Coherent spin dynamics of electrons and holes in semiconductor quantum wells and quantum dots under periodical optical excitation: resonant spin amplification versus spin mode-locking

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(Dated: December 30, 2011, file = rsa-modlock-29dec11.tex, printing time = 1:46)

The coherent spin dynamics of resident carriers, electrons and holes, in semiconductor quantum structures is studied by periodical optical excitation using short laser pulses and in an external magnetic field. The generation and dephasing of spin polarization in an ensemble of carrier spins, for which the relaxation time of individual spins exceeds the repetition period of the laser pulses, are analyzed theoretically. Spin polarization accumulation is manifested either as resonant spin amplification or as mode-locking of carrier spin coherences. It is shown that both regimes have the same origin, while their appearance is determined by the optical pump power and the spread of spin precession frequencies in the ensemble.

PACS numbers: 78.67.-n, 78.47.-p, 71.35.-y

I. INTRODUCTION

The coherent spin dynamics of carriers in semiconductor nanostructures attract considerable attention nowadays due to future quantum information technologies based on spintronics applications. With respect to fundamental studies this research field delivers exciting and unexpected results on the properties of spin systems and the possibility to control them by external fields or by structural parameters.

Optical pump-probe techniques for time-resolved measuring of Faraday and Kerr rotation are based on excitation by trains of laser pulses where the pulse durations range from hundreds of femtoseconds to a few picoseconds. They have been demonstrated to be among the most reliable tools for investigating coherent spin dynamics. The principle of these magneto-optical techniques is the following: an intense laser pulse of circularly polarized light (the pump) is used to orient spins and, therefore, to create a macroscopic spin polarization. This polarization is probed by the linearly polarized probe pulses through rotation of their polarization plane after propagation through the spin polarized medium (the Faraday effect) or reflection at this medium (the Kerr effect). The probe pulse is time-delayed relative to the pump pulse, and by tuning this delay one can measure the spin polarization dynamics. To study the coherent spin dynamics the sample is exposed to an external magnetic field, typically oriented perpendicularly to the light wave vector (Voigt geometry), which allows one to detect the precession of the optically induced spin polarization and monitor its decay. Application of these techniques to single spins, which is potentially possible for systems with a relatively small dispersion of precession frequencies, and spin mode-locking (SML) found for an ensemble of singly-charged QDs with a large dispersion of Larmor frequencies (see, e.g., Refs. and references therein).

For studying the RSA regime experimentally, scanning the magnetic field has been suggested instead of the commonly used scan of the pump-probe time delay, used also for tracing SML. The probe pulse arrival time in this case is fixed at a small negative delay prior to the pump pulse. The resulting RSA spectrum is a periodic function of magnetic field from which information such as carrier $g$ factor and dephasing time of the spin ensemble can be extracted.

In this paper we show that the RSA and SML are two different manifestations of the same phenomenon: spin accumulation caused by the periodic excitation with pump pulse trains. We elaborate the fundamental differences in conditions for appearance of these two regimes. The most important parameters in this regard are the pump power and the spin precession frequency spread causing spin dephasing. Differences of the two parameters, in turn, lead to different phenomenologies in experiment, providing significantly different capabilities for analyzing spin systems quantitatively.

The paper is organized as follows. In Section I we recall the basic concepts and equations for describing spin coherence generation. We discuss the difference between the classical and quantum mechanical approaches to describing carrier spin coherence generation for resonant trion excitation. Then we consider generation of long-lived spin coherence during the trion lifetime. We describe the spin dynamics of charged carriers and trions...
in magnetic field and discuss the effects of spin relaxation and spin precession of the trion spin on the long-lived spin coherence of resident carriers. We also consider here the long-lived dynamics after generation and the spin accumulation caused by the train of pump pulses. Section III is devoted to the RSA regime, for which we consider different conditional effects: trion spin relaxation, nuclear field fluctuations, and spin relaxation anisotropy. The conditions, which are important for observing RSA, and the characteristics, which one can extract from the analysis of RSA signals, are collected at the end of Sect. III. Section IV describes the main features of mode-locking of electron spin coherences. Then in Section V we compare the spin dynamics in the RSA and SML regimes, obtain conditions for the SML regime and discuss the transition to the RSA regime. In the Conclusions, we give a comparative description of the RSA and SML regimes and their applicability to investigations of long-lived spin dynamics in low-dimensional systems.

II. GENERATION OF SPIN COHERENCE

In the following we analyze the long-lived spin coherence of resident carriers (electrons and holes) generated by periodic light excitation in semiconductor quantum wells and quantum dots. We consider a situation with a low concentration of resident carriers, when the probability to have two charge carriers with significantly overlapping wavefunctions is low. In this case, mainly few-particle complexes, excitons (electron-hole pairs) and tri-ons (three particle complexes) can be optically excited, while other many-body correlations are negligible. For quantum wells this corresponds to typical carrier densities smaller than $10^{10}$ cm$^{-2}$, for which at liquid Helium temperatures carriers are localized on QW width fluctuations with respect to their in-plane motion. Only one carrier per localized site is typical for such concentrations and the distance between the localized carriers exceeds the extensions of neutral and charged exciton wavefunctions. For quantum dots the low concentration regime corresponds to occupation of a dot with only one resident carrier, i.e. to a regime of singly-charged QDs.

Here we consider the theoretical aspects of the problem. We do not discuss the experimental aspects of the observations (measurements) of long-lived spin coherences and features of ellipticity and Faraday rotation signals. We limit ourselves to the degenerate pump-probe regime, when the probe laser has the same photon energy as the pump one, and to resonant excitation of the trion states. We assume that the pulse duration is significantly shorter than all characteristic relaxation times of the considered spin system. Other regimes are studied in detail elsewhere. These conditions are typical for experiments with semiconductor nanostructures.

For low concentrations of resident carriers charged excitons (trions) play an important role in the generation process of carrier spin coherence. A negatively charged exciton ($T^-$ trion) is a bound state of two electrons and one hole, while a positively charged exciton ($T^+$ trion) is a bound state of two holes and one electron. The trion ground state at zero magnetic field has a singlet spin configuration, such that the spins of the two identical carriers are aligned opposite to each other and the trion Zeeman splitting is controlled by the $g$ factor of the unpaired carrier, e.g., the hole in $T^-$. Hereinafter we assume that only heavy holes with angular momentum projections $\pm3/2$ onto the growth axis are involved.

The theoretical analysis used in this paper can be equally applied to structures with resident electrons or resident holes. In order to do that we have introduced universal notations: the resident carrier spin $S$, the trion spin $S_T$, the Larmor frequency of the resident carrier $\omega = g_\mu_B B/\hbar$, and the Larmor frequency of the trion $\Omega = g_T h B/\hbar$. Here $B$ is the external magnetic field, $\mu_B$ is the Bohr magneton, $g$ and $g_T$ are the $g$ factors of the resident carrier and trion, respectively. Similarly universal notations are also used in what follows to denote characteristic time scales.

In $n$-type doped structures with resident electrons, $S$ is the electron spin, $S_T$ is a (pseudo) spin of the $T^-$ trion ($S_T = +1/2$ for $+3/2$ hole and $-1/2$ for $-3/2$ hole), $\omega$ is the electron Larmor frequency, and $\Omega$ is the $T^-$ Larmor frequency determined by the hole $g$ factor. Correspondingly, in $p$-type doped structures with resident holes, $S$ is the heavy hole pseudospin, $S_T$ is the spin of the $T^+$ trion, which corresponds to the electron spin in this trion, $\omega$ is the heavy hole Larmor frequency, and $\Omega$ is the $T^+$ Larmor frequency determined by the electron $g$ factor.

For the sake of simplicity we consider in most parts of this paper $n$-type doped structures with resident electrons, as there are more experimental data available for these structures. Wherever we analyze $p$-type doped structures this will be notified. Before we proceed to the analysis of spin precession in magnetic field and spin dephasing processes, let us inspect briefly the models of optical generation of spin coherence.

