Nonequilibrium nuclear spin distribution function in quantum dots subject to periodic pulses

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Electron spin dephasing in a singly charged semiconductor quantum dot can partially be suppressed by periodic laser pulsing. We propose a semiclassical approach describing the decoherence of the electron spin polarization governed by the hyperfine interaction with the nuclear spins as well as the probabilistic nature of the photon absorption. We use the steady-state Floquet condition to analytically derive two subclasses of resonance conditions excellently predicting the peak locations in the part of the Overhauser field distribution which is projected in the direction of the external magnetic field. As a consequence of the periodic pulsing, a nonequilibrium distribution develops as a function of time. The numerical simulation of the coupled dynamics reveals the influence of the hyperfine coupling constant distribution onto the evolution of the electron spin polarization before the next laser pulse. Experimental indications are provided for both subclasses of resonance conditions.

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I. INTRODUCTION

Combining traditional electronics with novel spintronic devices has led to an intensive investigation of semiconductor quantum dots (QD) using electrical [1,2] or optical probes [3,4]. High localization of the electron wave function in the QD reduces the decoherence facilitated by free electron motion, but simultaneously increases the hyperfine interaction strength between the confined electron spin and the surrounding nuclear spins [4–9]. Nevertheless, QD ensembles driven by periodic circular polarized laser pump pulses provide a promising route for optically controlled quantum functionality [3,10].

A steady state of the spin system emerges from the periodic pulsing of such QD ensembles that is substantially different from its equilibrium starting point. Floquet's theorem provides a periodicity condition for this nonequilibrium steady state, which translates into a mode-locking resonance condition for the electron spin dynamics. Early on, it was conjectured that this mode-locking condition [3] leads to a nuclei-induced frequency focusing of electron spin coherence. Although the short time dephasing remains unaltered, the resonance condition partially restores spin coherence via constructive interference before the next laser pulse arrives. Hole-spin initialization and relaxation times were studied in a similar pump-probe configuration in Ref. [11].

In this paper, we derive a semiclassical approach for the coupled dynamics of the electron spin and the nuclear spins in a single negatively charged QD. The external magnetic field is applied in the Voigt geometry, i.e., orthogonal to the optical axis, and the electron spin is subject to periodic pulsing with a circular polarized laser. The presented method takes into account the hyperfine interactions between the electron and nuclear spins [5,6,12] as well as the Zeeman terms for all spins. The method is used to calculate the emerging nonequilibrium steady state induced by the periodic pulsing.

Our approach is based on the observation that the Overhauser field generated by the large number of nuclei spin behaves as a classical variable in leading order [5,12–14], particularly in a large external magnetic field. Chen *et al.* [15] showed that the quantum-dynamics of the central spin model [5,16] can be accurately approximated by expanding the path-integral representation around its saddle-point, defined by a set of classical Euler-Lagrange equations of motion. Subsequently, the quantum mechanical trace is replaced by a configuration average over all classical spin configurations [15].

The effect of laser pulses as well as the decay of the created trion, however, require a fully quantum mechanical treatment of the electron spin dynamics reflecting the probabilistic nature of the photon absorption and emission processes. In order to accommodate these quantum effects, the unique correspondence between a quantum-mechanical expectation value for a spin 1/2 and the components of the density matrix of such a spin subject to a classical magnetic field is exploited. The quantum nature of the spin-pumping and the trion decay can therefore be included into an Ehrenfest equation for the electron spin expectation value. It has almost the same analytical structure as that of a classical spin, and the quantum mechanics is encoded in the nonconstant length of the classical spin vector to account for the effect of each laser pulse and the subsequent trion decay.

Although our theoretical simulations focus on a single QD, the considerations can be extended to a QD ensemble. In a single QD, the distribution of the hyperfine coupling constants, the electronic g factor as well as the oscillator strength is fixed while they typically vary from QD to QD in a real ensemble. Furthermore, the different geometries of different QDs yield different electronic confinement potentials and consequently, different laser energies are required to pump the trion states of each QD [17,18]. A monotonous connection between the trion excitation energy and the electron g-factor g_e has been experimentally used [19] to address subsets of a QD ensemble with differently colored laser light. Since the fluctuations of the Overhauser field define the short-time dephasing time T^* [4,5,20], our calculations can be interpreted as either simulations for an ensemble of identical QDs, or the accumulated time average of many consecutive measurements of the spin-polarisation on a single QD. Experimentally, the accumulated average of a QD ensemble is recorded [3]. To account for this, we have to average the results for a single QD over a typical distribution of g_e factors as well as a distribution of times T^* as given in the experimental situation.

