Interplay of spin mode locking and nuclei-induced frequency focusing in quantum dots

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We study the influence of nuclei-induced frequency focusing on the mode locking of spin coherence in quantum dots subjected to a periodic train of optical pulses. In particular, we address the question whether or not nuclei-induced frequency focusing always enhances the effect of spin mode locking. We combine two advanced semiclassical approaches and extend the resulting model by including the full dynamics of the optically excited trion state. In order to reduce the discrepancy to a full quantum model, we establish a non-deterministic pulse description by interpreting each pump pulse as a measurement. Both extensions lead to significant qualitative changes of the physics. Their combination yields an improved description of the corresponding experiments. Importantly, we observe the emergence of dynamic nuclear polarization, i.e., the formation of a non-zero average polarization of the nuclear spin bath, leading to a certain increase of the coherence time.

I. INTRODUCTION

The generation of well-controllable stable quantum states is an ever ongoing challenge in the context of quantum information because of decoherence due to the interaction with the environment. A promising candidate for a technological realization is an electron spin localized in a semiconductor quantum dot (QD) due to the established fabrication tools for semiconductor nanostructures and the possible scalability. The major challenge is the interaction of the quantum states with the environment, eventually leading to decoherence.

Recently, important progress has been made in this field by Gangloff et al. [2], who demonstrated the implementation and manipulation of coherent states in a nuclear spin ensemble coupled to a localized electron spin in a QD, which have achieved by exploiting the hyperfine interaction. This step can be seen as the “missing piece of the puzzle” for a semiconductor nanostructure platform.

In a related earlier experiment by Greilich et al. [4] on QD ensembles subjected to a transverse external magnetic field (Voigt geometry), it was demonstrated that the hyperfine interaction can be also exploited such that the nuclear spin bath acts as a correction field to the Zeeman term which varies from QD to QD. By means of the nuclei-induced frequency focusing (NIFF) effect, the nuclear spin bath is manipulated in such a way that the Larmor precession of the localized electron spins is focused onto very few resonances, enhancing the spin mode locking (SML) effect [5].

The SML effect is briefly described as follows. Usually, the optically induced polarization of the localized electron spins dephases quickly due to a broad and inhomogeneous spectrum of precession frequencies. By applying trains of periodic pulses to the QD ensemble, a revival of the spin polarization emerges before the arrival of the next pulse. The amplitude of these revivals, depending on many system parameters, can be strongly enhanced by frequency focusing in the nuclear spin bath when a very long train of pulses is applied.

By optimization of the experimental protocol, it is even possible to drive the spectrum of Larmor frequencies to only a narrow single mode such that all localized electron spins precess with almost the same frequency [6]. Effectively, this leads to a strong increase of the coherence time and thus, enables the coherent manipulation of the localized electron spins [7].

In the context of SML and NIFF, there are several open questions stemming from recent experiments [8–11]. A fundamental one is whether or not NIFF always acts constructively, i.e., does it always lead to an enhancement of the SML effect, and what is the influence of the external magnetic field strength on this interplay? Recent theoretical studies, both quantum [12] and semiclassical [9, 10, 13], suggest that this is not necessarily the case due to the possibility of additional resonances which act destructively, and the slow Larmor precession of the nuclear spins play a major role in this context. However, the full dynamics of the excited trion state is neglected in these studies, and we lift this simplification in the present paper and demonstrate that this extension influences the physics qualitatively.

The present paper is devoted to a better theoretical description and understanding of NIFF. The existing semiclassical precession models [9, 10, 13, 15] are improved by interpreting the optical pump pulse as a measurement [10, 13]. This allows us to apply a truncated Wigner approximation [16] to the action of each pulse, leading to a reduced discrepancy to a full quantum mechanical description. Moreover, we investigate the role of the dynamics of the optically excited trion state and the role of an inhomogeneous ensemble of QDs. Quantum models describing the effect of NIFF also exist [13, 10, 12, 13, 17, 20], but they are typically restricted to a very small number of nuclear spins due to the exponentially growing Hilbert space so that one needs to either resort to constant hyperfine couplings (box model) or to perturbation theory.
In the next section, we introduce the initial model for the description of a homogeneous ensemble of QDs in a transverse magnetic field subjected to periodic circularly polarized laser pulses, which is a combination of various approaches from the literature [9, 13, 21, 22]. However, we will show that this model does not describe the experimental results appropriately. We extend this initial model step by step in the subsequent sections, leading to the extended models (EM) I, II, and III. A non-deterministic description of the pulse model is introduced in Sec. III by interpreting the pulse as a measurement. This reduces the discrepancy to a full quantum model while still treating a large nuclear spin ensemble. Here, we emphasize the need for further experimental studies with explicit suggestions. The role of inhomogeneities in the QD ensemble is briefly discussed in Sec. V. Finally, a conclusion and an outlook is given in Sec. VI.

II. INITIAL MODEL: LOCALIZED ELECTRON SPIN IN A QUANTUM DOT SUBJECTED TO PERIODIC OPTICAL PULSES

In this section, we introduce and numerically analyze the initial model in which we combine an established pulse model often used to describe the excitation of a trion [9, 21, 23, 25] with an efficient approach to the dynamics of the nuclear spin bath [13, 22], which allows us to simulate the saturation behavior of the system. Note that this model does not yield an appropriate description of some experimental results, but it is a good starting point to introduce the basic phenomena of SML and NIFF.

A. Equations of motion

We consider a homogeneous ensemble of GaAs QDs, i.e., all QDs are equal. They are singly charged by electrons and subjected to a strong transverse magnetic field (Voigt geometry) of up to several Tesla. A sketch which shows the basic model and setup is shown in Fig. 1. In each QD, the internal spin dynamics are governed by the hyperfine interaction of the single localized electron spin $\hat{S}$ with the surrounding $N$ nuclear spins $\hat{I}_k$ of the host lattice [26–32]. Quantum mechanically, this interaction is described by the hyperfine Hamiltonian

$$\hat{H}_{HF} = \sum_{k=1}^{N} A_k \hat{S} \cdot \hat{I}_k = \hat{S} \cdot \hat{B}_{ov},$$  

with the hyperfine coupling constants $A_k$ together with the nuclear spins $\hat{I}_k$ forming the so-called Overhauser field

$$\hat{B}_{ov} = \sum_{k=1}^{N} A_k \hat{I}_k.$$  

This Hamiltonian, often referred to as the central spin or Gaudin model [33–51], is extremely hard to solve for a non-uniform distribution of the couplings $A_k$ so that one is typically restricted by either the bath size $N$ or the maximum simulation time in spite of the existence of a Bethe ansatz solution [33–55].

We treat the dynamics of the system in a semiclassical manner, i.e., we solve the corresponding classical equations of motion and average over an appropriate distribution for the initial conditions of the classical spins [43]. This corresponds to a truncated Wigner approximation [16], which is a semiclassical phase space method. Because a QD comprises $N = 10^4–10^6$ nuclear spins [26, 52, 53], it is well justified to consider the Overhauser field as a classical variable $\hat{B}_{ov}$ chosen randomly from a normal distribution [26, 40, 43, 54]. Since the temperature in experiments corresponds to a much larger energy than the hyperfine couplings, the nuclear spins are in a completely disordered state. Thus, each component $B_{ov}^\alpha$, $\alpha \in \{x, y, z\}$, is initially sampled from a normal distribution characterized by the expectation value $\langle B_{ov}^\alpha \rangle = 0$ and the variance $\text{Var}[B_{ov}^\alpha] = 2/(T_n^*)^2$. We choose a typical value of $T_n^* = 1 \text{ ns}$ [5, 23]. Note that we define the variance the Overhauser field via the dephasing time $T_n^*$ because this time is accessible in the related experiments. Physically it is defined via the

\[ B_{ext} \]
strength of the hyperfine interaction and the spin of the nuclei, see Appendix A for details. In the semiclassical picture, the dynamics of the localized electron spin $\mathbf{S}$ is determined by $(h$ is set to unity)

$$\frac{d}{dt}\mathbf{S} = (\mathbf{B}_{ov} + g_e\mu_B B_{ext} e_x) \times \mathbf{S} + \frac{1}{\tau_0} J^x e_z, \quad (3a)$$

$$\frac{d}{dt} J^z = -\frac{1}{\tau_0} J^z, \quad (3b)$$

$$\frac{d}{dt} I_k = (A_k \mathbf{S} + g_n\mu_B B_{ext} e_x) \times I_k, \quad (3c)$$

where $g_e = 0.555$ is the $g$ factor of the localized electron spin, $\mu_B$ the Bohr magneton, $B_{ext}$ the strength of the external magnetic field, and $e_x$ the unit vector in $x$ direction. The intermediate trion state $J^z$, which is excited by a pump pulse, has the radiative lifetime $\tau_0 = 400$ ps and eventually recombines with the ground state $\mathbf{S}$. The derivation of the trion recombination term follows, e.g., from a Lindblad approach developed by Jäschke et al. [9]. It also appears in several other works, e.g., in Refs. [21, 23–25, 55, 56].

Importantly, the Overhauser field $B_{ov}$ is also a dynamic object since the $I_k$ are dynamic as described by Eq. (3c).

We consider an exponential distribution of the hyperfine couplings [13, 22, 47],

$$A_k \propto \exp(-k\gamma), \quad (4)$$

which is a realistic choice for two-dimensional and flat QDs [22, 37]. The parameter $\gamma$ defines the number of effectively coupled nuclear spins $N_{eff} \approx 2/\gamma$ [13, 22, 47]. Since it is unfeasible to solve the corresponding equations of motion for each individual bath spin $I_k$ for a realistic number of bath spins $N$, we have to resort to a more efficient approach.