A. Resonant excitation of trion. Classical and quantum mechanical approaches to carrier spin coherence generation

The important quantity for spin coherence generation in the system is the singlet trion resonance that is excited optically. The trion generation probability for resonant excitation depends on the light polarization and the spin orientation of the resident carrier. For instance, in $n$-type doped structures a $\sigma^+$ polarized pump generates a hole with spin projection $+3/2$ onto the light propagation axis $z$ and an electron with spin projection $-1/2$. Therefore, trion formation is possible only when the resident electron has spin projection $+1/2$. As a result, the circularly polarized pump pulse selects electrons with particular spin orientation from the ensemble of resident electrons to form trions. This, in turn, leads to spin polarization of the resident electrons.

There are two approaches for describing the spin coherence generation by circularly polarized light pulses. The first one is essentially quantum mechanical: a singly-charged QD or a QW with a localized resident electron is modeled as a two-level system. The ground state corresponds to the resident electron, while
the excited state is the singlet trion, see Fig. 1(a).

The interaction of the two-level system with the resonant pump pulse depends on the pulse parameters (polarization, intensity and pulse duration) and on the level occupations. The pump pulse action time, $\tau_p$, is assumed to be the shortest of all timescales in the problem, namely the trion dephasing and scattering times, the electron Larmor precession period, the trion radiative lifetime, the spin dephasing/decoherence times, etc. Under usual experimental conditions the trion lifetime is much shorter than the pump pulse repetition period and, consequently, trion spin polarization is absent shortly before the next pump pulse, i.e., is not detectable at negative time delays. It follows then, that the resident carrier spin pseudovector $\mathbf{S} = (S_x, S_y, S_z)$ before the pump pulse, $\mathbf{S}^0$, and after the pump pulse, $\mathbf{S}^\pm$, are related to each other through (2):

$$S_z^\pm = S_z^0 \pm \frac{Q^2}{16} \tau_p^2, \quad (1a)$$

where $S_z^\pm$ is the component of the trion spin pseudovector after, e.g., a $\sigma^+$ pump pulse in a $p$-type structure or a $\sigma^-$ pump pulse in a $p$-type structure and $\tau_p$ is the pump pulse area, i.e., shows Rabi oscillations inherent to a two-level system, see e.g. Refs. [14, 15, 27].

The experimentally studied situation in $n$-type QW structures is different [28]. For low pump powers and resonant trion excitation the electron spin coherence increases linearly with the pump power, see Eq. (3), while at high powers the spin $z$ component saturates and Rabi oscillations are not observed [28]. Clearly, the two-level model is not sufficient for describing such a behavior. The most probable reason is related to the weaker localization of electrons and trions in quantum wells and, hence, to the presence of many trion states. Scattering between these states becomes possible, as schematically illustrated in Fig. 1(b). The optical coherence of the trion with the pump is lost due to this scattering, while spin coherence is preserved. As a result, if the scattering time between different trion states, $\tau_1$, is considerably shorter than $\tau_p$, the Rabi oscillations at high pump powers vanish [28], because the population of the excited state of the two-level system coupled to the optical pulse is small. At the same time, the spin polarization generated by the pump pulse can be substantial, because spin does not relax during scattering. With an increase of the pump pulse power the electron spin saturates at the value

$$S_{z, max} = \mp N/4, \quad (4)$$

where $N$ is the total number of resident electrons in the system. The amount of trions formed for resonant excitation of the initially unpolarized electron ensemble cannot exceed $N/2$, since only half of the resident electrons have suitable spin orientation to become excited to trion singlets. The other $N/2$ of the resident electrons, which are not captured to trions, have become fully polarized.

As will be shown below in Sec. III, the quantum mechanical and classical approaches give the same results at low pump powers. Subsequently, we will use the quantum mechanical approach because it gives good descriptions for spin coherence generation for QDs in any excitation power regime and for QWs in the low power excitation regime.

B. Generation of long-lived spin coherence during the trion life time. Spin dynamics of charged carriers in magnetic field

1. Spin dynamics of resident carrier and trion

Right after the excitation pulse the coupled dynamics of resident carrier spin, $\mathbf{S}$, and trion spin, $\mathbf{S}^T = (S_x^T, S_y^T, S_z^T)$, can be described by the following system

$$\begin{align*}
\dot{S}_x^a &= \mp \frac{1}{2} Q^2 \sin \Phi S^b_y + Q^2 \cos \Phi S^b_x, \\
\dot{S}_y^a &= \mp Q^2 \sin \Phi S^b_x - Q^2 \cos \Phi S^b_y, \\
\dot{S}_z^a &= \mp Q^2 \sin \Phi S^b_x + Q^2 \cos \Phi S^b_y,
\end{align*} \quad (1b)$$

where $P$ is the pump pulse power. One of the main predictions of the considered quantum mechanical approach is that for high pump powers the electron spin $z$ component depends periodically on the pump area, i.e., shows Rabi oscillations consistent with the classical approach.

For low pump powers and resonant trion excitation the electron spin coherence increases linearly with the pump power, see Eq. (3), while at high powers the spin $z$ component saturates and Rabi oscillations are not observed [28]. Clearly, the two-level model is not sufficient for describing such a behavior. The most probable reason is related to the weaker localization of electrons and trions in quantum wells and, hence, to the presence of many trion states. Scattering between these states becomes possible, as schematically illustrated in Fig. 1(b). The optical coherence of the trion with the pump is lost due to this scattering, while spin coherence is preserved. As a result, if the scattering time between different trion states, $\tau_1$, is considerably shorter than $\tau_p$, the Rabi oscillations at high pump powers vanish [28], because the population of the excited state of the two-level system coupled to the optical pulse is small. At the same time, the spin polarization generated by the pump pulse can be substantial, because spin does not relax during scattering. With an increase of the pump pulse power the electron spin saturates at the value

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As will be shown below in Sec. III, the quantum mechanical and classical approaches give the same results at low pump powers. Subsequently, we will use the quantum mechanical approach because it gives good descriptions for spin coherence generation for QDs in any excitation power regime and for QWs in the low power excitation regime.
of equations \([4, 13, 23, 26]\):

\[
\frac{dS^T}{dt} = \frac{\mu_B}{\hbar} [gT B \times S^T] - \frac{S^T}{\tau_s} - \frac{S^T}{\tau_r},
\]

\[
\frac{dS}{dt} = \frac{\mu_B}{\hbar} [gB \times S] - \frac{S}{\tau_s} + \frac{S^T e_z}{\tau_r}.
\]

Here \(e_z\) is the unit vector along the \(z\) axis. The magnetic field \(B\) is assumed to be parallel to the \(z\) axis, \(\tau_s^T\) is the trion spin relaxation time, \(\tau_s\) is the phenomenological spin relaxation time of the resident carrier \([24]\), and \(\tau_r\) is the trion radiative lifetime. It is worth to mention that carriers left behind after trion recombination are polarized parallel or antiparallel to the \(z\) axis due to the optical selection rules, see the last term \(\propto S^T e_z\) in Eq. \(\eqref{eq:5}\).

From Eqs. \(\eqref{eq:5}\) the carrier spin projection onto the magnetic field, \(S_z\), is conserved. Introducing the trion spin lifetime, \(\tau_r = \tau_s^T / (\tau_s^T + \tau_r)\), we arrive at the following expression for the transverse carrier spin component

\[
S_+(t) = S_{+0} e^{-i\omega t - t / \tau_s} + S_{+0}^T \left[ -\xi e^{-i\omega t - t / \tau_s} + e^{-i / \tau_r} (\xi \cos \Omega t + \chi \sin \Omega t) \right].
\]

Here the subscript 0 denotes the components at time \(t = 0\), when the pump pulse is finished, e.g., \(S_{+0} = S_z(0) + iS_y(0)\).

\[
\xi = \xi_1 + i\xi_2 = \frac{\omega - \gamma - 1}{\gamma \tau_r[(1 - \omega / \gamma) + (\Omega / \gamma)^2]},
\]

\[
\chi = \chi_1 + i\chi_2 = \frac{\Omega / \gamma}{\gamma \tau_r[(1 - \omega / \gamma) + (\Omega / \gamma)^2]},
\]

and \(\gamma = \tau_r^{-1} - \tau_s^T > 0\).