While the basic effect of the periodic pulsing was well understood in terms of a resonance condition for the electron spin dynamics [10], a direct experimental access to the properties of the nuclear spin bath is absent under these conditions. One of the main objectives of this paper is to clarify the dynamics of the emerging mode-locking conditions based on an analytical argument as well as a detailed analysis of the full numerical simulations. We show that some of the basic features of forming a nonequilibrium distribution function of the Overhauser field predicted by Petrov and Yakovlev [21] prevails for the proper quantum mechanical treatment of the trion decay based on a Lindblad approach.

Assuming a converged periodic Floquet state after infinitely many laser pulses, we analytically derive two steady-state resonance conditions: one is identical to the conjecture by Greilich *et al.* [10], while the second condition additionally depends on the ratio between the Larmor frequency and the trion decay rate. It turns out that these analytic predictions excellently agree with the full numerical simulations and can provide an upper bound of the maximal achievable spin polarization in such experimental setups.

Recently, a fully quantum-mechanical treatment of the problem [22] has addressed the question how the nuclear nonequilibrium distribution function emerges due to the periodic pump pulses of a QD. While Petrov and Yakovlev [21] used simplified assumptions that each pump pulse initializes the electron spin in a fully polarized state, the quantum mechanical treatment of trion excitation and the subsequent decay under reemitting a photon has been taken into account [22]. A very slow growth of a peak structure in an originally Gaussian Overhauser field distribution [5,12] has been reported where these emerging peaks can be understood in terms of resonance conditions [22,23]. In order to address the dynamics of a reasonably large spin bath consisting of N = 15-17 nuclei, however, the hyperfine interaction was perturbatively treated up to linear order in the spin-flip terms during each pulse interval. It remains unclear whether the slow growth of the nonequilibrium distribution function is related to the underestimation of the spin-flip term in the perturbation theory or is already representative for a QD comprising typically 10⁵ nuclear spins.

Since the characteristic frequency of the classical and the quantum mechanical treatment of the spin precession are identical and given by the effective Larmor frequency, we do not alter the relevant time scale by resorting to a classical treatment of the individual nuclear spins in order to treat (i) large numbers of nuclear spins and (ii) allow for an isotropic dynamics induced by the hyperfine interaction. We provide a simple scaling argument how to extrapolate the time scales for the evolution of the nonequilibrium distribution function to a realistic QD.

The theoretical simulations are augmented by experimental data recorded on an ensemble of n-doped quantum dots. Measurements address the magnetic field dependency of the mode locking amplitude as well as the Fourier transform of the electron spin dynamics, both obtained by Faraday rotation measurements [3,10]. From the Fourier transforms with sufficient resolution we indeed find clear indications for precession modes fulfilling the predicted second class of resonance conditions that have not yet been observed before. These modes become particularly prominent around 4 T, where the mode-locked spin amplitude that can be

assessed shortly before the impact of a pump pulse shows a minimum.

A. Plan of the paper

We use a semiclassical approximation to study the electron spin dynamics and the development of a nuclear spin distribution in a periodically pulsed QD system. A formalism for the simulation that incorporates a classical description for the hyperfine interaction and Larmor precession around the external magnetic field as well as for the trion decay is presented. Section II is divided in two parts: one covers the theoretical basics, the other addresses the methods used in the simulation. In the first part, the central spin model is introduced. In Sec. II B, the Lindblad formalism for the trion decay is discussed. In the following, coupled equations of motion (EOM) are found for the electron spin and the nuclear spin bath. A classical approach for the trion decay can be derived from the Lindblad formalism. Under the assumption of a frozen Overhauser field two sets of resonance conditions can be found. Lastly, the influence of the Overhauser field on the electron spin is introduced. Section III contains the results of the theoretical simulations. We start with a brief introduction of the default settings of the parameters. Those will be used to gain a fundamental understanding of the time evolution of the system. The rest of the section is devoted to the variation of parameters like the external magnetic field or the distribution of coupling constants. Section IV addresses the results of experimental studies for the electron spin precession frequency spectrum, which show clear indications for modes at both resonance conditions. The last section will summarize the results and give an outlook to further investigations.

II. MODELS AND METHODS

We aim to describe a single electron charged QD subjected to periodic laser pulses and to an externally applied magnetic field. The time scales of the system vary greatly: the time duration of the pulses (\sim 1.5 ps), the trion decay (\sim 0.4 ns) and the repetition time of the pulse (13.2 ns) [3,10]. Therefore the laser pumping will be treated quantum-mechanically, whereas we will use a semiclassical approach for the trion decay and the electron spin dephasing between two consecutive pulses.