By applying the spectral density (SD) approach to the Overhauser field dynamics [22], we reduce the number of dynamic variables from 3$N + 4$ to 3$N_{tr} + 4$, where $N_{tr} = O(75) \ll N$ is a truncation parameter. The essence of this approach is the replacement of the bath spins $I_k$ by appropriate sums of bath spins, represented by the auxiliary vectors $Q_k$, similar to an approach by Erlingsson and Nazarov [30]. These vectors follow the equation of motion

$$\frac{d}{dt} Q_k = (\epsilon_k \mathbf{S} + g_n\mu_B B_{ext} e_x) \times Q_k, \quad (5)$$

where $g_n\mu_B = g_e\mu_B/800$ is roughly the average magnetic moment of the nuclei in a GaAs QD [9, 10, 12, 13, 57], which is roughly 800 times smaller than the magnetic moment of the localized electron due to the much larger masses of the nuclei, and the $\epsilon_k \propto \sqrt{F_i/T_i}$ are effective coupling constants which emerge from the original couplings distribution [4] via application of the SD approach, see Appendix A.2. Finally, the Overhauser field is given by

$$B_{ov} = \sum_{k=1}^{N_{tr}} \sqrt{W_k} Q_k, \quad (6)$$

where the $W_k$ are weights also emerging from the SD approach. Since we focus on GaAs QDs, we choose $I = 3/2$ for the nuclear spins. Thus, we sample the components of the vectors $Q_k$ from a normal distribution around zero with variance $I(I + 1)/3 = 5/4$. For InGaAs QDs, one needs to also account for $I = 9/2$ of the indium isotopes. A detailed explanation of the SD approach is given in Appendix A.2 and in Refs. [13, 22].

B. Pulse model

The periodic optical pumping is carried out with the repetition period $T_R = 13.2$ ns [4, 5, 23]. We focus on resonant pumping by $\pi$ pulses with helicity $\sigma^-$, which have a typical duration of 1.5 ps in the experiments [4, 5, 7, 10, 11, 23, 55]. The pumping of the localized electron spins with this circularly polarized light leads to the excitation of a singlet $X^-$ trion, which decays completely until the arrival of the next pulse under the experimental condition $\tau_0 \ll T_R$. Because one Larmor period even in a magnetic field as large as 9 T lasts about 14 ps, we consider the pulse to act instantaneously since the pulse duration is shorter by one order of magnitude. We consider flat QDs where the lateral size by far exceeds their height, i.e., we can choose the growth axis $e_z$ to be the quantization axis [21].

Under these conditions, the pulse action can be described by a simple relation between the spin components before ($S_{bh}, J_0$) and after ($S_{h}, J_0$) the pulse [9, 21]

$$S_a^z = \frac{1}{4} + \frac{1}{2} S_b^z, \quad (7a)$$

$$S_a^x = S_b^x = 0, \quad (7b)$$

$$J_a^z = S_a^z - S_b^z, \quad (7c)$$

$$J_a^x = 0, \quad J_b^x = 0, \quad (7d)$$

where $J^z$ and $J^x$ are the transverse components of the trion pseudospin $J$. They have no relevance in this section as we neglect possible trion pseudospin dynamics here, but they become important in Sec. IV. Note that for longer pulse durations, the effectiveness of the pulse is reduced especially for very large magnetic fields [10, 59], but this is beyond the scope of the present work.

C. Results

We solve the coupled equations of motion (3a), (3b), and (3c) for the spin dynamics numerically for $M$ random initial fields $\{Q_k\}$ while applying the pulse relation (7) every $T_R = 13.2$ ns for $n_p$ pulses. The actual dynamics of the localized electron spin is given by the ensemble average over all $M$ configurations. In our simulations, we use $M = 4800 - 10000$ most of the time. Let us briefly review the basic phenomena of spin mode locking (SML) and nuclear-induced frequency focusing (NIFF), which can be already discussed quali-
tatively for the initial model. Typical time evolutions between consecutive pulses due to these two effects are shown in Fig. 2a. The first pulse creates a net spin polarization which precesses around the transverse magnetic field $B_{\text{ext}}$. Due to the normal distribution of the Overhauser field, this polarization dephases on the time scale $T^* = 1$ ns according to [26, 49]

$$S_{\text{deph}}(t) \propto \cos(g_e \mu_B B_{\text{ext}}) \exp \left[ -\left( \frac{t}{T^*} \right)^2 \right].$$

(8)

After applying only a few pulses, a revival of this polarization emerges just before the arrival of the subsequent pulse. This effect is called spin mode locking (SML) [5] and it emerges due to a selection of precession modes due to the properties of the pulse [7]. Qualitatively speaking, the modes corresponding to an integer number of Larmor periods between consecutive pulses lead to an enhancement of spin polarization while the spin polarization is destroyed for modes corresponding to a non-integer number. The physics behind this behavior is that the localized electron spin is optically inactive in the first case and optically active in the second case according to the selection rules. It can be shown analytically that a steady state for this revival amplitude emerges when neglecting the Overhauser field dynamics and also the trion recombination ($g_e \mu_B B_{\text{ext}} \gg 1/\tau_0$) [9, 10, 20]. The steady state follows from the condition $S^z(n_p T_R) = S^z(n_p T_R + T_R)$ in combination with the periodic application of the pulse [7]. After averaging over the Overhauser field distribution, the analytical steady state in the SML regime (without NIFF) takes the value [10, 20]

$$S_{\text{SML}} := \lim_{n_p \to \infty} S^z(n_p T_R) = \frac{1}{\sqrt{3}} - \frac{1}{2} \approx 0.07735,$$

(9)

which is also identical to the value of the envelope $S^z$ [Eq. (11)], i.e., the $x$ and $y$ components vanish. In the
following, we will refer to this value as the SML steady state value. This steady state is reached after about ten pulses, independent of the external magnetic field strength.

When driving the system by much longer train of pulses, the effect of nuclei-induced frequency focusing (NIFF) emerges [4], with a rate depending strongly on the parameters $B_{\text{ext}}$, $\gamma$, and $T_n^\ast$. This effect leads to a change of the SML steady state amplitude. The periodic driving of the localized electron spin is transferred to the nuclear spin bath via the hyperfine interaction such that the initial normal distributed Overhauser field evolves towards a comb-like structure, see Fig. 2c.

The position of the emerging peaks in the probability distribution of the Overhauser field corresponds to the two resonance conditions [9, 10]

\[
\Omega_{\text{eff}} T_R = 2\pi k, \quad \Omega_{\text{eff}} T_R = 2\pi k + 2\arctan(B_{\text{eff}} \tau_0) \approx (2k + 1)\pi,
\]

where $k$ is an integer number, $\Omega_{\text{eff}} = |B_{\text{ext}} + g_\mu_B B_{\text{ext}} e_x| \approx \Omega_{\text{eff}}/g_\mu_B$ created by the external magnetic field and the Overhauser field and $\tau_0$ is the radiative lifetime of the trion. We will refer to the first condition \(^{10a}\) as the even resonance condition (ERC) because $2k$ is an even integer. The approximation in the second condition \(^{10b}\) holds for $B_{\text{eff}} \tau_0 \gg 1$, which is the case in our theoretical considerations. Hence, we refer to it as the odd resonance condition (ORC) because $(2k + 1)$ is an odd integer.

Depending on which resonance conditions emerge, the revival amplitude is either increased or decreased with respect to the SML steady state value $S_{\text{SML}}$. The correct description of this interplay as a function of time scales only linearly with $\gamma$ and of the ratio

\[
g_\text{e} \mu_B B_{\text{ext}} T_R = \pi k,
\]

where $k$ is an integer number, which plays a crucial role for the magnetic field dependence of the saturated revival amplitude $S_{\text{lim}} \approx \text{lim}_{n_p \to \infty} S^\dagger(n_p)$. The values of $B_{\text{ext}}$ fulfilling this condition are highlighted in Fig. 2b as vertical dashed lines for two different ratios $g_\text{e} \mu_B / g_\text{n} \mu_B$. This nuclear resonance condition (NRC) describes the number of half-turn revovluions of the nuclear spins in the external magnetic field $B_{\text{ext}}$ between consecutive pulses. Note that the influence of the small Knight field, i.e., the additional field that a nuclear spin sees due to its coupling to the localized electron spin, is neglected. It might induce slight deviations from the expected resonance positions [31].

Let us discuss the details of Fig. 2a. We observe that the curve for $g_\text{e} \mu_B / g_\text{n} \mu_B = 500$ is essentially a horizontally rescaled version of the curve for $g_\text{e} \mu_B / g_\text{n} \mu_B = 800$. Maxima are found close to the values of $B_{\text{ext}}$ fulfilling the NRC [13]. The first maximum ($k = 1$, half turn) is rather broad while the second maximum ($k = 2$, full turn) is quite sharp and slightly shifted to the right from the expected resonance position. We identify the emergence of phase shifts while approaching the second maximum since $S_{\text{lim}} \approx S^\dagger$ and $S_{\text{lim}}^\dagger$ start to deviate from each other. Just after the second resonance is reached, this additional phase vanishes again. For $g_\text{e} \mu_B / g_\text{n} \mu_B = 500$, a third maximum ($k = 3$, full turn) is found, which is very similar to the first one and indicates a periodicity for larger values of $B_{\text{ext}}$. We expect a third maximum also for the ratio $g_\text{e} \mu_B / g_\text{n} \mu_B = 800$, but it is numerically out of reach. The heights of the broad maxima are very simi-
lar. The sharp maximum is slightly less pronounced for \( g_e \mu_B / g_n \mu_n = 800 \), but this is most likely due to the finite discretization of the magnetic field \( B_{ext} \).

The finding of a maximum at 3.9 T for \( g_e \mu_B / g_n \mu_n = 800 \) is the main downside of the initial model. Previous research has established theoretical models which predict minima at the values of \( B_{ext} \) fulfilling the NRC [10, 13], which is also in much better agreement with the experimental results at around \( B_{ext} = 4 \) T [9, 10].