In order to have a closed equation system \([4]\), we have to relate the carrier and trion spins at \(t = 0\). This can be done through Eqs. \(\eqref{eq:1}\) and \(\eqref{eq:2}\). After a single pump pulse \((S^T = 0)\) one has

\[
S_{z0}^T = -S_{z0}.
\]

The first term in the right hand side of Eq. \(\eqref{eq:5}\) describes the carrier spin precession. The term proportional to \(S_{z0}^T e^{-i\omega t}\) describes the spin polarization of the resident carrier after trion recombination. Below, we consider the relation of these two contributions as a function of spin system parameters and external conditions.

2. Effect of trion spin relaxation on spin coherence of resident carrier

In absence of an external magnetic field the efficiency of resident carrier spin generation is solely determined by the trion spin relaxation \([3, 13, 30]\). This becomes clear from Eq. \(\eqref{eq:4}\), which for \(B = 0\) reduces to

\[
S_z(t) = S_{z0} e^{-t / \tau_s} + S_{z0}^T \xi \left( e^{-t / \tau_s} + e^{-i / \tau_r} \right). \tag{9}
\]

It follows from Eq. \(\eqref{eq:4}\) that \(\xi = -(\tau_r \gamma)^{-1} \approx -1 + (\tau_s / \tau_s^T)\), provided that the carrier spin relaxation time exceeds by far both trion recombination time and trion spin lifetime. These conditions are readily fulfilled in experiment. Hence, the long-lived carrier spin coherence is given by

\[
S_z(t) = (S_{z0} - S_{z0}^T \xi) e^{-t / \tau_s}, \quad t \gg \tau_r. \tag{10}
\]

If spin relaxation in the trion is suppressed, i.e. \(\tau_s^T \gg \tau_r\), then \(\xi \to -1\). Therefore, since for a single pump pulse \(S_{z0}^T = -S_{z0}\), the contribution of the carrier left behind from the trion decay compensates exactly the spin polarization of the remaining, non-excited carrier component. As a result, no long-lived spin coherence for resident carriers is generated. In general, when the resident carrier has been polarized before pump pulse arrival, this carrier polarization will not be affected by the pump pulse and conserved after trion recombination. To conclude, trion spin relaxation is required to give rise to a non-zero long-lived spin coherence of the resident carriers in absence of a magnetic field.

3. Spin precession of resident carrier

The carrier spin precession about an external magnetic field results in an imbalance of resident and returning spins. Hence, long-lived spin coherence can be excited even in the absence of trion spin relaxation. Provided that the trion spin does not precess \([31]\), \(\Omega = 0\), the long-lived carrier spin coherence is given by \([4]\)

\[
S_z(t) = \text{sign}(S_{z0})|S_{z0} - S_{z0}^T \xi| e^{-t / \tau_s} \cos(\omega t - \phi), \quad t \gg \tau_T \tag{11}
\]

where \(\phi\) is the initial phase, which can be related to the parameter \(\xi\), see Ref. \([4]\) for details. Note, that in Ref. \([4]\) the phase is shifted by \(\pi/2\) with respect to our definition in Eq. \(\eqref{eq:11}\). The amplitude of the long-lived spin coherence \(A_\phi\) after a single pump pulse can be recast as

\[
A_\phi = |S_{z0} - S_{z0}^T \xi| = |S_{z0}(1 + \xi)| \approx |S_{z0}| \frac{|\omega \tau_r|}{\sqrt{1 + (\omega \tau_r)^2}}, \tag{12}
\]

where the latter approximate equality is valid for a long spin relaxation time fulfilling the relation \(\tau_s^T \gg \tau_r\). According to Eq. \(\eqref{eq:12}\) the long-lived spin amplitude first increases with growing magnetic field \(\propto \omega \tau_r\) and then saturates in strong fields.

The general case of arbitrary \(\omega \tau_r\) and \(\tau_s^T / \tau_r^T\) is illustrated in Fig. \(\text{2}\). Panel (a) demonstrates the dependence of the long-lived spin coherence amplitude \(A_\phi\) on magnetic field (expressed as \(\omega / \gamma\)) for different values of the ratio \(\tau_s^T / \tau_r^T\). Depending on the parameter \(\tau_s^T / \tau_r^T\), the change of amplitude \(A_\phi\) as a function of magnetic field (through \(\omega \sim B\)) occurs for different field values since \(\gamma\) itself is determined by \(\tau_r\) and \(\tau_s^T\). The panels (b) and (c) in Fig. \(\text{2}\) show the carrier spin coherence \(S_z(t)\) calculated for fast \((\tau_s^T / \tau_r^T = 10)\) and slow \((\tau_s^T / \tau_r^T = 0.01)\) spin relaxation of the trion. The solid and dashed lines show \(S_z(t)\) in zero and finite magnetic field, respectively. One can see from Fig. \(\text{2b}\), that at \(\tau_s^T / \tau_r^T = \)
the maxima of the solid line, (240). As one can see in Fig. 2(c) the dashed line at zero magnetic field leads to the coincidence of the amplitudes of long-lived spin coherence. This is, however, not the case for the smaller ratio of \( \tau_0/\tau_s = 0.01 \) (\( \omega/\gamma = 240 \)). As one can see in Fig. 2(c) the dashed line at zero magnetic field leads to the coincidence of the amplitudes of long-lived spin coherence, \( A_s(0) \), with the maxima of the oscillating solid line (finite field, \( \omega/\gamma = 0.24 \)). In other words, the application of magnetic field here does not change the efficiency of spin coherence generation. This is, however, not the case for the smaller ratio of \( \tau_0/\tau_s = 0.01 \) (\( \omega/\gamma = 240 \)). As one can see in Fig. 2(c) the dashed line at longer delays has considerably smaller amplitude than the maxima of the solid line, \( A_s(0) \ll A_s(240) \). This means that the amplitude of long-lived spin coherence, \( A_s \), can be strongly increased by external magnetic fields. To conclude, even in the absence of spin relaxation in the trion the application of an external magnetic field leads to appearance of long-lived spin polarization of the resident carriers.

4. Effect of spin precession in trion on spin coherence of resident carrier

Spin precession of the trion, characterized by the frequency \( \Omega \), also provides a mechanism for generating long-lived carrier spin coherence. Although the in-plane hole g factor in quantum wells and in self-assembled quantum dots is rather small \([32, 34]\), the spin precession of the hole in the T- trion may become important in tilted magnetic fields \([33] \), and in the case of the T+ trion excited in p-doped structures \([36] \).

Allowing for \( \Omega \neq 0 \) results in the following expression for the amplitude of the long-lived spin coherence \( A_s \) \( [c.f. Eqs. (11) and (12)]:

\[
A_s = |S_{z,0} - S_{z,0}^T \xi| = |S_{z,0}(1 + \xi)| \approx |S_{z,0}| \frac{(\Omega \tau_s)^2}{1 + (\Omega \tau_s)^2}
\]

where in the latter equality we assume a trion spin relaxation time, \( \tau_s^T \gg \tau_s \), and neglect the resident carrier spin precession, \( \omega \ll \Omega \). It follows from Eq. (13) that the spin precession in the trion acts similar to the trion spin relaxation. Here it does not matter whether the spin of the unpaired carrier in the trion was rotated by the magnetic field or flipped due to spin relaxation: in both cases long-lived carrier spin polarization arises.

The situation becomes richer when spin precession of both resident carrier and trion occurs. Figure 3 shows the spin dynamics of T- trion and resident electron [panel (a)], n-type and T+ trion and resident hole [panel (b), p-type]. The black curves give the difference \( S_{z,0}^T - S_z \). The gray (red) curves give only the trion contribution to the signal, \( S_{z,0}^T \).