A. Central spin model

The Fermi contact hyperfine interaction between the central electronic spin and the nuclear spins in the QD provides the largest contribution to electron spin dephasing [4] in a singly charged semiconductor QD. Other interactions such as dipole-dipole interaction [4] or the electrical quadrupolar nuclear interactions [24–27] are several orders of magnitude smaller and, therefore, will be neglected in the following [4,5,8].

The Hamiltonian of the central spin model (CSM) accounts for the effect of the external magnetic field on the electron and nuclear spins as well as the hyperfine interaction between nuclear spin bath and electron spin:

$$H_{\rm CSM} = g_{\rm e} \mu_{\rm B} \vec{B}_{\rm ext} \vec{\hat{S}} + \mu_{\rm N} \vec{B}_{\rm ext} \sum_{k=1}^{N} g_k \vec{\hat{I}}_k + \sum_{k=1}^{N} A_k \vec{\hat{I}}_k \vec{\hat{S}}.$$
 (1)

The operators \hat{S} and \hat{I}_k denote the electron spin and the k^{th} nuclear spin. N labels the number of nuclear spins.

All spins precess around the external magnetic field \vec{B}_{ext} with the Larmor frequency $\omega_e = g_e \mu_B |\vec{B}_{ext}|$ for the electron spin and $\omega_{N,k} = g_k \mu_N |\vec{B}_{ext}|$ for the k^{th} nuclear spin, respectively. The last term in (1) encodes the hyperfine interaction between central spin and nuclear spins via the Overhauser field, $\vec{B}_N = \sum_k A_k \vec{I}_k$. The feedback to the k^{th} nuclear spin is given by the Knight field $\vec{B}_k = A_k \vec{S}$. The strength of the coupling constants A_k are determined [4,5] by the probability of an electron being present at the position of the k^{th} nucleus $|\psi(\vec{R}_k)|^2$.

The fluctuations of the Overhauser field in absence of an external magnetic field, $\langle \hat{B}_N^2 \rangle$, define a characteristic time scale of the system,

$$(T^*)^{-2} = \sum_{k} A_k^2 \langle \vec{l}_k^2 \rangle, \tag{2}$$

governing the short-time electron spin decoherence. Throughout the paper, we use T^* as the characteristic time or inverse energy scale in all calculations. Note that we have absorbed \hbar in the definition of time in the numerical calculations; at the end, the time is converted back to physical units using $T^* \approx 1$ ns in order to connect with the experiments.

It is useful to introduce dimensionless coupling constants $a_k = T^*A_k$ and dimensionless magnetic fields $|\vec{b}_{ext}| = g_e \mu_B T^* |\vec{B}_{ext}| = \omega_e T^*$. This leads to the dimensionless Hamiltonian

$$\bar{H} = H_{\rm CSM} T^* = \vec{b}_{\rm ext} \hat{S} + \zeta \vec{b}_{\rm ext} \sum_k \vec{l}_k + \sum_k a_k \vec{l}_k \hat{S}, \quad (3)$$

where ζ denotes the ratio between the nuclear Zeeman and the electronic Zeeman energy. For simplicity, g_k is taken as equal for all nuclear spins. For $\ln_x \operatorname{Ga}_{1-x} \operatorname{As} \operatorname{QDs}$, $\zeta = \frac{g_k \mu_N}{g_e \mu_B} \approx$ $(800)^{-1}$ replaces the small difference in the Ga, In, and As Zeeman energies by an averaged value ζ . Recently, the effect of different nuclear spin species on the dynamics has been investigated employing a quantum mechanical perturbation theory [23]. This requires a nuclei dependent ratio ζ_k in Eq. (3), which is beyond the scope of this paper. In the experiments, the external magnetic field is applied in *x* direction, in the Voigt geometry, while the laser beam direction, which is perpendicular to this, defines the *z* direction.

B. Methods

The major challenge for the description of the pulse dynamics and the build-up of a nonequilibrium steady state in a QD ensemble subject to periodic laser pulses is the large separation of the time scales. While the laser pulse duration typically is given by $T_P = 1 - 4$ ps, and can be treated as instantaneous to a good approximation, the dephasing time due to the hyperfine interactions is three orders of magnitude larger while the pulse repetition time is $T_R = 13.2$ ns in the experiments. Since the experiments are performed at magnetic fields of the order of 1–6 T, the electronic Larmor frequency $|\vec{b}_{ext}|$ is large compared to the hyperfine interaction energy fluctuation $1/T^*$.