The structure of the dependence \( S_{lim}^\perp(B_{ext}) \) in Fig. 2d can be understood by studying the corresponding quasi-steady distributions of the effective magnetic field \( p(B_{ext}) \). The most relevant examples for \( g_e \mu_B / g_n \mu_n = 800 \) are shown in Fig. 2f. For almost any external magnetic field \( B_{ext} \), indicated by the orange vertical line in the plots, we find sharp peaks at the even (vertical solid black lines) and odd (vertical dashed black lines) resonance conditions, but with different weights. Since the ERC corresponds to full Larmor periods between consecutive pulses, the steady state condition following from the pulse relation (7a) is \( S_n^\perp = S_n^z = 1/2 \). For the ORC, the steady state is determined by \( S_n^\perp = -S_n^z = -1/6 \). For this reason, odd resonances dominate with a three times smaller weight when all contributions from the Overhauser field distribution are summed.

Note that for the same reason the ERC dominates the SML regime if there is no NIFF because the Overhauser field is normally distributed so that this distribution by itself does not favor ERC over ORC or vice-versa.

Interestingly, we find strong deviations from the even and odd resonances at around \( B_{ext} \approx 7.8 \) T, which is the value corresponding to \( k = 1 \) in the NRC [13]. This finding explains the deviations of the \( z \) component from the envelope in Fig. 2a. When increasing the magnetic field just slightly to \( B_{ext} = 8 \) T, sharp peaks are found at the ERC, but small side peaks remain which do not correspond to the expected resonances.

D. Discussion

The initial model shows the fascinating coherent spin phenomena SML and NIFF and also a non-monotonic dependence \( S_{lim}^\perp(B_{ext}) \), which was first published in Refs. [9, 10] and studied in more detail in Refs. [10, 13]. However, the results for the initial model contradict the findings of previous theoretical research [10] where minima instead of maxima were found at the NRC, which is also in much better agreement with the experimental results [9, 10]. While there is experimental evidence that both even and odd modes can appear simultaneously in the frequency spectrum of the localized electron spin [9], this seems to be not the case for every magnetic field. Moreover, the modes found in the experiments appear to be generally much broader. In the following sections, we address these issues by extending the initial model step by step.

III. EXTENDED MODEL I: NON-DETERMINISTIC PULSE DESCRIPTION

As we are modeling the system and the pulses in a semi-classical picture, it is not clear how to treat the excitation of the localized electron spin by the circularly polarized laser pulse. In fact, the pulse model [7] was derived quantum mechanically [21], but the relations are only valid for the expectation values of the spins. One could argue that applying the pulse relation [7] to the spin polarization after calculating the ensemble average, but this approach destroys any correlation otherwise present in a single configuration such that no SML appears when there is no NIFF. However, such correlations are preserved in a quantum mechanical approach.

We extend the pulse model [7] by interpreting it as a measurement. This leads to a non-deterministic pulse description in the sense of a truncated Wigner approximation [10] and reduces the discrepancy to the fully quantum mechanical behavior. The same principle was applied in Refs. [10, 13] to a simpler pulse model, which led to a minimum of the magnetic field dependence \( S_{lim}^\perp(B_{ext}) \) at around 4 T as found in the experiments [9, 10]. Alternative non-deterministic pulse descriptions are discussed in Appendix B but they turn out to be less reliable.

A. Non-deterministic pulse description

The essence of simulating quantum mechanics by classical equations of motion is the choice of the correct initial conditions. Typically, one tries to fulfill the quantum mechanical moments of the corresponding operators, in our case of the spin operators. We already apply this principle to the Overhauser field by sampling it from the proper normal distribution. In this case, the huge number of bath spins provides a valid justification of the approach based on the central limit theorem [43]. In contrast, this argument does not hold for the single localized electron spin which is excited by a pump pulse, so any semi-classical treatment is always an approximation. Nevertheless, we will show that this procedure leads to promising results in our application.

The truncated Wigner approximation (TWA) is the theoretical foundation of the following approach [10]. In this semiclassical phase space method, the initial conditions are sampled from the appropriate Wigner distribution, which is eventually truncated by taking only leading order quantum corrections in \( \hbar \) into account. In leading order, quantum fluctuations appear only through the Wigner distribution of the initial conditions, but they do not affect the equations of motion themselves, i.e., they are classical. Finally, the quantum mechanical time evolution is mimicked by the ensemble average over all classical trajectories.

The main requirement is that the non-deterministic pulse retains the properties of pulse [7] in the SML regime.
We consider each pulse to act as a quantum mechanical measurement \[\text{[13]}\]. In particular, the pulse needs to fulfill the quantum mechanical property for spin-1/2 operators \((S^z)^2 \approx 1/4\). Hence, the deterministic pulse model \[\text{[7]}\] is extended to a non-deterministic description in which the electron spin \(S_e\) after the pulse is sampled from normal distributions characterized by

\[
\begin{align}
E[S_e^z] &= \frac{1}{4} + \frac{1}{2} S_e^z, \\
E[S_e^x] &= E[S_e^y] = 0, \\
\text{Var}[S_e^\alpha] &= \begin{cases} \\
\frac{1}{4} - E^2[S_e^\alpha], & \text{if } E^2[S_e^\alpha] \leq \frac{1}{4}; \\
0, & \text{else}.
\end{cases}
\end{align}
\]  

(14a) (14b) (14c)

The distribution is solely determined by the value \(S_e^z\), i.e., the distribution is different for every pulse unless a steady state is reached. We have to set the variance to zero in some cases because we treat the spins as classical vectors, i.e., a spin component can be larger than 1/2 due to the sampling from a normal distribution. Practically, this issue only arises for the \(z\) component, but for about 25% of the pulses. This alters the effective variance to a certain extent, but it leaves the expectation value unchanged, which is responsible for the correct steady state in the SML regime.

The validity of this non-deterministic pulse description is established in Appendix \[\text{B}\] where several non-deterministic pulse descriptions are benchmarked in the SML regime against the deterministic pulse model \[\text{[7]}\] and its quantum mechanical realization \[\text{[10]}\].

### B. Results

As pointed out above and studied in detail in Appendix \[\text{B}\] the non-deterministic pulse description \[\text{[14]}\] does not alter the behavior in the SML regime besides adding additional fluctuations to the spin polarization. In the following, we study the interplay of SML and NIFF when applying long trains of pulses.

Figure \[\text{3a}\] shows the influence of NIFF on the revival amplitude after the SML regime has been reached for various external magnetic fields \(B_{\text{ext}}\). All curves reach a saturation value after approximately the same number of scaled pulses \(n_p/B_{\text{ext}}^2\) \[\text{[10]}\] \[\text{[3]}\], but the saturation value depends significantly on the magnetic field strength. Due to the additional fluctuations added to the data because of the non-deterministic pulse description, we extract the saturation value \(S_{\text{lim}}^+\) by fitting an appropriate function to the data. It turns out that

\[
S(n_p) = A_{\text{NIFF}} \frac{2}{\pi} \arctan \left( \frac{n_p}{\eta} \right) + B_{\text{SML}} \tag{15}
\]

is a suitable fit function, which describes a \(1/n_p\) convergence towards saturation. This is different from the exponential saturation fit used in Ref. \[\text{[10]}\], which would slightly underestimate (overestimate) the saturation value, e.g., for \(B_{\text{ext}} = 1\ T\) \((B_{\text{ext}} = 4\ T)\). Fits of type \[\text{[15]}\] are included in Fig. \[\text{3a}\] as black dashed lines. Finally, the saturation value for \(n_p \to \infty\) is given by \(S_{\text{lim}} = B_{\text{SML}} + \text{sgn}(\eta) A_{\text{NIFF}}\). Since the fit error turns out to be fairly small, we use the mean squared error of the last 10% datapoints as error estimate.

The quasi-stationary distributions of the effective magnetic field shown in Fig. \[\text{3b}\] have much broader peaks than in the initial model of Sec. \[\text{II}\] where the deterministic pulse \[\text{[7]}\] is applied. They are located at the values of \(B_{\text{eff}}\) corresponding either to the ERC or to the ORC, i.e., only one class of resonances emerges, not both simultaneously as previously in Fig. \[\text{2b}\].

In this extended model, the parameter \(\gamma\), which is proportional to the inverse effective bath size, plays an important role. While the number of pulses required to reach saturation still increases linearly with \(\gamma\), the saturation value \(S_{\text{lim}}^\perp\) changes. A similar behavior is found in Ref. \[\text{[10]}\] for a different, but also non-deterministic, pulse when increasing the effective bath size. It turns out that the typical hyperfine coupling strength, which is proportional to \(\sqrt{B}/T_n^*\), determines the saturation value. Figure \[\text{3c}\] plots \(S_{\text{lim}}^\perp\) against the typical coupling strength for various combinations of \(B_{\text{ext}}^*\) and \(T_n^*\) while varying \(\gamma\). Especially for \(B_{\text{ext}} = 1\ T\) and \(4\ T\), there appears to be a linear dependence which we exploit for a linear extrapolation \(\sqrt{\gamma} \to 0\), i.e., to an infinite effective bath size, which is the limit of interest for QDs with \(10^4\) \(-\) \(10^6\) nuclear spins, i.e., \(\sqrt{\gamma} \approx 10^{-3} - 10^{-2}\). Furthermore, the exact choice of the dephasing time \(T_n^\perp\), which is an input parameter from the experiments, appears to be not important as long as it is significantly shorter than the pulse repetition period \(T_R\). Otherwise, we would approach the regime of resonant spin amplification instead of SML, showing different qualitative physics \[\text{[2]}\]. The data and its extrapolation is not as robust for \(B_{\text{ext}} = 1.95\ T\) where almost no NIFF emerges, but this uncertainty is represented by the fit error which is significantly larger than the error of a single saturation value.