![Figure 2](image_url)  
**Figure 2:** (Color online) (a) Dependence of the long-lived spin coherence amplitude \( A_s \) on the carrier Larmor precession frequency for different values of \( \tau_0/\tau_s^0 \). (b), (c) Carrier spin coherence \( S_z(t) \) normalized to \( S_z(0) \) for two different values of \( \tau_0/\tau_s^0 \). The spin dynamics at zero magnetic field are shown by the dashed lines. The solid lines show \( S_z(t) \) at finite magnetic field (\( \omega\tau_s = 2.4 \)). The arrows show the corresponding amplitudes \( A_s(\omega/\gamma) \) for these conditions.

![Figure 3](image_url)  
**Figure 3:** (Color online) Spin dynamics of resident carriers and trions for (a) negatively charged trions T-, n-type and (b) positively charged trions T+, p-type. The black curves show the temporal evolution of \( S_{z,0}^T - S_z \). The gray (red) curves give only the trion contribution to the signal, \( S_{z,0}^T \).
C. Spin accumulation induced by a train of pump pulses

In experiments on coherent spin dynamics periodic trains of pump pulses are commonly used. When the spin relaxation time of the resident carrier is comparable or longer than the repetition period of the pump pulses, i.e. \( \tau_s > T_R \), the steady-state carrier spin polarization results from the cumulative contribution of multiple pump pulses. In external magnetic fields applied in the Voigt geometry, the steady-state situation is reached for each result from the cumulative contribution of multiple pump pulses. When the spin precession period of the resident carrier is comparable with the pump pulse repetition frequency, the steady-state carrier spin polarization is not efficient, as seen from the comparison of the amplitudes in Figs. 4(a) and 4(b).

Eq. (14) should be averaged over their distribution \( \text{PSC} \), see below Sec. IV D and Eq. (28).

\[
S_z^b(\omega) = \pm \frac{1}{2} \frac{K}{1 + Qe^{-2\omega T_R/\tau_s} - e^{-\omega T_R/\tau_s}(1 + Q)\cos(\omega T_R) - K},
\]

where the signs \( \pm \) correspond to different polarizations of optical pumping and different types of resident carriers, cf. Eqs. (1), and

\[
K = \frac{(1 - Q^2)e^{-\omega T_R/\tau_s}}{2} \left\{ (1 + \xi_1)(Qe^{-\omega T_R/\tau_s} - \cos(\omega T_R)) - \xi_2\sin(\omega T_R) \right\}.
\]

Equation (14) shows that the spin z component before the next pump pulse arrival, \( S_z^b \), is a periodic function of magnetic field (see Fig. 5) with maxima of \( |S_z^b| \) at frequencies \( \omega \) satisfying the phase synchronization condition (PSC) \( [5, 14, 37] \):

\[
\omega = N\omega_R = \frac{2\pi N}{T_R}, \quad N = 0, 1, 2, \ldots
\]

Here \( \omega_R = 2\pi/T_R \) is the repetition frequency of the pump pulses. Indeed, one can see from time-resolved signals shown in Fig. 4 if the spin precession period of the resident carrier is commensurable with the pump pulse repetition period, then the spin coherence generated by the pump is always in phase with that from the previous pulse [see signal around zero time delay, Fig. 4(a)], and carrier spin polarization is accumulated. Let this phase, \( \phi \), be zero. Otherwise, if the spin precession and pump repetition periods are not commensurable, the accumulation of spin polarization is not efficient, as seen from the comparison of the amplitudes in Figs. 4(a) and 4(b).

In general, the electron spin precession has a particular phase, see Eq. (11), which we determine here as the difference \( (\omega T_R - 2\pi N) \), where \( N \) is the largest integer satisfying the condition \( (\omega T_R - 2\pi N) \geq 0 \). The phase can be expressed as:

\[
\cos(\phi) = -S_{zb}^b/\sqrt{(S_{zb}^b)^2 + (S_{yb}^b)^2},
\]

\[
\sin(\phi) = S_{yb}^b/\sqrt{(S_{zb}^b)^2 + (S_{yb}^b)^2}.
\]

Note, that in Fig. 4 and further on in this paper we show for convenience the inverted signal \(-S_z \) (in order to have positive signals for \( \sigma^+ \) pumping). This sign change does not affect the obtained results but is more suitable for their graphic presentation. For an ensemble of resident carriers with different spin precession frequencies, \( \omega \), precessing spin by relatively long trains of pump pulses: the decay of the spin polarization is then balanced by the pumping. As a result, the carrier spin after each repetition period, \( S(T_R) \), given by Eq. (11), should be equal to the carrier spin right before the pump pulse arrival, which we denote by \( S^0 \) (see Fig. 4). Using the connection between the carrier spins before and after the pump pulse, Eq. (11), and assuming that the pump pulse is resonant with the trion transition, \( \Phi = 0 \), one immediately comes to the following expression for the carrier spin z component before pump pulse arrival:

\[
S_z^b(\omega) = \pm \frac{1}{2} \frac{K}{1 + Qe^{-2\omega T_R/\tau_s} - e^{-\omega T_R/\tau_s}(1 + Q)\cos(\omega T_R) - K},
\]

Figure 4: (Color online) Dependencies of resident carrier spin polarization \( S_z \) on pump-probe delay for a carrier spin precession frequency which is (a) commensurable with the pump pulse repetition frequency \( \omega = 2\omega_R \) and (b) not commensurable with this frequency \( \omega = 2.5\omega_R \). Parameters of calculations are: \( \tau_s = 3T_R \), \( \Theta = 0.1\pi \). Thick vertical arrows show the arrival times of the pump pulses. Phase \( \phi \) of the oscillating polarization, \(-S_z\), is \( \phi = 0 \) in panel (a) and \( \phi = \pi \) in panel (b).

Figure 5 shows \( S_z^b \) calculated after Eq. (14) for different pump pulse areas \( \Theta \) in the case of fast trion spin relaxation. As x axis scale in Fig. 5 we take the ratio of spin precession frequency \( \omega \) and \( \omega_R \), which represents...
the resident carrier spin polarization $S_z^b$ and its phase on magnetic field expressed by $\omega/\omega_T = g_\nu B/(h\omega_T)$. Data are shown for zero time delay (right before the pump pulse arrival), calculated for different ratios $\tau_s/T_R$ at $\Theta = 0.1\pi$ (a,c) and for different pump pulse areas $\Theta$ at $\tau_s/T_R = 3$ (b,d).

Figure 5: (Color online) Dependence of the resident carrier spin polarization $S_z^b$ and its phase on magnetic field expressed by $\omega/\omega_T = g_\nu B/(h\omega_T)$. Data are shown for zero time delay (right before the pump pulse arrival), calculated for different ratios $\tau_s/T_R$ at $\Theta = 0.1\pi$ (a,c) and for different pump pulse areas $\Theta$ at $\tau_s/T_R = 3$ (b,d).

The phases of the signals from Figs. 5(a) and 5(b) are shown in panels (c) and (d), respectively. One clearly sees the zeros of the phase correspond to maxima of spin polarization, $-S_z^b$, and the values $\phi = \pm \pi$ correspond to its minima.

One should note, that the magnitude of the accumulated spin polarization, as well as the width of the resonance peaks in the magnetic field dependence of $-S_z^b$, are determined not only by the pump pulse power and the carrier spin relaxation time, but also by the mechanism of long-lived spin coherence generation and the spin dephasing time. We present the analysis of these effects in the following Sections.