Electronic spin polarization is generated by resonant circular σ^+ laser pulses exciting the electron state $|\uparrow\rangle$ to a trion state $|\uparrow\downarrow\uparrow\rangle$. Spin conservation and the formation of an electron singlet prevent the excitation of the electron $|\downarrow\rangle$ state for σ^+ circular polarization. The effective *g* factor of the trion is dominated by the hole spin and turns out to be negligibly small. Therefore precession of the trion state $|\uparrow\downarrow\uparrow\rangle$ to $|\uparrow\downarrow\downarrow\downarrow\rangle$ in the external magnetic field is omitted. During the Larmor precession of light at a decay time $1/\gamma$, which is typically $0.1 - 0.2T^*$. Clearly, this process must be treated quantum mechanically by a Lindblad approach even though a simplified approach has recently been proposed [21].

The experimentally relevant time scales allow us to separate the time evolution between two pulses into two steps: (i) the laser pulse, which is treated by an instantaneous unitary transformation of the electronic part of the density operator, and (ii) the decay of the trion is accounted for by a Lindblad formalism and the simultaneous time evolution of the coupled nuclear electronic system.

For the last step, one could remain within a fully quantum mechanical description [13,20] but is limited to a relative small number of nuclear spins [20,22], or to short-time dynamics [13] using a TD-DMRG approach [28]. Alternatively, one can map the dynamics onto a set of classical equations of motion [5,15,29], which shows remarkably good agreement with the full quantum mechanical treatment [13] but is easily extendable to a large number of spins. Below, we address the key challenge of how to combine quantum and classical calculations in a systematic way to incorporate the formation on a nonequilibrium density distribution of the Overhauser field [21], which is the origin of the self-focusing experimentally observed by Greilich *et al.* [10].

1. Lindblad approach

We start from an Ising basis for the nuclear spins defined parallel to the external field denoted by $\vec{m} = (m_1, \ldots, m_N)$ where m_k is the eigenvalue of I_x for the *k*th nuclear spin, and σ for the two spin orientations of the electron spin. In that basis, the matrix elements of the density operator of the coupled nuclear-electronic system $\vec{\rho}(t)$ are denoted by

$$\rho_{(\sigma,\vec{m}),(\sigma',\vec{m'})}(t) = \langle \sigma,\vec{m} | \rho(t) | \sigma',\vec{m'} \rangle.$$
(4)

Since the nuclear Zeeman energy as well as a single coupling constant a_k are very small, the nuclear spin configurations can be treated as frozen on the very short time scale of the pulse duration [10]. For each frozen nuclear configuration $\alpha =$ $(\vec{m}, \vec{m'})$, the basis of the electron spin can be freely chosen. The states $|\uparrow\rangle, |\downarrow\rangle$ will denote the eigenvectors of σ_z with the eigenvalues ± 1 , while the electron spin Ising basis parallel to the external field is assigned to $|\uparrow\rangle_x, |\downarrow\rangle_x$. Hence we interpret $\rho_{(\sigma, \vec{m}), (\sigma', \vec{m'})}(t)$ as a matrix element of mixed Ising bases: an Ising basis for the nuclear spins defined parallel to the external field and an Ising basis for the electron spins in the *z* direction.

The eigenstates of the central spin are given by the two ground states $|\uparrow\rangle$ and $|\downarrow\rangle$ as well as its two trion states $|\uparrow\downarrow\uparrow\rangle$ and $|\uparrow\downarrow\downarrow\downarrow\rangle$. Since only σ^+ polarized light is taken into account the $|\uparrow\downarrow\downarrow\downarrow\rangle$ state will never be excited. This reduces the dimension of the electronic subspace from four to three.

Combined with an assumed effective spin 1/2 for the nuclear spins this leads to a Hilbert space dimension of $3 \cdot 2^N = 3D$, where *D* is the dimension of the Hilbert space of the nuclear spin bath.

In this paper, we restrict ourselves to ideal π pulses, which instantaneously excite the trion state $|\uparrow\downarrow\uparrow\rangle$ from the electron state $|\uparrow\rangle$. Since the simulation is done for only one QD the pulses have the same effect in each configuration. Such an ideal pulse can be described by the unitary transformation

$$\hat{T} = \mathbf{i}|\uparrow\downarrow\uparrow\rangle\langle\uparrow| + \mathbf{i}|\uparrow\rangle\langle\uparrow\downarrow\uparrow| + |\downarrow\rangle\langle\downarrow|$$
(5)

converting the initial density operator ρ^{bp} to $\rho^{\text{ap}} = \hat{T} \rho^{\text{bp}} \hat{T}^{\dagger}$. Since the pulse only affects the electronic subsystem, this transformation holds for each frozen nuclear configuration α so that we obtain D^2 independent transformations:

$$\rho_{\alpha}^{\rm ap} = \hat{T} \rho_{\alpha}^{\rm bp} \hat{T}^{\dagger} \,. \tag{6}$$

In Eq. (6), $\rho_{\alpha}^{\text{bp}}$ and $\rho_{\alpha}^{\text{ap}}$ denote 3 × 3 matrices in the enlarged electronic Hilbert space including the trion state $|\uparrow\downarrow\uparrow\rangle$ —for details, see Appendix A.