Interestingly, for large values of \(\sqrt{\gamma}/T_n^*\) almost no NIFF emerges and the linear scaling is not applicable. Physically, the effective hyperfine couplings \(\epsilon_k \propto \sqrt{\gamma}/T_n^*\) becomes too large in comparison to the nuclear Zeeman term \(g_n\mu_B B_{\text{ext}}\) in Eq. \[\text{[5]}\]. Hence, the linear scaling for \(\sqrt{\gamma} \to 0\) is expected to work even for larger ratios \(\sqrt{\gamma}/T_n^\perp\) when a larger magnetic field is applied. This is evident for \(B_{\text{ext}} = 3.9\ T\) in Fig. \[\text{3c}\].

We use this new insight in Fig. \[\text{3d}\] by plotting the saturation value \(S_{\text{lim}}^\perp\) as a function of \(B_{\text{ext}}\) for decreasing values of \(\gamma\). Furthermore, we extrapolate the saturation value to an infinite bath \((\gamma \to 0)\) and compare the results to the SML regime. During this extrapolation process, we enforce the physical lower bound \(S_{\text{lim}}^\perp \geq 0\). This is realized by setting \(S_{\text{lim}}^\perp = 0\) if the extrapolation yields a negative value, but we check that the actual extrapolation value and its error are in agreement with the bound. We find two minima at the values of \(B_{\text{ext}}\) which fulfill the NRC \[\text{[13]}\], with the second one being much narrower. The minima and maxima become more pronounced for...
smaller values of $\gamma$, and the minima for $\gamma \rightarrow 0$ correspond to $S_{\lim}^\perp \approx 0$. At around $B_{\text{ext}} = 2$ T and 6 T, almost no NIFF emerges for any choice of $\gamma$. Between these two values, we find a destructive interference of SML and NIFF, leading to a decrease of the revival amplitude, and we also find this behavior in a narrow interval around $B_{\text{ext}} = 7.8$ T. For the other values of $B_{\text{ext}}$, NIFF increases the revival amplitude, i.e., it acts constructively by enhancing the SML already present after a few pulses.

C. Discussion

We mimic the quantum mechanical behavior of the system by a non-deterministic pulse description. As a result, we find the expected minima in the magnetic field dependence of $S_{\lim}^\perp$, similar to the experimental and theoretical results of Kleinjohann et al. [10], and in contrast to the initial model in the previous section. Overall, our results are qualitatively very similar to their quantum mechanical approach. Differences could stem from the considered bath sizes since the quantum mechanical approach is limited to only $N = 6$ nuclear spins.

Summarizing, the quasi-stationary distribution of the effective magnetic field shows much broader peaks than previously in Sec. III. This is in much better agreement with what is found experimentally [9, 11]. Moreover, only one class of resonances emerges, which corresponds to either an integer (ERC) or a half-integer (ORC) number of Larmor periods between consecutive pulses. Depending on the magnetic field, either the ERC or the ORC is fulfilled, which in turn leads to an increase or a decrease of the revival amplitude relative to the SML regime, respectively. Importantly, the minima and maxima of the magnetic field dependence become more pronounced for...
larger bath sizes while the dephasing time $T^*_n$ only has a small influence in the limit of an infinite bath size.

IV. EXTENDED MODEL II: TRION PSEUDOSPIN DYNAMICS

Up to this point, we treated the trion only as an intermediate state $J^e$ which eventually recombines on the timescale $\tau_0 = 400 \text{ ps}$ [see Eq. 3]. However, we neglected the dynamics of its pseudospin $J$, which is very similar to the dynamics of the ground state of the localized electron spin $S$ described by the equation of motion (16). In the recent theoretical studies of Refs. [9, 10, 13], this dynamics was not considered either. But the description of its dynamics, especially the coupling to the external magnetic field, is crucial for the correct description of the time evolution between consecutive pulses. In the context of spin inertia and polarization recovery measurements, where rather small magnetic fields up to a few 100 mT are applied in Faraday geometry, the detailed description of the trion pseudospin dynamics is absolutely mandatory [25, 65, 62]. In the following, we will show that its inclusion will also alter the behavior of NIFF as a function of the magnetic field, and importantly, we find evidence for dynamic nuclear polarization (DNP), i.e., the emergence of a non-zero average polarization of the nuclear spin bath, in the model.

A. Equations of motion for trion pseudospin

The optical transition induced by a $\sigma^-$ pump pulse leads to the excitation of a singlet $X^-$ trion. In this case, the trion consists of two electrons in a spin singlet state and a heavy hole with unpaired spin such that the effective type of charge carrier in the excited state (hole) is opposite to the type in the ground state (electron). The dynamics of the resulting trion pseudospin $J$ is induced by the effective magnetic field, but we need to consider that the hyperfine interaction is much weaker and anisotropic for hole spins because it is caused by the dipole-dipole interaction [63, 64]. Then, this hyperfine interaction is described by the anisotropic hyperfine Hamiltonian [65]

$$ \hat{H}_{\text{HF,anisotropic}} = \sum_{k=1}^{N} \left[ \chi A_k \hat{J}^z \hat{I}_k^z + \frac{\chi}{\lambda} A_k (\hat{J}^x \hat{I}_k^x + \hat{J}^y \hat{I}_k^y) \right]. $$

(16)

The resulting classical equation of motion for the trion pseudospin has the form

$$ \frac{d}{dt} J = \chi \left( B_{ov}^z e_z + \frac{1}{\chi} B_{ov}^x \right) \times J $$

$$ + g_n \mu_B B_{\text{ext}} e_z \times J - \frac{1}{\tau_0} J, $$

(17)

$$ \frac{d}{dt} Q_k = \epsilon_k S \times Q_k + \chi \epsilon_k \left( J^z e_z + \frac{1}{\chi} J^x e_x \right) \times Q_k $$

$$ + g_n \mu_B B_{\text{ext}} e_z \times Q_k, $$

(18)

with $J^y = J^z e_z + J^x e_x$. The $\epsilon_k$ and $W_k$ do not change as the hyperfine couplings $A_k$ are still parameterized by the exponential distribution [41] and the Overhauser field is still given by Eq. (A9).

B. Results

The numerical implementation of the extended equations of motion [17] and [18] is straightforward and only leads to a negligible increase of computational complexity in comparison to the equations of motion considered in the two previous sections. In the following, we will discuss the main changes which emerge in comparison to the results of Sec. III.

1. Spin dynamics

In the pump-probe experiments under consideration, the Faraday rotation or ellipticity is measured by weak linearly polarized pulses. The probed signal is proportional to $S^z - J^z$ [27], i.e., the spin polarization of the system is measured. Figure 4a shows the corresponding time evolution in our model between two pulses in the saturation regime for the two magnetic fields $B_{\text{ext}} = 1 \text{ T}$ and $4 \text{ T}$. As expected, the initial dephasing shows additional modulations, stemming from the trion pseudospin which precesses around the external magnetic field with a slightly different Larmor frequency than the electron spin. The modulations decay on the timescale $\tau_0 < T_R$ so that they do not appear in the revival before the next pulse.

2. Nuclei-induced frequency focusing

The most prominent difference to the EM I of Sec. III becomes evident when studying the magnetic field dependency of the revival amplitude, which is plotted in Fig. 4b.
for various values of the parameter $\gamma$. While we still find two minima at values of $B_{\text{ext}}$ fulfilling the NRC \cite{13}, the first broad minimum hints at the emergence of even resonances instead of the previous odd ones because the value of the revival amplitude is larger instead of smaller compared to $S_{\text{SML}}$. This is supported by the corresponding distribution of the effective magnetic field $p(B_{\text{eff}})$ for $B_{\text{ext}} = 3.9$ T in Fig. 4d where peaks are found at the values of $B_{\text{ext}}$ fulfilling the ERC \cite{10a}. In this regime, the values of the revival amplitude are larger than the mere SML steady state value $S_{\text{SML}}$, which is plotted as a black horizontal dashed line in Fig. 4b. However, the the overall degree of NIFF in this regime is small. This is different from our findings for the EM I where the revival amplitude approaches zero and the effect of NIFF becomes more pronounced for small values of $\gamma$. Furthermore, the second minimum in Fig. 4b still corresponds to the ORC \cite{10b} in the probability distribution $p(B_{\text{eff}})$ as shown in Fig. 4c for $B_{\text{ext}} = 7.8$ T. This minimum is narrower compared to the second minimum found for the EM I.

The linear extrapolation $\sqrt{\gamma} \to 0$ for the saturation value $S_{\text{lim}}^+$, which we have established in Fig. 3c of Sec. 11, is still valid when including the trion pseudospin dynamics, and the exact choice of the dephasing time $T_n^*$ is also unimportant here. We apply this extrapolation procedure for the magnetic field dependence $S_{\text{lim}}^\pm(B_{\text{ext}})$ in Fig. 4b. Overall, the structure becomes more pronounced in the limit $\gamma \to 0$, but the revival amplitudes around the minimum at $B_{\text{ext}} = 3.9$ T are almost independent of $\gamma$. Moreover, the maxima have a similar height as in Fig. 4b. This means that the degree of NIFF under optimal conditions is very similar. Under these conditions, the revival amplitude is more than three times larger than in the SML regime without NIFF. Experimentally, a ratio of 3.6 for the revival amplitude with vs. without NIFF is
found for $B_{\text{ext}} = 2$ T \cite{11}. For this particular magnetic field, we find a ratio of only 2.

The influence of the parameter $\chi = 0.2$, which characterizes the hyperfine interaction strength between the trion pseudospin and the Overhauser field, is minor. Since the trion only has a lifetime of 400 ps and the hyperfine interaction is much weaker than for the electron spin, this coupling could be neglected for its smallness. In our simulations, we find very similar NIFF for $\chi = 0$, i.e., without the coupling of the trion pseudospin to the Overhauser field. At best, NIFF is marginally more pronounced when this coupling is neglected because it acts as a small additional perturbation. However, the deviations between the results are of the order of the estimated error so that no reliable conclusion can be given.