III. RESONANT SPIN AMPLIFICATION

We begin with the classical expression for carrier spin polarization under RSA conditions [18, 39]. The underlying assumptions are the following: (i) only carrier spin polarization is considered, and (ii) it is supposed that each pump pulse generates only a $z$ component of spin polarization, whose magnitude is $S_0$. All non-additive effects of the pump pulse [28] are disregarded. After single pump pulse excitation the carrier spin dynamics are described by a decaying cosine function periodic with the Larmor precession frequency $\omega$ and decay with time $\tau_s$. The effect of a long train of pump pulses on the carrier spin polarization can be calculated as:

$$S_z(\omega, t) = \sum_{k=0}^{\infty} S_0 e^{-(t+kT_R)/\tau_s} \cos[\omega(t+kT_R)],$$

where $t$ is the pump-probe delay and $k = 0, 1, 2, \ldots$. This equation can be rewritten [37, 38] as:

$$S_z(\omega, t) = \frac{S_0}{2} \sum_{k=0}^{\infty} e^{-t/\tau_s} \cdot e^{-T_R/\tau_s} \cos[\omega(t+kT_R)] = \cosh(T_R/\tau_s) - \cos[\omega(t+T_R)].$$

It follows from Eq. (15) that for sufficiently long decay times $\tau_s \geq T_R$ the carrier spin has sharp resonances as a function of magnetic field. As will be shown by our calculations, this corresponds to the solid line in Fig. 5(a) and gives the RSA signals presented in Fig. 5. The peak positions at zero pump-probe delay correspond to spin precession frequencies which are commensurable with the pump repetition frequency $\omega_R = 2\pi/T_R$. The expression (13) near commensurable frequency $(|\omega T_R - 2\pi N| \ll 1)$ and at a zero time delay can be written as:

$$S_z^b \sim \frac{1}{(\omega T_R - 2\pi N)^2 + (T_R/\tau_s)^2}.$$

Here we assume that $T_R/\tau_s \ll 1$. The peak width is determined by the relaxation time of the electron spin polarization. Note, that for the spin ensemble the time $\tau_s$ should be changed to the dephasing time $T_2^\rho$ [39]. This allows one to measure spin relaxation and spin dephasing times exceeding $T_R$, i.e., for conditions where direct determination by time-resolved methods becomes inapplicable. The equations (13) and (14) describe a number of experiments well, see, e.g., Refs. [28, 37, 40, 41], and facilitate evaluation of carrier $g$ factors and spin dephasing times [39].

However, one sees that the spin polarization in Eqs. (13) and (14) increases to infinity if $\tau_s$ becomes larger and larger. Moreover, such an approach disregards completely the spin dynamics of trions and the specifics of carrier spin dephasing in external magnetic fields. This case requires a special treatment. There are also experiments which reveal a complicated shape of RSA spectra or a complete absence of RSA despite of very long spin relaxation times, which cannot be described by this simple model [37, 42]. The general analysis required for such cases is presented below.

A. Fast spin relaxation in trion

If the spin relaxation of the unpaired carrier in the trion is fast, $\tau_s^T \ll \tau_r$, the trion spin dynamics does not
affect the spin polarization of the resident carrier, see Sec. II B 2. In this case the carrier polarization induced by the pump pulse is not compensated by the carriers left after trion recombination, as these carriers are unpolarized. Then χ = 0 and the parameter K in Eq. (14) has the simple form [13, 23]

\[ K = \frac{(1 - Q^2)e^{-\frac{TR}{\tau_s}}}{2} \left[ Qe^{-\frac{TR}{\tau_s}} - \cos(\omega TR) \right]. \tag{20} \]

The detailed analysis of Eqs. (14) and (20) for this case is given in Refs. [14, 23]. If, moreover, the pump pulse area Θ is small, so that 1 - Q ≪ 1, Eq. (14) together with Eq. (20) go over into the classical expression of Eq. (18) for carrier spin polarization under RSA conditions.

It follows, that for frequencies near the phase synchronization condition of Eq. (14), the spin z component of the resident carrier can be recast as [23]:

\[ S^z_s \sim \frac{1}{(\omega TR - 2\pi N)^2 + (TR/\tau_s + (1 - Q))^2}, \tag{21} \]

where we assume that TR/τs ≪ 1, 1 - Q ≪ 1 and |ωTR - 2πN| ≪ 1. One sees from Eq. (21) that the RSA peak width is determined by TR/τs or 1 - Q, whichever is larger.

Figure 6 shows RSA signals calculated for a small pump power, Θ = 0.1π, at two different delays. The shape of the RSA signal at large negative delay (t = −0.1TR) differs from the one at zero delay due to the different phases of the spin precession.

An increase of the pump pulse area results in broadening of the RSA peaks, as was already shown in Fig. 6(b). For increasing pump pulse area the RSA peaks are no longer Lorentzians and, therefore, it is instructive to start from the situation in which the spin relaxation time can be comparable or even longer than its recombination time. It is worth to stress, that we can use the same system of equations (5) to describe the spin dynamics in n-type (resident electron and T− trion) and p-type (resident hole and T+ trion) structures. Figure 7(a) illustrates the situation that is typical for n-type QWs [31, 42], in which trion spin precession is absent. Figure 7(b) shows the RSA signal with a trion spin precession frequency Ω = 4ω, which may correspond to the T+ trion case in p-type QWs [36, 44]. The analysis shows that small Ω, i.e., \( \Omega \leq \omega \), leads to no significant changes of the RSA signal shape as compared with one in Fig. 7(b). A fast precession of the trion spin results in a faster appearance of long-lived spin coherence with increasing magnetic field, compare Figs. 7(a) and 7(b).

**B. Slow spin relaxation in trion: effect of trion spin dynamics**

Let us now turn to the general case, in which the trion spin relaxation time can be comparable or even longer than its recombination time. It is instructive to start from the situation in which t_{s}^{T} ≫ τ_r and long-lived spin coherence appears only due to carrier or trion spin precession about the magnetic field. Clearly, the peaks in the S^z_s(ω) dependence are suppressed for ωτ_r, Qτ_r ≪ 1 due to inefficient spin generation, and they increase significantly with an increase of magnetic field. This is illustrated in Fig. 7(c) where the calculated RSA signals are shown for t_{s}^{T} = 30τ_r. Note, that such unusual RSA spectra with suppression of the peak amplitudes in weak magnetic fields have been observed experimentally in both n-type and p-type QWs [22, 36, 42, 44].

**C. Effect of spin relaxation anisotropy**

To make our analysis of RSA complete, we briefly discuss here another effect, which is relevant for weak magnetic fields. It addresses the situation in which the carrier spin relaxation or the dephasing times are anisotropic. Spin relaxation anisotropy is an inherent feature of semiconductor quantum wells [45, 49]. For simplicity, we con-
Figure 7: (Color online) Impact of slow spin relaxation of the unpaired spin in the trion: RSA signals at zero delay without (Ω = 0) and with (Ω = 4ω) trion spin precession (panels (a) and (b), respectively). Parameters of calculations: τs,z = 30τs, τs = 0.01τR, τs = 3τR and ωτs = 4.4 at B = 1 T and Θ = 0.1π.

Figure 8: Effect of an anisotropy of the carrier spin relaxation times. The electron spin z component right before pump pulse arrival (t = 0) is calculated as function of magnetic field after Eq. (22). τs,z = 4τR.

D. Spin decoherence and dephasing

The spin relaxation time of localized carriers can be extremely long reaching up to microseconds for electrons in QDs, for example [51]. This is related with quenching of the orbital motion and the corresponding suppression of spin relaxation mechanisms contributed by spin-orbit coupling [52, 53]. The coherence time of an individual spin is typically much longer compared with the spin dephasing time of an inhomogeneous spin ensemble. The inhomogeneity, which leads to a spread of carrier spin precession frequencies, results in spin dephasing characterized by the T2* dephasing time. This time measured, e.g., from the decay of spin beats in external magnetic field is in the few nanoseconds range for QD ensembles [14, 15, 54] and in the tens of nanoseconds range for QWs containing diluted carrier gases [9, 22, 50, 40].

One of the main origins for the inhomogeneity of a spin ensemble is related to the g factor spread of localized carriers. For electrons the g factor variation can arise from changes of the effective band gap for different localization sites [14, 54, 56]. For localized holes the variations are mainly related to changes in the mixing of heavy-hole and light-hole states [57]. The spread of g factors in a spin ensemble, Δg, is translated into a spread of spin precession frequencies, Δωg, and, therefore, results in a spin dephasing rate [38, 40]

\[ \frac{1}{T_{2,\Delta g}^*} \sim \frac{\Delta g \mu_B B}{\hbar} = \Delta \omega_g, \]  

which is accelerated with increasing magnetic field.