The trion decays under emission of a photon, which is accounted for by the Lindblad equation [30]

$$\dot{\rho} = \mathcal{L}\rho(t) = -i[H_{\rm S},\rho] - \gamma(s_2s_1\rho + \rho s_2s_1 - 2s_1\rho s_2).$$
(7)

The second term describes the trion decay into the electron state $|\uparrow\rangle$ by a constant decay rate γ where the two transition operators, s_1 and s_2 , are given by the projectors $s_1 := |\uparrow\rangle\langle\uparrow\downarrow\uparrow\rangle|$ and $s_2 := |\uparrow\downarrow\uparrow\rangle\langle\uparrow\downarrow\rangle|$. In an exact treatment of the CSM, the system Hamiltonian H_S would be H_{CSM} . In the frozen Overhauser field approximation (FOA), H_S only accounts for the electronic degrees of freedom.

Clearly, the Lindblad equation cannot be solved exactly for a CSM comprising of large numbers of nuclear spins since the Hilbert space grows exponentially. We are either restricted to small nuclear system sizes [20,22] or we employ the frozen nuclear approximation [5], and arrive at the independent Lindblad equations

$$\dot{\rho}_{\alpha}(t) = \mathcal{L}_{\alpha} \rho_{\alpha}(t), \qquad (8)$$

where the Liouvillian \mathcal{L}_{α} in each Overhauser field configuration includes the system Hamiltonian $H_{\rm S}(\alpha) = g_{\rm e}\mu_{\rm B}\vec{S}\vec{B}_{\rm ext} + \Delta H(\alpha)$, which describes the electronic precession in the external magnetic field and a static, configuration dependent Overhauser field. Note that $\rho_{\alpha}^{\rm ap}$ serves as starting density operator, and \mathcal{L}_{α} is time independent. The trion decay in the Liouvillian is independent of the nuclear bath configuration. Equation (8) can be formally solved via

$$\rho_{\alpha}(t) = \mathrm{e}^{\mathcal{L}_{\alpha}(t-t_0)} \rho_{\alpha}(t_0). \tag{9}$$

2. Semiclassical approximation (SCA)

The requirement to solve $D^2 = (2^N)^2$ matrix equations (9) in the frozen nuclear field approximation drastically limits the number of bath spins which can be included in a numerical simulation [22] to N < 20. For large numbers of nuclear spins contributing to an Overhauser field of a finite length, however, the central limit theorem has been used to calculate very accurately the short-time dynamics of the spin-spin correlation function using a Gaussian distributed statical classical Overhauser field [5].

Chen *et al.* systematically derived corrections to the frozen Overhauser field approximation [15] starting from the quantum mechanical path integral formulation of the problem. The path integral for expectation values uses spin coherent states for each spin which are parameterized by the solid angle. The saddle point approximation leads to (N + 1) coupled Euler-Lagrange equations [13,15,29],

$$\frac{\mathrm{d}}{\mathrm{d}t}\vec{S} = (\vec{b}_{\mathrm{N}} + \vec{b}_{\mathrm{ext}}) \times \vec{S}, \qquad (10a)$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\vec{I}_{k} = (a_{k}\vec{S} + \zeta\vec{b}_{\mathrm{ext}}) \times \vec{I}_{k}.$$
(10b)

with a remaining integral over all possible initial spin configurations. These equations describe the dynamics of coupled classical spin vectors representing the central spin \vec{S} and the nuclear spin \vec{I}_k by classical vectors. Neglecting the dynamics of the nuclear spins given by Eq. (10b) recovers the FOA of Merkulov *et al.* [5,12,31], where the average over all initial nuclear spin configurations has been replaced by a configuration average over a Gaussian distributed Overhauser field entering Eq. (10a).