The distribution of the hyperfine couplings is also not essential for the qualitative NIFF behavior. When we employ a simple box model, i.e., we choose all hyperfine couplings to be the same with $A_k \propto (\sqrt{N} T^*_{\alpha})^{-1}$, the magnetic field dependence of the revival amplitude $S_{\text{lim}}(B_{\text{ext}})$ shows the same qualitative shape as in Fig. 4b with a slightly more pronounced NIFF. Since all nuclear spins precess with the same frequency in this simplified model, the nuclear spins cannot change their mutual angles. We will see, however, that the alignment of nuclear spins is one of the two essential mechanisms leading to dynamic nuclear polarization.

3. Dynamic nuclear polarization

When studying the probability distributions of the effective magnetic field $p(B_{\text{eff}})$ in Fig. 4c, one can discern a small shift of the distribution to the right for $B_{\text{ext}} = 1$ T and 8 T. In order to highlight this shift, we include the mean value $B_{\text{eff}}$ of the distribution as a green vertical line in the plots. The applied magnetic field $B_{\text{ext}}$ is indicated in orange. Remember that at the beginning of each simulation, $B_{\text{eff}} \approx B_{\text{ext}}$ holds, with small deviations stemming only from the statistical nature of our approach.

The shift results from dynamic nuclear polarization (DNP) in the Overhauser field, i.e., the nuclear spins align along the axis of the external magnetic field $B_{\text{ext}} e_x$ to a certain extent. In order to analyze this phenomenon, we define the DNP as

$$\Delta B_{\text{DNP}} := \frac{B_{\text{ov}}(n_p) - B_{\text{ov}}(n_p = 0)}{g_\sigma \mu_B}$$

and study it as a function of the magnetic field for several values of $\gamma$ in Fig. 4d. The number of pulses $n_p$ is chosen such that $S_{\text{lim}}$ is approximately in saturation. The dots and solid lines represent $\Delta B_{\text{DNP}}$, the triangles and dashed lines its absolute value. Interestingly, there is a very similar shape as in Fig. 4c when studying the absolute value of the shifts, and we find no DNP at the magnetic fields fulfilling the NRC \cite{13}.

The difference between the DNP for different values of $\gamma$ is minor for most magnetic fields. Note that the shifts $\Delta B_{\text{DNP}}$ plotted in Fig. 4d do not represent the stationary values even though the values of $S_{\text{lim}}$ are approximately saturated because the DNP approaches its limit much slower than the revival amplitude. This leads to slightly different results for the different values of $\gamma$. It appears that for the magnetic fields for which the shift $\Delta B_{\text{DNP}}$ is most prominent, smaller values of $\gamma$ correspond to a stronger DNP. Unfortunately, it is not possible to reach the stationary DNP regime for the large magnetic fields due to its slow convergence, but we analyze the DNP behavior in detail for small magnetic fields in the following. In Fig. 5 we analyze the saturation behavior of DNP and the internal mechanism leading to its emergence.

Figure 5. Analysis of the DNP behavior in the EM II for $B_{\text{ext}} = 0.5$ T and $\gamma = 0.004$. All fits (black dashed lines) are of the type \cite{20}. (a) Build-up of the DNP $\Delta B_{\text{DNP}}$ (blue line), defined by Eq. \cite{19}, due to periodic driving with pulses. The orange line shows the simultaneous increase of the average Overhauser field length $B_{\text{ov}}$. (b) Decrease of the standard deviation of the Overhauser field components $B_{\text{ov}}, \alpha \in \{x,y,z\}$, due to periodic driving with pulses. (c) Average angles $\theta$ between the Overhauser field and the unit vectors $e_\alpha$, as a function of the number of pulses. (d) Probability distributions $p(\theta)$ after different numbers of pulses.
Figure 5a shows the build-up of the DNP $\Delta B_{\text{DNP}}$ due to periodic driving with pulses for $B_{\text{ext}} = 0.5$ T and $\gamma = 0.004$, eventually reaching a steady state of about 100 mT after more than $10^5$ pulses. This DNP is fairly large in comparison to the initial standard deviation of the Overhauser field components of about 29 mT. Note that the revival amplitude $S_{\text{lim}}$ is already saturated after 5000 pulses for this set of parameters. A more than 200 times longer train of pulses is required to reach the saturation regime for DNP.

The DNP build-up and saturation as a function of the number of pulses can be described via

$$f_{\text{DNP}}(n_p) = A_{\text{DNP}} \frac{2}{\pi} \arctan \left( \frac{n_p}{\eta} \right) + B_{\text{DNP}},$$

where $A_{\text{DNP}}$, $B_{\text{DNP}}$, and $\eta$ are fit parameters. This function shows a $1/n_p$ convergence towards saturation.

In parallel, the average length of the Overhauser field

$$\overline{B}_{\text{ov}} := \left| \frac{B_{\text{ov}}}{g_\text{e}\mu_B} \right|$$

also increases, and its dependence on the number of pulses can be described by the fit (20) as well. This lengthening alone does not explain the emergence of DNP completely, see below.

Figure 5a shows the average angles

$$\overline{\theta}^\alpha := \arccos \left( \frac{\overline{B}_{\text{ov}}^\alpha}{\overline{|B_{\text{ov}}|}} \right).$$

$\alpha \in \{x, y, z\}$, between the Overhauser field components $B_{\text{ov}}^\alpha$ and the unit vectors $e_\alpha$ as a function of the number of pulses. The average of the initial angle is given by $\pi/2$ for all components $\alpha$ since the components $B_{\text{ov}}^\alpha$ are sampled from a simple normal distribution. Driving the system with periodic pulses does not influence the average angles $\overline{\theta}$ and $\overline{\theta}^z$, but the average angle $\overline{\theta}^x$ is reduced to about $\pi/10$, implying that the Overhauser field aligns along the $x$ axis, i.e., the axis of the external magnetic field. The dependence of $\overline{\theta}^x$ on the number of pulses can be described again by the function (20). Note that $\overline{\theta}^x$ is not reduced to zero because of the finite components $B_{\text{ov}}^y$ and $B_{\text{ov}}^z$. These components still follow a normal distribution but with a standard deviation reduced by about 40%.

The corresponding probability distributions of the angles $\theta^\alpha$ after different number of pulses are plotted in Fig. 5b. Initially, all components follow the same distribution with a maximum at $\pi/2$. Due to the continuous driving with pulses, the distributions of all components become narrower, i.e., more focused around a certain angle. The angles $\theta^y$ and $\theta^z$ remain centered around $\pi/2$, but the angle $\theta^x$ becomes significantly smaller as also shown in Fig. 5c.

An important consequence of DNP is a narrowing of the Overhauser field distribution which can be seen in Fig. 5c shows the average angles $\overline{\theta}^\alpha$ after different number of pulses are plotted in Fig. 5b. Initially, all components follow the same distribution with a maximum at $\pi/2$. Due to the continuous driving with pulses, the distributions of all components become narrower, i.e., more focused around a certain angle. The angles $\theta^y$ and $\theta^z$ remain centered around $\pi/2$, but the angle $\theta^x$ becomes significantly smaller as also shown in Fig. 5c.

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For $\Delta B_{\text{DNP}}$, this extrapolation yields a value of 117 mT.

Figure 6. Extended model II: Limiting values of the DNP $\Delta B_{\text{DNP}}$, the average Overhauser field length $\overline{B}_{\text{ov}}$, the Overhauser field standard deviation $\sigma_{\text{ov}}^\alpha$, and the average angle $\overline{\theta}^\alpha$, calculated by applying a fit of type (20) to the data for $B_{\text{ext}} = 0.5$ T (circles) and 1 T (stars) for various values of $\gamma$. The fit errors are usually too small to be discernible. The dashed ($B_{\text{ext}} = 0.5$ T) and dash-dotted ($B_{\text{ext}} = 1$ T) lines represent linear fits, allowing for extrapolations $\gamma \to 0$.
for $B_{\text{ext}} = 0.5$ T and a value of 98 mT for $B_{\text{ext}} = 1$ T. For $B_{\text{ext}} = 0.5$ T, the standard deviations $\sigma_{ov}^{B}$ and $\sigma_{ov}^{\gamma}$ are reduced by more than one third from their initial value of about 29 mT to only 18.7 mT. The standard deviation of $B_{ov}$ increases for smaller $\gamma$ to about 23 mT, but this is still smaller than the initial value. For $B_{\text{ext}} = 1$ T, this anisotropy disappears and the standard deviation of all components are reduced to about 21.3 mT.

Comparing the results for $\gamma \to 0$ with the data for finite values of $\gamma$, we can conclude that all main effects are already present for, e.g., $\gamma = 0.01$. The size of the spin bath only influences the precise values for the observables, but the order of magnitude turns out to be independent of $\gamma$. Let us briefly discuss the role of the dephasing time $T_{n}^{\gamma}$ for DNP. The typical fluctuation strength of the Overhauser field is proportional to $1/T_{n}^{\gamma}$ such that we also expect a linear dependence of the maximum DNP on this parameter. For quantum dots with, e.g., $T_{n}^{\gamma} = 2$ ns, we find a DNP of about 45 mT for $B_{\text{ext}} = 0.5$ T and $\gamma \approx 0.01$. For the same parameters but $T_{n}^{\gamma} = 1$ ns, the DNP of 94 mT is around twice as large, and for $T_{n}^{\gamma} = 0.5$ ns, we find a DNP of 123 mT. For $\gamma \to 0$ and $T_{n}^{\gamma} \approx 4$ ns, which corresponds to the QD sample discussed in Ref. [67], we estimate a DNP of about 30 mT based on the assumption of a linear scaling with $(T_{n}^{\gamma})^{-1}$. In any case, the DNP is expected to be significantly larger than the average fluctuations of the Overhauser field. It would be very interesting to verify or falsify these predictions experimentally.