Another origin of spin dephasing typical for electrons is related to random nuclear fields in the quantum dots [58]. Each localized electron is subject to a hyperfine field of a particular nuclear spin fluctuation, Bn, and, therefore, precesses about this field at a frequency ωn. These fluctuations are different for localization sites, causing dephasing of the electron spin ensemble. The dephasing
rate can be estimated by the root mean square of the electron spin precession frequency in the field of frozen nuclear fluctuations [33]:

\[
\frac{1}{T_{2,n}^*} \sim \sqrt{\langle \omega_n^2 \rangle}.
\]  

(25)

Assuming a normal distribution of \( B_n \), Eq. (25) can be rewritten as:

\[
\frac{1}{T_{2,n}^*} \sim \frac{g\mu_B \Delta B}{\hbar} = \Delta \omega_n.
\]  

(26)

where \( \Delta B \) is the dispersion of the nuclear spin fluctuation distribution [33].

Estimates show that \( T_{2,n}^* \) is on the order of several nanoseconds for GaAs quantum dots [58, 59]. Hence, in weak magnetic fields (e.g., \( B \lesssim 0.3 \) T for \( g = 0.5 \) and \( \Delta g = 0.005 \) [43]) the spin beat decay for resident electrons is determined by the hyperfine interaction, and in higher fields the dephasing is caused by the spread of \( g \) factors [60].

In quantum wells with a diluted electron gas the electron localization on well width fluctuations is considerably weaker compared to the QD case. As a result, \( \Delta g \) is smaller and the hyperfine interaction is weaker. Therefore, the spin dephasing times can reach \( \sim 30 \div 50 \) ns in weak magnetic fields and at low temperatures [44]. Below the effect of a spin precession frequency spread on RSA signals is analyzed.

1. Spread of \( g \) factors

For a more realistic approach we need to take into account the precession frequency spread, \( \Delta \omega \), in the spin ensemble. Here for distinctness we consider only the frequency spread caused by \( \Delta g \) (the spread related with the nuclear spin fluctuations is considered below). For ensemble of carrier spins with a spread of \( g \) factors, \( \Delta g \), the spread of Larmor precession frequencies, \( \Delta \omega_g \), is proportional to the magnetic field:

\[
\Delta \omega_g(B) = \Delta g \mu_B B / \hbar
\]  

(27)

To model the ensemble RSA signal one has to sum the contributions of the individual spins [38] over the \( g \) factor distribution function:

\[
\rho(g) = \frac{1}{\sqrt{2\pi \Delta g}} \exp \left[ -\frac{(g - g_0)^2}{2(\Delta g)^2} \right],
\]  

(28)

where \( g_0 \) is the average \( g \) factor value in the spin ensemble, resulting in an average Larmor frequency: \( \omega_0 = g_0 \mu_B B / \hbar \).

RSA spectra calculated by means of Eqs. (14) and (28) for short trion spin relaxation, i.e. dependencies of the carrier spin polarization, \( -S_z \), on magnetic field in terms of \( \omega_0 / \omega_R \) are shown in Fig. [4] for two negative time delays. An increase of magnetic field leads to broadening of the RSA resonances and decrease of their amplitudes. This reflects the acceleration of the spin dephasing rate \( 1/T_{2,R}^* \sim B \), in accordance with Eq. (24).

Figures (10a) and (10b) show RSA signals for long trion spin relaxation, \( \tau^*_T = 30 \tau_T \), with and without spin precession in the trion. An ensemble spread of \( \Delta \omega_g = 0.02 \omega_0 \) results in a broadening of the RSA peaks and a decrease of their amplitudes with increasing magnetic field, similar to Fig. [9]. This results in the characteristic bat-like shape of the RSA signal [22, 50, 42, 44] compared with Fig. [4] where the spin dephasing was absent, \( \Delta \omega_g = 0 \). Accounting for the spread of \( \Omega \) does not change the signals significantly.

Figure (10a) corresponds to a situation that is obtained for resident electrons oriented by excitation of the \( T^- \) trion in \( n \)-type (In,Ga)As/GaAs QWs [22, 42]. In such structures the in-plane hole factor is small compared with the electron \( g \) factor, and consequently \( \Omega \ll \omega \), so that the spin precession of the \( T^- \) trion can be neglected.

Figure (10b) corresponds to the long-lived hole spin orientation for excitation of the \( T^+ \) trion in \( p \)-type GaAs/(Al,Ga)As QWs [38]. For the \( T^+ \) trion the ratio \( \Omega / \omega \) is opposite, i.e. \( \Omega \gg \omega \). In Ref. [45] \( \Omega = 4.5 \omega \) and the spin precession in trion affects the RSA signal.

The results of the calculations shown in Figs. (10a) and (10b) are in good agreement with available experimental data for quantum well structures [22, 50, 42]. All calculations were done for a small pulse area \( \Theta = 0.1 \pi \). The analysis of the case of high pump power, which results in saturation effects, shows that an increase of the pump power results in an increase of the signal amplitude and broadening of all peaks, similar to the case discussed in Sec. (11A) see also Fig. [5]. The bat-like shape of the RSA signal envelope is conserved even for \( \Theta = \pi \) pump pulses.
2. Nuclear field fluctuations and resonant spin amplification in weak magnetic fields

Interaction of the nuclear spins with hole spins is weak and in many cases can be neglected. At the same time, for localized electrons the hyperfine interaction with the nuclei can considerably contribute to the spin dynamics. Therefore, in this subsection we will focus on n-type structures containing resident electrons.

In weak magnetic fields the electron spin dephasing time related to the spread of g factor values, Eq. (24), proportional to $1/B$, becomes very long and nuclear field fluctuations play an important role. The hyperfine fields acting on the electrons due to these nuclear fluctuations can be as large as $B_n \sim 0.5$ mT for GaAs QWs [30] and an order of magnitude larger in (In,Ga)As QDs [61].

For $B \gtrsim B_n$ the only important component of the nuclear field fluctuation is the one parallel to the external field $B$. It results in a spread of Larmor precession frequencies, damping of the spin beats and broadening of the RSA peaks, provided $B_n > |\Delta g/g|B$.

The situation becomes different in weak magnetic fields $B < B_n$. In this case all components of the nuclear fluctuation field become important. For illustration we consider a homogeneous electron spin ensemble ($\Delta g = 0$) in a magnetic field which is the sum of the external magnetic field $B$ and the fluctuation field $B_n$. For simplicity, we consider the regime of fast spin relaxation in the trion ($\tau_s^T \ll \tau_s$). To model the dynamics of the electron spin ensemble one can assume a normal distribution of $B_n$:

$$\rho_n(B_n) = \frac{1}{(\sqrt{2\pi}\Delta B)^3} \exp \left( -\frac{B_n^2}{2(\Delta B)^2} \right),$$

where $\Delta B$ is the isotropic dispersion of the nuclear fluctuation field distribution ($\Delta B_x = \Delta B_y = \Delta B_z$). The spread of the Larmor precession frequencies, $\Delta \omega_n$, does not depend on the external magnetic field:

$$\Delta \omega_n = g \mu_B \Delta B / \hbar.$$  \hspace{1cm} (30)

The average Larmor frequency of the spin ensemble in this case is equal to the spin precession frequency in an external magnetic field without nuclear fluctuations: $\omega_0 = g \mu_B B / \hbar$.

Figure 11 shows RSA signals at zero time delay averaged over $B_n$ for different $\Delta B$ values. One sees that indeed, an increase of the frequency spread $\Delta \omega_n$ leads to an increase of the dephasing rate evidenced via broadening of the RSA peaks. For weak magnetic fields $B < \Delta B$ the $y$ component of the nuclear fluctuation field, $B_{n,y}$, which is perpendicular to $S_z$ and to the external field, can additionally destroy the long-lived carrier spin polarization. This is manifested in an additional broadening and a decrease of the amplitude of the zeroth RSA peak (compared to the ±1 peaks), as is clearly seen in Fig. 11(a,b). The enhancement of $S_z$ in the vicinity of zero field for large fluctuations, see Fig. 11(c), is due to the fact that the $z$ component of the spin polarization can not destroy by a parallel component of the nuclear fluctuation field $B_{n,z}$.