A word is in order concerning the spin length. While the quantum mechanical electron spin has S = 1/2 and also a spin length of I = 1/2 is assumed for the nuclear spins we use a classical spin vector of $|\vec{I}_k| = 1$ in the numerical simulations below. Clearly, Eq. (10a) remains unaltered after replacing $\vec{S} \rightarrow \vec{S}' = \vec{S}/S$. In Eq. (10b), we replace $\vec{I}_k \rightarrow \vec{I}_k/I$ to justify the classical spin vector of $|\vec{I}_k| = 1$. This requires $a_k \vec{S} \rightarrow (Sa_k)\vec{S}/S$ and $b_N = I \sum_k a_k I_k/I$. The modified equations of motion of the SCA are then given by

$$\frac{\mathrm{d}}{\mathrm{d}t}\vec{S}' = (\vec{b}_{\mathrm{N}} + \vec{b}_{\mathrm{ext}}) \times \vec{S}', \qquad (11a)$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\vec{I'}_k = (a'_k\vec{S'} + \zeta\vec{b}_{\mathrm{ext}}) \times \vec{I'}_k, \qquad (11\mathrm{b})$$

$$\vec{b}_{\rm N} = I \sum_{k} a_k \vec{I'}_k \,, \tag{11c}$$

where all primed spin vectors are classical vectors of length unity. As long as the Overhauser field \vec{b}_N remains unaltered, the electron spin Larmor frequency is invariant of the spin length. Equation (11c) reveals that the fluctuation of the Overhauser field is proportional to the spin length [5], classically I^2 and quantum mechanically I(I + 1), which becomes identical for large *I*. Defining $\omega_{\text{fluc}} = \sqrt{\ll \vec{B}_N^2 \gg}$ the spin lengths can be absorbed into the definition of T^* or ω_{fluc} [5,26]. For S = I, the change of the classical spin length leads to a modified coupling constant $a'_k = a_k/2$ in Eqs. (11b) and (11c).

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The averaging over all Overhauser field configurations has been interpreted as an averaging over an ensemble of identical QDs [32] each characterized by a classical spin vector. In experiments, however, variations of the QD size leads to different characteristic time scales T^* . Therefore we view the averaging procedure as being equivalent to averaging over repetitive measurements [10] and will perform the averaging over different T^* values of the QDs in the ensemble in a second step.

3. Combining the Lindblad and the semiclassical approach

In order to connect the quantum mechanical treatment of the pulsed excitation and the stochastic decay of the trion with a semiclassical description between the pulses, we recall that the trace over the full Hilbert space was replaced by an integration over all initial spins in the SCA. We discretize the integration over all initial spins by generating N_C configurations comprising N different nuclear spins and one central spin, each equally distributed over the Bloch sphere, each weighted by a factor $1/N_C$.

Since the thermal energy in the experiments typically exceeds all other energy scales of H_{CSM} , the initial quantum mechanical density matrix ρ_0 is isotropic and proportional to the unity matrix: $\rho_{\alpha}(t=0) = (1/2)\underline{1}(1/D)\delta_{\vec{m},\vec{m}'}$. By resorting to an average over N_C classical configurations, we essentially replace the factor $(1/D)\delta_{\vec{m},\vec{m}'}$ by the factor $1/N_C$ and identify the label α by the classical configuration index,

$$\langle \hat{\hat{S}} \rangle = \frac{1}{D} \sum_{\alpha} \operatorname{Tr}[\bar{\rho}_{\alpha} \hat{\hat{S}}]$$
$$\approx \frac{1}{N_C} \sum_{\mu} \operatorname{Tr}[\rho_{\mu} \hat{\hat{S}}] = \ll \hat{S} \gg , \qquad (12)$$

where the trace is calculated with the 2 × 2 density matrix $\rho_{\mu} = \bar{\rho}_{\alpha} = D \times \rho_{\alpha}$ whose initial value is $(1/2)\underline{1}$. In the second line of the equation, μ labels the classical configuration and $\ll \cdots \gg$ denotes the configuration average.

In a purely classical simulation, the classical spin S_{μ} is averaged directly. Interpreting a classical spin vector \vec{S} with $|\vec{S}| = 1/2$ as the expectation value of a quantum mechanical spin 1/2 uniquely defines the corresponding 2×2 density matrix

$$\rho_{S} = \begin{pmatrix} \frac{1}{2} + S_{z} & S_{x} - iS_{y} \\ S_{x} + iS_{y} & \frac{1}{2} - S_{z} \end{pmatrix}.$$
 (13)

While a purely classical spin has a fixed length, the quantum mechanical expectation value \vec{S}_{μ} ,

$$\vec{S}_{\mu} = \text{Tr}[\rho_{\mu}\hat{S}] \tag{14}$$

can have arbitrary length reflecting the requirement for a quantum ensemble description: the effect of the laser pump pulse is an inherent statistical process.