### C. Discussion

The inclusion of the trion pseudospin dynamics is a crucial step towards the correct description of the underlying pump-probe experiments. It turns out to also have an important qualitative influence on the behavior of NIFF. In Sec. [11], we found a broad regime where pronounced peaks in the probability distribution $p(B_{\text{ext}})$ emerge where the ERC is met. However, when including the trion pseudospin dynamics, the majority of magnetic fields $B_{\text{ext}}$ lead to the emergence of peaks at the ERC, with the only exception being the very narrow but apparently robust feature around $B_{\text{ext}} = 7.8$ T. Around the broad minimum at $B_{\text{ext}} = 3.9$ T, we find very weak frequency focusing in the Overhauser field (see Fig. [13]), which is also independent of the effective bath size. In contrast, the frequency focusing without inclusion of the trion pseudospin dynamics is much more pronounced at this magnetic field (see Fig. [5]), but with the ORC instead of the ERC. As a result, we find an increase of the revival amplitude in comparison to the SML regime without NIFF due to the constructive interplay of NIFF and SML. In contrast, the revival amplitude is strongly suppressed for the EM I of Sec. [11].

In the quantum mechanical model with $N = 6$ bath spins used by Kleinjohann et al. [10], odd resonances emerge around the NRC for $B_{\text{ext}} = 3.9$ T ($k = 1$) with an accompanied minimum of the revival amplitude. They also find a minimum at $B_{\text{ext}} = 7.8$ T ($k = 2$), but is much broader than in the simulations presented here and neither the ERC nor the ORC is fulfilled, similar to what we found in Fig. [26] for the initial model. However, the trion pseudospin dynamics was not accounted for in Ref. [10]. Hence, the ERC in its original form does not hold anymore because the trion dynamics becomes more complex when including its full dynamics as in Eq. [17]. At present, however, the origin of the persistent sharp minimum at $B_{\text{ext}} = 7.8$ T remains unclear.

The experimental situation on this issue is unclear, but the tools for a systematic study are available. By applying a radio frequency field, Evers et al. [11] were able to scramble the nuclear spins in the QDs such that they do not contribute to the revival amplitude by means of NIFF. By applying this approach to a broad range of magnetic fields, a systematic comparison between SML without NIFF and SML with NIFF is possible. Such an experimental study is likely to clarify whether or not NIFF always leads to an increase of the revival amplitude. Moreover, a detailed search for sharp features around the NRC [13], especially for $k = 2$, is promising. Note that multiple NRCs are possible when considering all individual magnetic moments of the isotopes of the QD sample instead of an average one. The consideration of several isotopes is likely to lead to a more complex structure of the revival amplitude as a function of the magnetic field strength. This issue is beyond the scope of the present work, but will be the topic of future research.

The finding of evidence for DNP of about 120 mT in the simulation of these experiments is fascinating because it implies an increase of the coherence time. Indications for DNP in similar but simpler simulations can be seen in the results of Ref. [13], but the effect remained unnoticed. Note that the non-deterministic pulse is not responsible for DNP. We also find DNP when we use the deterministic pulse model [7] in combination with the full trion dynamics. For this combination, DNP as a function of the magnetic field shows a fairly similar behavior as depicted in Fig. [14]. However, almost perfect NIFF fulfilling the ERC with $S_{\text{lim}}^{\perp} \to 0.5$ emerges when studying this particular combination. Only for $B_{\text{ext}} = 7.8$ T, the ORC with $S_{\text{lim}}^{\perp} \to -1/6$ emerges instead of the ERC. But since the
broad minimum around $B_{\text{ext}} = 3.9$ T is missing, we do not study this combination in more detail.

As argued above, the model studied by Glazov et al. [13] is similar to our model because the trion pseudospin effectively decouples from the ground state for large magnetic fields. In their model, DNP is predicted analytically by studying the stability of the fixed point given by the ERC [10a]. Without any additional nuclear spin relaxation, the resonance condition turns out to be an unstable fixed point so that DNP is possible. In agreement with Ref. [13], changing the helicity of the pulses does not change the DNP direction. Note that the emergence of DNP could be less efficient in the experiments due to weak nuclear spin relaxation [13].

Experimental hints for DNP in this type of experiment already exist. In the measurements of Ref. [13], the distribution of Larmor frequencies, extracted from the measured real-time evolution via pump-probe spectroscopy, is shown, and one can discern a shift from the bare Larmor frequency resulting from the external magnetic field, possibly due to DNP.

Since about 200 times more pulses are required to reach the saturation of the DNP in comparison to the steady state of the revival amplitude due to NIFF, it is well possible that the DNP steady state is not yet reached in the experiments so far. Experimentally, it takes around a minute to reach a strong revival amplitude for a magnetic field of 6 T [1]. By applying the suggested scaling with $B_{\text{ext}}^{-2}$, which has yet to be confirmed experimentally, we estimate a strong DNP to emerge within 5 minutes for a magnetic field of 1 T and within 3 hours for a magnetic field of 6 T. A systematic experimental study of this fascinating feature is definitely called for.

V. EXTENDED MODEL III: INHOMOGENEOUS ENSEMBLE OF QUANTUM DOTS

In the previous section, we found modulations in the measured time evolution between consecutive pulses shown in Fig. 4a, which survive for the trion lifetime $\tau_0 = 400$ ps. However, it was measured that these modulations die out much quicker, e.g., on a timescale of about 170 ns [23]. In addition, the ensemble dephasing time $T_2^*$ shows a strong magnetic field dependence in the experiments [5, 23, 67], which cannot be explained by the Overhauser field.

Until now, we considered a homogeneous ensemble of QDs with a dephasing time $T_2^* = 1$ ns. This is, however, a simplification. The $g$ factor of the localized electron spin varies slightly from QD to QD because they are not identical, leading to a faster dephasing for large magnetic fields. We consider resonant optical pumping in this paper, i.e., the $g$ factor of the electron spin in each QD can be modeled by a normal distribution with expectation value $g_e$ and standard deviation $\Delta g_e = 0.005$ [5, 67], leading to the ensemble dephasing time $T_2^*$ defined by

$$\left(T_2^* \right)^{-2} = \left(T_{\text{n}}^* \right)^{-2} + \left(T_{\text{inh}}^* \right)^{-2}, \quad (24)$$

with the dephasing time $\left(T_{\text{inh}}^* \right)^{-1} = \Delta g_e \mu_B B_{\text{ext}} / \sqrt{2}$ due to the inhomogeneities of the QD ensemble. This total dephasing time decreases for large magnetic fields while its upper bound is given by $T_{\text{n}}^*$ for $B_{\text{ext}} \to 0$.

We apply the same modeling to the $g$ factor of the trion pseudospin with standard deviation $\Delta h_e = 0.016$ to account for the fast vanishing of the modulations in the time evolution observed in the experiments. Note that this value is chosen empirically such that the spin dynamics resembles the experimental data. When including a
finite spread $\Delta g_\|$ for the $g$ factor of the trion pseudospin, the polarization of the ensemble dephases on a timescale which can be shorter than the radiative lifetime $\tau_0$. The average magnetic moment of the nuclei does not change, i.e., it is still chosen as $g_n \mu_B = g_n \mu_B / 800$.

The implementation of the spread of the $g$ factors is straightforward in our simulations. It is realized by sampling the $g$ factors $g_n$ and $g_\|$, from the aforementioned normal distribution around their expectation values 0.555 and 0.66, respectively.

Let us discuss which results of the previous section change when studying an inhomogeneous ensemble of QDs. Figure 7a shows the overall faster dephasing for larger magnetic fields $B_{\text{ext}}$, as expected from Eq. (24).

The modulations also vanish much quicker, which is in better agreement with the actual experiments [5, 23, 58]. An even better agreement can be achieved by explicitly fitting the system parameters to experimental results, e.g., the $g$ factor of the trion pseudospin $g_\|$, but this is not the goal of this paper.

Instead, we are interested if and how an inhomogeneous ensemble of QDs alters the interplay of SML and NIFF. Obviously, modeling the $g$ factor of the electron spin by a normal distribution leads to a broadening of the distribution of the effective magnetic field $p(B_{\text{eff}})$ for large magnetic fields as demonstrated in Fig. 7b, but the width of each individual peak due to frequency focusing does not change noticeably.

Without the spread of $g$ factors, peaks also appear in the probability distribution of the mere Overhauser field $p(B_{\text{ov}})$ due to NIFF. They are slightly shifted from the expected resonance positions and they are also slightly broader compared to the probability distribution of the effective magnetic field $p(B_{\text{eff}})$ [13]. In contrast, for the inhomogeneous ensemble of QDs under study even a rather small magnetic field of 0.5 T is enough to smear out the resonances in the distribution $p(B_{\text{ov}})$. The minimal width of the peaks is limited by the $g$ factor spread. Once this width is larger than the distance $2\pi / T_R$ between adjacent resonances, i.e., $T_R^{-1} \lesssim \Delta g_n \mu_B B_{\text{ext}}$, no peaks can occur. Hence, we find no peaks in the mere Overhauser field distribution $p(B_{\text{ov}})$ here, only the distribution of the effective magnetic field $p(B_{\text{eff}})$ shows a comb-like structure.

In Fig. 8 we compare the final results for the magnetic field dependence of the revival amplitude $S_{\text{lim}}(B_{\text{ext}})$ of the EMs I, II, and III in the limit of an infinite bath size ($\gamma \to 0$). It turns out that no significant difference can be found between EM II (homogeneous) and III (inhomogeneous), i.e., the qualitative interplay of SML and NIFF does not change upon inclusion of a finite $g$ factor spread. From the comparison we conclude that the minimum at $B_{\text{ext}} = 7.8$ T is even narrower for the EMs II and III in comparison to EM I. Possibly, this narrow feature remains unmeasured in experiments when the discretization of the magnetic field is chosen too large; we only find it when calculating data for $B_{\text{ext}} = 7.8$ T. For this reason, a systematic experimental search for this narrow feature would be very interesting.