E. Analysis of RSA signals and evaluation of spin dephasing times and $g$ factors

To conclude our analysis of RSA we emphasize that in spite of the possibly complex shape of RSA signals, especially in case of a long spin relaxation in the trion, the analysis allows one to obtain various parameters with high accuracy. This is due to the fact that these parameters are responsible for different features in the RSA spectrum:
• the $g$ factor of the resident carriers gives the magnetic field positions of the RSA peaks.

• The $g$ factor spread, $\Delta g$, determines the amplitude decrease of the RSA peaks with increasing magnetic field.

• The spin relaxation/dephasing time $\tau_s$ is related to the RSA peak widths $12$.

• The ratio of spin relaxation time $\tau_s^T$ and radiative lifetime $\tau_r$ of the trion determines the possible increase of RSA peak amplitudes with increasing magnetic field. If $\tau_r$ is obtained from an independent time-resolved measurement, then $\tau_s^T$ can be extracted from fitting the RSA spectrum.

• For long spin relaxation in the trion (when the RSA signal has a bat-like shape) the symmetry of the RSA peaks at zero pump-probe delay can indicate the fact that the trion $g$ factor is larger than that of the resident carrier ($|gT| \gg |g|$). However, the value of the trion $g$ factor should be obtained from another experiment.

• Finally, the amplitude and the width of the zero-field RSA peak can contain information on the anisotropy of the spin relaxation of delocalized carriers and the nuclear effects for localized carriers.

The spin dynamics parameters considered above can be extracted only for sufficiently homogeneous ensembles and at weak excitation powers (small pump pulse areas), which is typical for semiconductor QWs.

It is worth to mention, that there are other generation mechanisms of long-lived spin coherence for nonresonant optical excitation $8, 22, 44$. In this case, the RSA signal can change its shape dramatically. However, a detailed analysis allows one to identify the generation and relaxation mechanisms of carrier spin polarization and obtain the corresponding quantitative information about relaxation processes.

IV. MODE-LOCKING OF CARRIER SPIN COHERENCES

Now we turn to strongly inhomogeneous spin systems, for which the spread of the spin precession frequencies is so large that

$$ T_s^2 < T_R. \quad (31) $$

Still, the spin relaxation time of the resident carrier is assumed to exceed by far the repetition period, $\tau_s \gg T_R$. In this case the ensemble spin polarization generated by a pump pulse decays within the time $T_s^2$, i.e., disappears before the next pump pulse arrival. Figure 12 presents model calculations, which show the dynamics of the carrier spin polarization excited by a train of the pump pulses. Indeed, the polarization decays quite rapidly after the pump pulses, but thereafter reemerges at negative delays $-T_s^2 \lesssim t < 0$. Such a behavior has been explained in terms of mode-locking of carrier spin coherences that are synchronized by the periodic train of pump pulses $14 \ [20]$.

If the condition $31$ is fulfilled, the pump pulse excites a broad distribution of spin precession frequencies, among which there are several frequencies satisfying the phase synchronization condition of Eq. $15$. The carrier spins with such precession frequencies are excited much more efficiently, i.e., accumulate more spin polarization than the other ones. As a result, the main contribution to the signal is given by the commensurable spin beat frequencies. In other words, the spins satisfying the PSC become resonantly amplified, while others are not, and the synchronized spins contribute mostly to the experimentally measured signal of carrier spin polarization. Such behavior of the spin signals, characteristic for the mode-locking of carrier spin coherences, has been observed in $n$-type singly-charged (In,Ga)As QDs $14 \ [20] [21]$.

The calculations shown in Fig. 12 are carried out after Eqs. $14$ and $50$ assuming, for simplicity, that the trion spin relaxation is fast, $\tau_s^T \ll \tau_r$, and the spread of the carrier spin precession frequencies $\Delta \omega = \omega_R$ does not depend on the magnetic field strength.

Let us have a closer look on the signals in Fig. 12. It is remarkable, that the phase of the spin beats before the next pump pulse arrival is fixed for any magnetic field. The average precession frequency of spin ensemble, $\omega_0$, satisfies the PSC in Fig. 12(a) while it does not in Fig. 12(b). The phase, however, in both cases is exactly the same and it also coincides with the one after the pump pulse, $\phi = 0$. This is in strong contrast with the regime of weak dephasing ($T_s^2 \gtrsim T_R$), see Fig. 12(c), and can be considered as the principle difference of the SML and RSA regimes of carrier spin accumulation. Note that

\[
\begin{align*}
\text{(a)} & \quad \omega_0 = 10 \omega_R & \quad T_R \\
\text{(b)} & \quad \omega_0 = 7.5 \omega_R \\
-1.0 & \quad -0.5 & \quad 0.0 & \quad 0.5 & \quad 1.0 \\
-0.2 & \quad 0.0 & \quad 0.2 & \quad 0.4 \\
-0.2 & \quad 0.0 & \quad 0.2 & \quad 0.4 \\
\end{align*}
\]

Pump - probe delay, $t/T_R$
the regime of weak dephasing is similar to the dynamics of a single spin presented in Figs. 4 and 5.

It is worth to mention, that the ratio of the signal amplitudes at negative and positive delays depends strongly on the generation efficiency and conservation of spin polarization, i.e., on the pump pulse area, the trion spin relaxation, and the ratio of carrier spin relaxation time $\tau_s$ to $T_R$ \cite{14, 20}.

V. RSA VERSUS MODE-LOCKING

In this Section we discuss how one can distinguish the RSA and SML regimes and what parameters are responsible for separating these regimes. This separation is based on the common basic mechanism of the RSA and SML effects, which is the accumulation of carrier spin polarization under periodic pump pulse excitation. The key difference between the regimes is the ratio of the Larmor frequency broadening to the repetition frequency of the pump pulses: $\Delta \omega/\omega_R$. This is schematically illustrated in Fig. 13(a,b) by the frequency spectrum of the spin ensemble in a finite magnetic field. Here few PSC modes satisfying Eq. 14 from $(N-2)\omega_R$ to $(N+2)\omega_R$ are indicated in by the dashed vertical lines.

In the RSA regime $\Delta \omega \ll \omega_R$ and only one PSC mode (or even none) can fall into the distribution of Larmor frequencies. When the PSC mode coincides with the distribution maximum, as it is shown in Fig. 13(a), one obtains a peak in the RSA spectrum. And when the overlap between the mode and the distribution is absent the RSA spectrum has minimum.

For the SML regime involvement of at least two PSC modes is necessary. Therefore, the condition for this regime is $\Delta \omega \gtrsim \omega_R$, see Fig. 13(b). The calculations given in this Section show that in fact the transition to the SML regime happens already for $\Delta \omega \gtrsim 0.5\omega_R$, when the tails of the Larmor frequency distribution overlap with more than one PSC mode.

![Figure 13: Larmor frequency distribution function](image)

Figure 13: Larmor frequency distribution function (multiplied for convenience by $\sqrt{2\pi \Delta \omega}$) of a spin ensemble for RSA (a) and SML (b) conditions. (c) Parameter diagram showing schematically the regimes where RSA and SML occur, see text for details.

Deeper insight in the separation between the RSA and SML regimes is collected below in Figs. 14, 15, and 16. Here the carrier spin polarization amplitude, $S^b_z$, and the signal phase at zero negative delay are analyzed as functions of magnetic field, time delay, Larmor frequency spread, and pump pulse area. We also consider the effect of resident carrier spin relaxation taking it into account via the parameter $\tau_s/T_R$. For most figures a pump pulse area $\Theta = \pi$ is chosen as it provides efficient spin accumulation. Let us go step by step through this data set.