Since classical equations of motion (10) are norm conserving for any vector \vec{S} , the restriction of a fixed spin length of the central spin is not required. The classical equations of motion (10) only faithfully replace the unitary time evolution of a quantum system under the influence of H_{CSM} .

This unitary time evolution, however, is violated by the Lindblad equation. It accounts for the build-up of spin polarization due to the laser pulse and consecutive trion decay: the length of the spin expectation value quantum mechanically calculated with $\bar{\rho}_{\alpha}$ will result in different spin polarizations from the initial spin length. This reflects the fact that even an initially pure quantum mechanical state typically will end up in a mixed state after the trion decay.

The quantum mechanical evolution of the electronic density matrix including the trion decay in a static magnetic field is determined by Eq. (7). This requires the solution of eight differential equations for the 3×3 matrix since the trace remains conserved at all times. We will show below, that these

equations are partially decoupled and are equivalent to those of the spin and trion expectation values.

In order to connect the quantum mechanical treatment of the laser pulse with the semiclassical equations of motion (10), we start from the FOA, i.e., treat \vec{b}_N as static. After the laser pulse, the expectation value of any given local observable \hat{O} in the electronic subspace can be calculated from the dynamics of the density matrix (7):

$$\frac{d}{dt}\langle \hat{O} \rangle = \mathrm{i} \mathrm{Tr}[\rho(t)[H_{\mathrm{S}}, \hat{O}]] - \gamma \mathrm{Tr}[\Delta \rho_L \hat{O}], \qquad (15)$$

where

$$\Delta \rho_L = |\uparrow\downarrow\uparrow\rangle\langle\uparrow\downarrow\uparrow|\rho(t) + \rho(t)|\uparrow\downarrow\uparrow\rangle\langle\uparrow\downarrow\uparrow| - 2|\uparrow\rangle\langle\uparrow|P_{\rm T}(t)$$
(16)

and $P_{T(t)} = \langle \uparrow \downarrow \uparrow | \rho(t) | \uparrow \downarrow \uparrow \rangle$ denotes the trion occupation probability. It is straightforward to derive the equation of motion for the electron spin expectation values

$$\frac{d}{dt}\langle \hat{\vec{S}} \rangle_{\mu} = \vec{b} \times \langle \hat{\vec{S}} \rangle_{\mu} + \gamma P_{\mathrm{T}\mu}(t) \vec{e}_{z} , \qquad (17)$$

which has a very intuitive interpretation. While the trion decays back into the spin-up state contributing only to the spin polarization in z direction, the electronic spin precesses around the effective magnetic field $\vec{b} = \vec{b}_{\rm N} + \vec{b}_{\rm ext}$.

The solution of this set of equations requires the dynamics of the source term determined by the differential equation

$$\frac{d}{dt}P_{\mathrm{T},\mu}(t) = -2\gamma P_{\mathrm{T},\mu}(t), \qquad (18)$$

which also is derived from (15). It has the simple analytic solution

$$P_{\mathrm{T},\mu}(t) = P_{\mathrm{T},\mu}(0)e^{-2\gamma t}, \qquad (19)$$

where $P_{T,\mu}(0)$ is the trion occupation directly after the laser pump pulse. These define the first four equations determining the evolution of the nine matrix elements of the quantum mechanical density operator.

Since the trace is conserved, there are four more differential equations required for the full solution of the density matrix. The remaining four other differential equations only involve trion off-diagonal matrix elements and also have a trivial exponential decaying solution. Furthermore, these off-diagonal matrix elements do not couple to the differential equations determining the spin dynamics and can be neglected.

Consequently, we can include the Lindblad decay into the SCA replacing (10a) by

$$\frac{d}{dt}\vec{S}(t) = (\vec{b}_{\rm N} + \vec{b}_{\rm ext}) \times \vec{S}(t) + \gamma P_{\rm T\mu}(0)\vec{e}_z e^{-2\gamma t} , \quad (20)$$

where the time t is measured relative to the last pulse.

Within the FOA, this differential equation can be even solved analytically. Without the source term, the homogeneous solution reads [5]

$$\vec{S}_{\text{hom}} = (\vec{A}\vec{n})\vec{n} + [\vec{A} - (\vec{A}\vec{n})\vec{n}]\cos(\omega_L t) + \vec{n} \times [\vec{A} - (\vec{A}\vec{n})\vec{n}]\sin(\omega_L t), \qquad (21)$$

where the Larmor frequency is given by $\omega_L = |\vec{b}| = |\vec{b}_N + \vec{b}_{ext}|$, and $\vec{n} = \vec{b}/\omega_L$ denotes the unit vector in the direction of the effective magnetic field. The solution is parametrized by

the three component vector \vec{A} , which would be equal to $\vec{S}(0)$ in the absence of the source term.