DNP occurs even when studying an inhomogeneous ensemble of QDs (not shown), with an almost identical DNP behavior as in Fig. [13]. This is expected because the additional small variances of the $g$ factors barely change the dynamics from QD to QD, i.e., the qualitative physics remains the same as in the EM II. The same argument holds for the similarity of the results for the EMs II and III in Fig. 8.

However, the influence of DNP on the dephasing time is smaller. DNP only implies a decrease of the dephasing time $T_n^\text{ph}$, which is determined by the strength of Overhauser field fluctuations. The inhomogeneous dephasing time $T_{n}^\text{ph}$ is not altered by DNP. Hence, the relative influence of the narrowed Overhauser field distribution due to DNP on the total dephasing time $T_n^\text{ph}$ as defined by Eq. (24) is diminished, especially for large magnetic fields.

VI. CONCLUSION

We developed an improved semiclassical model for the spin mode locking (SML) effect in combination with nuclei-induced frequency focusing (NIFF) in QDs which yields an improved numerical description of various experimental results. The final model is the result of a combination of several key points while exploiting various scaling arguments.

First, we combined an established semiclassical pulse model often used to describe the excitation of a trion [9, 21] with an efficient approach to the spin dynamics of the Overhauser field [13, 22]. However, the results do not match our expectations gained from a quantum mechanical description of the problem, and they also disagree with the experimental results [9, 10].
Consequently, we improved the pulse model via a nondeterministic description (EM I) in which we interpret the pulse as a measurement in order to reduce the discrepancy to quantum mechanical results while being able to cope with large nuclear spin baths. This step to considerably improved results which are in qualitative agreement with what is found in the quantum model of Ref. [10], where only a small nuclear spin bath is considered. In this improved model and in agreement with Ref. [10], both even and odd resonances are found in the probability distribution of the effective magnetic field due to NIFF for different strengths of the external magnetic field. Importantly, the two kinds of resonances do not appear simultaneously and the corresponding peaks are rather broad due to the mimicked quantum fluctuations. The emergence of odd resonances in the distribution of the effective magnetic field leads to a reduction of the SML effect in comparison to the case without NIFF. This means that NIFF leads to a reduction of the revival amplitude in this model for a broad range of magnetic fields.

We improved our theory further by including the full dynamics of the trion pseudospin, resulting in the extended model II (EM II). This step is necessary to describe the measured time evolution between two consecutive pulses correctly. We found that the fast Larmor precession of the trion pseudospin acts as a perturbation which suppresses the odd resonances such that the observed behavior of NIFF in the system as a function of the external magnetic field is qualitatively different from the one of the previous model (EM I). Here, NIFF acts only constructively except for a very narrow regime around a resonance condition for the nuclear spins where their Larmor period between consecutive pulses matches the pulse repetition time.

Furthermore, we observed the emergence of dynamic nuclear polarization (DNP) of the order of 100 mT, i.e., the formation of a non-zero average polarization of the nuclear spin ensemble, which can be much larger than the typical fluctuations of the Overhauser field. Similar behavior can be inferred from the experimental results presented in Ref. [9] where the spectrum of Larmor frequencies of the localized electron spins is studied. Importantly, the saturation of the DNP takes about 200 times longer than the saturation of the NIFF. Its emergence leads to a slight narrowing of the Overhauser field distribution by about one third and thus, also to a slight increase of the dephasing time. Moreover, we find a similar dependence of the DNP on external magnetic field as for the NIFF. The absolute value of the DNP is minimal around the nuclear resonance conditions where the nuclear Larmor period corresponds to a multiple of the half pulse repetition time. For a typical experiment, we estimate the maximum DNP to be 30 mT for a magnetic field of 1 T, which is be reached after about 5 minutes. This DNP is significantly larger than the average fluctuation of the Overhauser field.

The consideration of an inhomogeneous ensemble of QDs led to the extended model III (EM III). This step is crucial to achieve a correct description of the measured spin dynamics between two consecutive pulses. It does not lead to a qualitatively different DNP behavior, and also not to a different interplay of SML and NIFF.

In this final model (EM III), the peaks in the Overhauser field distribution are fairly broad compared to, e.g., the initial model discussed in Sec. II. This is similar to what is found in the quantum mechanical model of Ref. [10]. Thus, we attribute this behavior to quantum fluctuations captured by the randomness of the pulse model introduced in Sec. IV.

Note that the width of the peaks is not determined by some additional relaxation time induced by further interactions such as the quadrupolar interaction or dipole-dipole interaction, but it is intrinsic to the studied model at hand. But indeed, such further interactions are another possible mechanism which could hinder the efficiency of NIFF. Moreover, weak nuclear spin relaxation due to such interactions could lead to a reduced DNP efficiency.

Further extensions of the model are conceivable. First, the model should be extended to account for the various isotopes in InGaAs QDs so that several resonance conditions for the nuclear spins act in a combined way. We expect that this extension leads to a more complex structure in the magnetic field dependence of the revival amplitude, similar to the experimental results.

In the present study, we have focused only on the resonant excitation of a trion. However, the applied pulse model can be easily generalized to detuned pulses [21]. Thereby, one can account for the influence of the inhomogeneous broadening of the QD sample on the trion excitation [8, 17, 68] and explicitly calculate the Faraday rotation and ellipticity [21], which show different dependencies on certain parameters [8]. Moreover, this step would enable us to simulate two-color pump-probe experiments [8, 21, 68]. Detuned pulses can lead to the emergence of different resonances as demonstrated in Refs. [11, 69]. There, the influence of a positive and negative detuning on NIFF is discussed, but a different model is used for the spin dynamics. In another recent study by Dominguez et al. [70], the optical Stark effect induced by detuned pulses turns out to be very important to accurately describe DNP in the system.

A third relevant aspect is the inclusion of a finite pulse duration, which leads to a reduced efficiency of the pulse for large magnetic fields and can also lead to phase shifts for the resonances [10, 59].

From the experimental side, several clarifications can stimulate progress in understanding the relevant physics. First, different models suggest different scaling laws for the rate of NIFF as a function of the magnetic field; in our case the rate is reduced by $B^2_{\text{ext}}$. Second, a systematic comparison of the revival amplitude as a function of the magnetic field for the cases with and without NIFF is helpful, accompanied by an analysis of the Larmor frequency spectrum with respect to the class of resonances. Such an experimental study can also reveal the influ-
of motion each bath spin follows the classical equation of motion (A1), which is the most efficient choice [22].

The intervals become exponentially small for increasing \( k \), which is the most efficient choice [22].

The prefactor \( \lambda \) is determined from the relation

\[
\lambda = \left( \frac{N_{\text{tr}}}{\sqrt{2\gamma A_Q} t_{\text{max}}} \right)^{1/(N_{\text{tr}} - 1)},
\]

where \( t_{\text{max}} = n_p T_R \) is the total simulation time. For \( \lambda = 1 \), the discretization is simply equidistant. It turns out that such experiments can be performed by applying an appropriate radiofrequency field to the system which hinders the frequency focusing of the nuclei. But so far measurements at various strengths of the magnetic field have not been performed. The occurrence of DNP in the system for which there is some evidence in the experimental data in Ref. [9] is another subject calling for further investigation.

The improved description of the spin dynamics in quantum dots paves the way for better and better coherent manipulation of this quantum degree of freedom. This is a prerequisite for applications of quantum dot systems in quantum information and quantum sensing. Hence, this promising route of research needs to be pursued further.

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Appendix A: Overhauser field dynamics

1. Standard semiclassical approach

The Overhauser field is defined as the weighted sum of all nuclear bath spins \( B_{\text{ov}} = \sum_{k=1}^{N} A_k I_k \), with \( I_k \) being the \( k \)th bath spin, \( N \) the number of bath spins, and \( A_k \) the hyperfine coupling constants defined by (4). In a semiclassical TWA approach [15] (4), we apply this classical equation of motion

\[
\frac{d}{dt} I_k = (A_k S + g_n h n B_{\text{ext}} e_z) \times I_k,
\]

while the initial conditions are chosen randomly according to normal distributions with expectation value zero and variance \( I(I + 1)/3 \) for each component \( I^k \) [40] [43].

As a result, the initial Overhauser field also follows a normal distribution with expectation value zero and variance

\[
\text{Var}[B_{\text{ov}}^k] = \frac{I(I + 1)}{3} A_Q^2 = \frac{2}{(T_n^\alpha)^2},
\]

with \( A_Q^2 := \sum_{k=1}^{N} A_k^2 \), which defines the dephasing time \( T_n^\alpha \) of the electron spin due to the hyperfine interaction with the nuclear spins with spin \( I \). In our case, we focus on GaAs QDs so that we have \( I = 3/2 \). When studying InGaAs QDs, one needs to also account for \( I = 9/2 \) of the indium isotopes. This requires a slightly more complicated definition of the variance which includes the relative abundances of the isotopes in a QD.

2. Spectral density approach

Since QDs consist of \( N = 10^4 - 10^6 \) bath spins [26] [52] [53], the numerical treatment of the \( N \) equations of motion (A1) is infeasible. For this reason, we briefly explain the more efficient spectral density (SD) approach in the following, which we apply to calculate the dynamics of the Overhauser field consisting of an infinite number of bath spins. It is introduced in Ref. [22] and applied successfully for the description of QDs subjected to periodic pulses in Refs. [10] [13].