First, for demonstration purposes, we assume again that a spread of the carrier spin precession frequencies is $\Delta \omega = \omega_R$, and it does not depend on magnetic field. For $n$-type structures this corresponds to the case when the $\Delta \omega$ of the resident electrons is dominated by the random fields of the nuclear spin fluctuations: $\Delta \omega_m \propto B_{n,x}$. For $B > B_n$ only the $B_{n,x}$ component parallel to the external magnetic field should be considered, see Sec. III D 2.

Magnetic field dependencies of the carrier spin polarization, $-S^b_z$, and the signal phase are shown in Figs. 13(a) and 13(b) for different $\Delta \omega$ and $\tau_s/T_R = 300$. For a small frequency spread of $\Delta \omega = 0$ and $0.2\omega_R$ the polarization amplitude and phase are periodic functions of magnetic field, which is characteristic for the RSA regime, for comparison see Figs. 5(b) and 5(d). An increase of $\Delta \omega$ to $0.5\omega_R$ drastically changes the character of these functions: both of them become independent of magnetic field. The spin polarization amplitude has a finite value (in this case it is equal 0.08), while $\phi = 0$. These are characteristics of the SML regime.

Details of separating the RSA from the SML regime with increasing frequency spread are presented in Fig. 14(c). The peak amplitudes of the spin polarization at the PSC frequencies are plotted for different pump pulse areas there. The amplitude initially decreases with an increasing spread and approaches a saturation level for larger spreads. Independence of the amplitude on the spread is characteristic for the SML regime, therefore, one can see from the Fig. 14(c) that the regimes cross over at $\Delta \omega \sim 0.5\omega_R$.

The spin polarization amplitude in the SML regime depends critically on the pump pulse area, see also Fig. 14(d). It is close to zero for $\Theta < 0.3\pi$, but strongly increases for $\Theta$ exceeding this value, approaching a maximum at $\Theta = 2\pi$ for sufficiently large $\tau_s/T_R = 3000$. The dependence of $S^b_z$ for a large spread, which corresponds to a constant plateau level, can be written as:

$$S^b_z = \frac{1 - Q}{1 + Q} \left[ 1 - \frac{M^2 - 1}{L^2 - 1} \right], \tag{32}$$

where $M = Q e^{-T_R/\tau_s}$ and $L = e^{-T_R/\tau_s} (1 + Q^2)/2$. The calculations in Fig. 14(d) show that with increasing electron spin relaxation time $\tau_s$ the maximum signal amplitude shifts to a pulse area of $2\pi$ [unlike the dependence of spin polarization on pulse area for excitation by a single pulse, for which Rabi oscillations occur with maximum at $\Theta = \pi$].

The fact that the separation between RSA and SML is controlled by the ratio $\Delta \omega/\omega_R$ offers the instructive opportunity to realise a changeover between these two regimes by tuning the magnetic field. This would be possible for the case when the Larmor frequency spread is controlled by $\Delta \omega$, see Sec. III D 2, because in this case $\Delta \omega$ increases linearly with $B$. Results of corresponding
calculations for $\Delta \omega_g = 0.1 \omega_0$ are given in Fig. 15a. In analogy with Figs. 14(a) and 14(b), one can identify the RSA regime in low magnetic fields ($|\omega_0/\omega_R| < 3$), where both the polarization amplitude and the phase change with $B$, and the SML regime in larger magnetic fields ($|\omega_0/\omega_R| > 5$), where these parameters do not vary anymore.

Figure 15(c) shows the range of parameters in which the different spin accumulation regimes can be obtained. The dashed curve corresponds to the condition $\Delta \omega = 0.5 \omega_R$, which may serve as approximate boundary between the RSA and SML regimes. Indeed, if the $g$ factor spread is small, the spin frequency distribution contains only one phase synchronized mode in a broad range of magnetic fields, the latter are expressed via $\omega_0(B)/\omega_R$. It corresponds to the RSA regime for which the parameter space is placed below the dashed curve in Fig. 15(c). On the contrary, if the $g$-factor spread is large, several phase synchronized modes become involved already at weak magnetic fields and, for relatively efficient optical pumping, SML occurs [the parameter space above the dashed curve].

The time evolution of the spin polarization for the magnetic fields in Fig. 15(a) are given in Fig. 15a. Panel (a) corresponds to the RSA regime (weak magnetic fields). One can see that the spin polarization phase and amplitude at small negative delays depends on the relation to the PSC. However, in the ML regime, shown in panel (b), both values are constant irrespective whether the PSC are fulfilled or not.

From the results of Secs. IV and V one can conclude about the two main features of the SML regime. The first one is a fixed phase of the spin signal at very small negative delays, which is independent of the magnetic field. This reflects the primary amplification of spins with
commensurable spin beat frequencies in a strongly inhomogeneous ensemble. The second one is a characteristic revival of the dephased signal before the next pump pulse arrival shown in Fig. 11(c).

It is also interesting, that contrary to the RSA regime in the SML regime the magnetic field dependence of the spin polarization at zero negative delay is smooth. The dependence is similar to that presented by the dashed line in Fig. 11(c). The width of this bell-like curve is determined by the nuclear field fluctuations and is approximately equal to $4\Delta B$.

Let us summarize the conditions for the SML regime. Apart from the obvious condition $\tau_s \gg T_R$ it requires:

1. A significant spread of carrier spin precession frequencies, $\Delta \omega_R > 0.5 \omega_R$. The spread can be caused by the nuclear fluctuation fields or by the spread of $g$ factors.

2. The frequency spread $\Delta \omega > 0.5 \omega_R$ leads to a dephasing of the spin signal within the time $T \sim T_R/\pi$, i.e. faster than the time interval between subsequent pump pulses.

3. One can see from Figs. 14(c) and 14(d) that the pump pulse area should be sufficiently large, $\Theta \gtrsim \pi/2$. Otherwise the frequency spread $\Delta \omega > 0.5 \omega_R$ would cause only a decay of the spin polarization without its revival before the next pump pulse arrival.

**VI. CONCLUSIONS**

To conclude, we have performed a comprehensive theoretical study of carrier spin coherence in spin ensembles subject to periodic optical pumping. The effect of spin accumulation has been analysed for singly-charged quantum dots and quantum wells with a low density carrier gas. The accumulation results in two regimes of carrier spin coherence: resonance spin amplification and spin mode-locking. These regimes, while being different in their phenomenological appearances and realization conditions, have the same origin and occur for spin ensembles for which the carrier spin coherence time exceeds by far the pump repetition period. The resonance spin amplification and spin mode-locking are mutually exclusive regimes because of the requirement on excitation power and precession frequency spread.

For the RSA regime sufficiently homogeneous spin ensembles and small excitation powers (small pump pulse areas) are required. These conditions are experimentally realized in QW structures with electron or hole resident carriers of low density, i.e. for the regime, where negatively or positively charged trions play an important role. In this case the spin dephasing times for resident carriers can be extracted with high accuracy, even when they exceed the pulse repetition period. The spreads of $g$ factors and nuclear spin fluctuations are less important for the long-lived spin coherence compared to the case of strongly inhomogeneous QD ensembles.

In contrast to the RSA regime the SML regime requires a strong inhomogeneity of the spin precession frequency in the spin ensemble and high excitation powers (pump areas close to $\pi$ and more). By now the SML regime has been observed experimentally and studied in great detail for ensembles of (In,Ga)As/GaAs QDs each singly charged with a resident electron. In principle it may be also observable for quantum dots singly-charged with a resident hole, if the respective conditions are met.

**Acknowledgments**

The authors thank A. Greilich, Al. L. Efros, I. V. Ignatiev, and E. L. Ivchenko for valuable discussions. This work was supported by the Deutsche Forschungsgemeinschaft, the Russian Foundation of Basic Research and the EU Seventh Framework Programme (Grant No. 237252, Spin-optronics). IAY is a Fellow of the Alexander von Humboldt Foundation. MMG acknowledges support of the “Dynasty” Foundation—ICFPM.

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