_x, J_y \rangle = (J_x, J_y + J_y, J_x)/2$. Note that a similar combination of electron spin operators (although symmetry allowed) does not contribute to the spin Faraday and ellipticity effects, since second powers of electron spin operators reduce to the first powers\footnote{In the regime linear in $\mathbf{E}^{pr}$ the probe-induced dielectric polarization does not contain contributions such as $\langle J_x, \sigma_\tau \rangle$, either.}. The contributions due to the trion spin polarization vanish if the pump–probe delay exceeds $\tau_{NC}$, the radiative lifetime of an electron–hole pair in the NC; therefore, pump–probe signals at long enough delays are determined by the electron spin $z$ component only. It is assumed below that the pump–probe delay exceeds the trion lifetime in the NC (S$^+\tau$) the linearly polarized pulse are related as $S^+ = QS^-$, where, e.g., for a resonant pulse $Q = \cos^2(\theta_0 / \sqrt{2})$, here $\theta_0$ is the linearly polarized pulse area.
and we focus solely on the electron spin contribution to the Faraday and ellipticity signals.

We note that due to the degeneracy of the heavy- and light-hole states the spin Faraday, Kerr and ellipticity signals are proportional to the electron spin projection onto the light propagation axis. Hence, by changing the direction of light propagation one can study the dynamics of all spin pseudovector components, making spherical NCs most suitable for the spin state tomography measurements [37].

Before we proceed with the discussion of spin Faraday and ellipticity signal temporal behavior, let us estimate the typical spin signal strengths. For the NC array with a concentration $N_{NC} = 10^{15} \text{ cm}^{-3}$, thickness $L = 10^{-4} \text{ cm}$, the Faraday rotation angle can be estimated according to equation (22) as $\sim 10 \text{ mrad}$ for $\tau_{NC} = 400 \text{ ps}$, $\hbar \omega_0 = 1.4 \text{ eV}$, $\hbar \Delta \omega_0 = 6.15 \text{ meV}$ [16]. Parameters $A$ and $C$ in equation (25) are $A = -1.75 \text{ eV}^{-1}$, $C = 2.99$ [16].

**Figure 3.** (a) Electron spin polarization, $S^{+}_z$, created by a train of Rosen and Zener $\sigma^+$ polarized pump pulses with the repetition period $T_R = 13.2 \text{ ns}$, as a function of NC optical transition frequency $\omega_0$ (black/solid line). The real (red/dashed) and imaginary (blue/dotted) parts of function $G(\omega - \omega_p)$ are shown in panels (a) and (d). Panels (b), (c) represent the time resolved dependence of the spin ellipticity and Faraday rotation signals in the QD ensemble for the degenerate pump–probe regime ($\omega_p = \omega_R$). Panels (d)–(f): the same as in (a)–(c), but for the nondegenerate regime ($\omega_p - \omega_0$) $T_R = 0.8 \pi$. Calculations are carried out for pump pulses of area $\Theta = \pi$, in-plane magnetic field $B = 1 \text{ T}$, $\tau_p = 100 \text{ fs}$, $\tau_{se} = 1 \mu s$, $\hbar \omega_0 = 1.4 \text{ eV}$, $\hbar \Delta \omega_0 = 6.15 \text{ meV}$ [16]. Parameters $A$ and $C$ in equation (25) are $A = -1.75 \text{ eV}^{-1}$, $C = 2.99$ [16].

where $A$ and $C$ are constants. In accordance with equation (22) the electron spin Faraday and ellipticity signals as functions of the time delay between the probe and the pump pulses $t$ in the quantum dot ensemble are given by (cf [13, 19])

$$
\mathcal{E} + i \mathcal{F} = -2 \frac{\pi N_{NC} L}{e^2 \tau_{NC}^2} |E_0|^2 \left[ \int S_z(\omega_0, \omega_p; t) \times G(\omega_p - \omega_0) \rho(\omega_0) \ d\omega_0 \right].
$$

where $S_z(\omega_0, \omega_p; t)$ is the temporal dependence of the electron spin in the quantum dot on the resonance frequency $\omega_0$ induced by the pump with the carrier frequency $\omega_p$, which can be found from equations (12) and (6). The variation of $\tau_{NC}$ with the variation of resonance frequency is neglected in equation (26), since this dependence is very smooth on the scale of the inverse pulse duration.

The spread of spin precession frequencies caused by the $g$-factor spread gives rise to important consequences. First, for some quantum dots in the ensemble the spin precession frequency and the pump pulse repetition frequency are commensurable (see equation (17)), resulting in the strong enhancement of the $S_z$ in these dots. The distribution of the electron spin $z$-component right after the pump pulse arrival as a function of the quantum dot optical transition frequency is shown by the solid curve in figures 3(a) and (d). The peaks on these curves correspond to the dots which satisfy the phase synchronization condition (17). This yields the mode-locking of electron spin precession [16]: the total spin of the ensemble induced by the pump pulse becomes dephased due to the spread of the spin precession frequencies on the nanosecond time scale, and before the next pulse arrival the spin polarization of the ensemble emerges due to the contribution of the NCs where the spin precession is synchronized. This is clearly seen in figures 3(b), (c), (e) and (f), where the temporal dependence of the spin ellipticity ((b), (e)) and Faraday rotation ((c), (f)) are presented for different detunings between the pump and probe pulses. It is worth stressing that the allowance for the nuclei-induced electron...
spin precession frequency focusing effect can result in even higher amplitudes of the signals at negative delays [17, 34, 35].

The second consequence of the g-factor spread is clearly manifested in figure 3(c), where the Faraday rotation signal for degenerate pump and probe pulses is demonstrated. Notably, the Faraday rotation is absent at \( t = 0 \), and the signal amplitude is a non-monotonic function of the time delay: firstly it grows with time and afterwards it decays. This is a feature of the spectral sensitivity of the Faraday effect which is described by an odd function of the detuning; see the blue dotted curve in figure 3(a). The dependence of electron spin z-component on the detuning, \( \omega - \omega_p \), is symmetric right after the pump pulse arrival (\( t = 0 \)) but becomes asymmetric as time goes by due to the correlation of the electron g-factor and the resonance frequency of the NC. It results in the growth of the Faraday rotation signal with time. For even higher time delays the Faraday rotation amplitude decays due to the spin dephasing [19]. The pump–probe detuning itself introduces the asymmetry of the spin distribution with respect to the probe frequency (see figure 3(d)) and the Faraday rotation signal shows behavior similar to that of the ellipticity, namely, damped oscillations. The interaction of electron spins with nuclei may result in breaking of the direct link of the electron spin precession mode-locking and lead to the complete suppression of the ellipticity signal at negative delays [17, 34, 35].

Three-dimensional inhomogeneous arrays of NCs are demonstrated in figure 3(e) and (f). The results of this work can also be applied to the concept of spin coherence generation and detection for electrons in colloidal NCs or for electrons localized on donors in the bulk GaAs-type semiconductors where the donor-bound exciton can be optically excited.

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