The SD approach allows us to study an infinite spin bath while the number of effectively coupled nuclear spins is finite and given by \( N_{\text{eff}} \approx 2/\gamma [13] [22] [17] \). Instead of calculating the time evolution of each bath spin \( I_k \) individually, we consider \( N_{\text{tr}} = \Theta(75) \) auxiliary vectors \( Q_k \) which evolve according to the equation of motion

\[
\frac{d}{dt} Q_k = (\epsilon_k S + g_n h n B_{\text{ext}} e_z) \times Q_k,
\]

where \( \epsilon_k \) is an effective coupling constant derived via application of the SD approach (see below). In the derivation of this approach [22], the hyperfine interaction of the bath spins given by the exponential parameterization (1) is represented by the linear spectral density \( W(\epsilon) = (\epsilon/\gamma) \Theta(\sqrt{2\gamma A_Q} - \epsilon) \), where \( \Theta(\epsilon) \) is the Heaviside function. This spectral density is discretized according to the following procedure. First, we divide the energy range \([0, \sqrt{2\gamma A_Q}]\) into \( N_{\text{tr}} \) intervals \( T_k := [\hat{\epsilon}_k, 1, \hat{\epsilon}_k] \) with

\[
\hat{\epsilon}_k = k \times N_{\text{tr}} \frac{\sqrt{2\gamma A_Q}}{N_{\text{tr}}}, \quad k \in \{0, 1, 2, \ldots, N_{\text{tr}}\}.
\]

The intervals become exponentially small for increasing \( k \), which is the most efficient choice [22].
out that a good choice for the number of intervals $N_{\text{tr}}$ is obtained when ensuring that $\lambda \approx 0.87$ holds for long simulations. For short simulations, we use a minimal number of $N_{\text{tr}} = 44$ auxiliary vectors $Q_k$ to ensure a minimal discretization density which still captures the correct physics. Each interval has the weight
\[ W_k := \int_{t_k}^{t_{k+1}} W(\epsilon) d\epsilon \quad \text{(A6)} \]
and the corresponding coupling strength $\epsilon_k$ is given by the average energy
\[ \epsilon_k := \frac{1}{W_k} \int_{t_k}^{t_{k+1}} \epsilon W(\epsilon) d\epsilon, \quad k \in \{1, 2, \ldots, N_{\text{tr}}\}. \quad \text{(A7)} \]
The $N_{\text{tr}}$ auxiliary vectors $Q_k$ represent the sums of the bath spins whose couplings lie within the interval $I_k$. Due to the central limit theorem, each initial component of the vectors $Q_k$ can be drawn from a normal distribution because they represent large linear sums of the bath spins, and they are uncorrelated for different $k$. Thus, we can initialize the $3N_{\text{tr}}$ components $Q^\alpha_k$, $\alpha \in \{x, y, z\}$, according to normal distributions around zero with variance
\[ \text{Var}[Q_k^\alpha] = \frac{(I + 1)}{3}. \quad \text{(A8)} \]
Finally, the Overhauser field is given by the weighted summation
\[ B_{\text{ov}} = \sum_{i=1}^{N_{\text{tr}}} \sqrt{W_k} Q_k, \quad \text{(A9)} \]
which leads to the same variance as required by Eq. [A2].

Appendix B: Alternative non-deterministic pulse descriptions

Establishing a valid non-deterministic semiclassical pulse description, which on average keeps the properties of the deterministic pulse model (7), is not straightforward. In this appendix, we discuss several alternatives to the non-deterministic pulse description (14) introduced in Sec. [II] and benchmark them against the deterministic semiclassical pulse (7) and its quantum mechanical realization used in Ref. [10] in the SML regime without NIFF. Note that in each approach, the relations (7c) and (7d) for the trion pseudospin $J_\alpha$ remain unchanged.

1. Discrete truncated Wigner approximation

As a first alternative, we apply the discrete truncated Wigner approximation (DTWA) [21] to the deterministic pulse (7). This phase space method only acts on a discrete phase space, which, in turn, gives rise to certain benefits.

In this approach, we sample each spin component $S^\alpha$ from the discrete phase space $\{+1/2, -1/2\}$ so that all quantum mechanical moments of the spin of the same component are taken into account correctly. Moreover, the spin length after a pulse is always given by $|S_a| = \sqrt{3}/2$.

The ensuring discrete distribution function is defined by
\[ P \left( S^\alpha_a = \pm \frac{1}{2} \right) = \frac{1}{2} \pm E[S^\alpha_a]. \quad \text{(B1a)} \]
\[ P \left( S^\alpha_a = -\frac{1}{2} \right) = \frac{1}{2} - E[S^\alpha_a], \quad \text{(B1b)} \]
where $E[S^\alpha_a]$ is the the mathematical expectation value of this probability distribution, which is given by Eq. (14). This approach works well as long as $|S^\alpha| \leq 1/2$, e.g., for the first pulse. But since the spin with initial length $\sqrt{3}/2$ precesses according to the equation of motion (3a), this condition does not necessarily hold for every pulse, leading to the appearance of negative probabilities in Eq. (B1). Our heuristic solution consists of effectively truncating the probability distribution, i.e., we set $P(S^\alpha_a = 1/2) = 1$ when $S^\alpha_a > 1/2$. However, this alters the resulting expectation value of the distribution and thereby, also the SML steady state. Another drawback of the DTWA consists in the broken rotational spin symmetry because certain spin axes are treated in a special way.

2. Trion probability

In this approach, we use $S^\alpha_b$ to determine the probability for the excitation of a trion. In this interpretation, the system realizes either the ground state electron spin $S$ or the trion pseudospin $J$ directly after the pulse. The circularly polarized laser pulse $\sigma^-$ only excites the electron spin if it is in the state $|\downarrow\rangle$. In the classical representation, this means that $S^\alpha_b = -1/2$ leads to the excitation of a trion, thus $S^\alpha_a = 0$ and $J^\alpha_a = -1/2$. For $S^\alpha_b = +1/2$, no trion is excited so that $S^\alpha_a = +1/2$ and $J^\alpha_a = 0$ follows. More general, the probability to find the spin in the state $|\downarrow\rangle$ and therefore to excite a trion is given by
\[ P_t = \frac{1}{2} - S^\alpha_b. \quad \text{(B2)} \]
If no trion is excited, the $z$ component of the electron spin simply takes the value $S^\alpha_z = +1/2$ while the $x$ and $y$ component are sampled from a normal distribution with expectation value zero and variance $1/4$ to account for the second moment of spin-$1/2$ operators. Alternatively, sampling from the discrete phase space introduced in Appendix [B1] is possible, but the results are worse.
Mathematically, this procedure leads to expectation values which are identical to the deterministic pulse relation \( S \). However, the same issue as discussed in Appendix \( B \) arises. Since negative probabilities can appear, the probability distribution requires needs to be truncated, i.e., we set \( P_z = 1 \) if \( S_{\perp} > -1/2 \) and \( P_z = 0 \) if \( S_{\perp} < 1/2 \). Eventually, this leads to a deviation of the expectation value from (14) and accordingly, also to a deviation from the SML steady state.

A possible solution consists of scaling the spin \( S_{\perp} \) to the Bloch sphere of spin length 1/2, before applying the pulse. We will see, however, that this procedure leads to the emergence of an unwanted phase shift.

Figure 9 shows the revival amplitudes \( S_{\perp} \) (upper panel) and the corresponding \( z \) components \( S_z \) (lower panel) for the following pulse descriptions: quantum mechanical (QM, black), deterministic semiclassical (DS, green dashed) [Eq. (7)], TWA [Eq. (14), orange dotted], DTWA [Eq. (15), blue], trion probability approach (TPA) [Eq. (B2), red dashed], and TPA with scaling to the Bloch sphere (see end of Appendix B 2, brown dash-dotted). As expected, the QM and the DS results are almost identical. Small deviations stem from the fact that the QM results are obtained for only \( N = 6 \) bath spins, which requires an additional ensemble average to get rid of finite size effects. These serve as our benchmark. They show the expected revival amplitude of the SML steady state value \( S_{SML} \approx 0.07735 \) and there is almost no difference between \( S_{\perp} \) and its \( z \) component, i.e., there is no phase shift.

The results for the TWA pulse, which we introduce and apply in the EM I of Sec. III, are in perfect agreement with the benchmark results (QM and DS). Small deviations stem mainly from the statistical nature of the ensemble average. We use \( M = 10^6 \) configurations here to calculate the ensemble average; the statistical deviations decrease with \( 1/\sqrt{M} \).

For the remaining non-deterministic pulse description, we find no agreement with the benchmark results. As expected, the DTWA and TPA pulse show a too small steady state revival amplitude. Interestingly, the results are identical apart from small statistical fluctuations in the SML regime, but the behavior in the NIFF regime is extremely different (not shown).

By scaling the spin vector \( S_{\perp} \) to the Bloch sphere before the application of the TPA, the revival amplitude \( S_{\perp} \) reaches the correct steady state. However, the revival amplitude is about two times larger than its \( z \) component, i.e., a significant phase shift is introduced by scaling to the Bloch sphere, but such a phase shift does not appear in the benchmark data.

3. Comparison to established pulse descriptions

Let us compare the various non-deterministic pulse descriptions to the established deterministic pulse relation (7) and its quantum mechanical realization used in Ref. 10 in the SML regime. Note that we do not include the trion pseudospin dynamics here because it has no important influence on the SML regime.

Figure 9 shows the revival amplitudes \( S_{\perp} \) (upper panel) and the corresponding \( z \) components \( S_z \) (lower panel) for the following pulse descriptions: quantum mechanical (QM, black), deterministic semiclassical (DS, green dashed) [Eq. (7)], TWA [Eq. (14), orange dotted], DTWA [Eq. (15), blue], trion probability approach (TPA) [Eq. (B2), red dashed], and TPA with scaling to the Bloch sphere (see end of Appendix B 2, brown dash-dotted). As expected, the QM and the DS results are almost identical. Small deviations stem from the fact that the QM results are obtained for only \( N = 6 \) bath spins, which requires an additional ensemble average to get rid of finite size effects. These serve as our benchmark. They show the expected revival amplitude of the SML steady state value \( S_{SML} \approx 0.07735 \) and there is almost no difference between \( S_{\perp} \) and its \( z \) component, i.e., there is no phase shift.

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