The inhomogeneous solution has the form

$$\vec{S}_{\rm in} = \vec{C} e^{-2\gamma t}.$$
(22)

Defining the rotation matrix $\underline{\underline{M}}$ such that $\underline{\underline{M}}\vec{v} = \vec{n} \times \vec{v}$, we obtain

$$\vec{C}_{\mu} = -\frac{P_{\mathrm{T}\mu}(0)}{2} \left[\underline{\underline{1}} + \frac{\omega_L}{2\gamma} \underline{\underline{M}}_{\mu} \right]^{-1} \vec{e}_z.$$
(23)

From the total solution $\vec{S}_{\mu}(t) = \vec{S}_{\text{hom},\mu}(t) + \vec{S}_{\text{in},\mu}(t)$ and the initial condition, we determine the

$$\vec{A}_{\mu} = \vec{S}_{\mu}(0) - \vec{C}_{\mu},$$
 (24)

where $\vec{S}_{\mu}(0)$ is the electronic spin expectation value of the configuration μ after the pulse. Since \vec{C}_{μ} has a negative sign, the spin polarization grows from $|\vec{S}_{\mu}(0)|$ directly after the pulse to $|\vec{A}_{\mu}|$, once the trion is completely decayed.

Within the SCA, we can even relax the constraint of a constant Overhauser field by allowing $\vec{b}_N \rightarrow \vec{b}_N(t)$. Then, the feedback of the central spin onto the nuclear spins and visa versa is included at any time. But the analytic solution derived above is no longer valid.

Let us summarize the individual steps of our hybrid quantum-classical approach to a OD subject to periodic laser pulses. Initially, (i) we generate N_C classical spin configurations labeled by μ and comprising a central spin and N nuclear spins, each equally distributed over the Bloch sphere. (ii) We freeze the nuclear spins and convert each central spin S_{μ} of the classical configuration into a 2 × 2 density matrix ρ_{μ} using Eq. (13). This matrix is extended to a 3 \times 3 matrix spanned by the enlarged Hilbert space including the trion. (iii) Then, the laser pulse is applied, described by Eq. (6), and quantum mechanical expectation values \vec{S}_{μ} and P_T are calculated directly after the pulse, which define the initial conditions for solving the coupled equations (11b), (11c), and (20) for the time interval up to $t = T_{\rm R}$. For the next pulse, we go back to step (ii). In order to calculate expectation values, we average the quantity of interest over all configurations μ for the given time *t*.

Our quantum-classical hybrid approach clearly reveals, that by the necessary quantum mechanical treatment of the laser pulses the simplified quantum to classical mapping of the spin degree of freedoms does not hold for the electron spin. \vec{S} loses its classical interpretation even within a single configuration. The requirement for a density matrix description has its deeper root in the statistical nature of the photon absorption which is linked to the quantum efficiency of the process. Although we only consider resonant photon absorption, the theory can be simply extended to nonresonant absorption by replacing *T* in Eq. (6) by the appropriate unitary time evolution operator.

4. Resonance conditions

Before we present the full numerical solution in Sec. III, we analytically extract a steady-state solution from the differential equation (20) using simplified approximations. As we demonstrate below, the resonance conditions obtained

in such a way agree remarkably well with our simulations providing an a posteriori justification of these simplifications.

Since the central spin dynamics is much faster than the nuclear spins, we treat the nuclear spin dynamics as nearly frozen on the time scale T_R , i.e., the nuclear Zeeman term is neglected. Furthermore, only the *x* component of the Overhauser field is taken into account since the *y* and *z* components can be viewed as small perturbations transversal to the large external magnetic field.

For the π pulses discussed in this paper, we relate the electron spin expectation values prior to the $N_{\rm P}$ th pulse, $\vec{S}^{\rm bp}(N_{\rm P}T_R)$, to the one after the laser pulse,

$$\vec{S}^{ap}(N_{\rm P}T_{\rm R}) = \left(0, 0, \frac{1}{2}\left(S_z^{\rm bp} - \frac{1}{2}\right)\right)^{\rm T},$$
 (25)

by applying the pulse operator (5). The corresponding trion occupation probability $P_{T,\mu}(0) = (S_z^{bp} + \frac{1}{2})$ is generated by the spin-up component of the density matrix. Therefore the trion and the electron spin state after each pulse depend only on S_z^{bp} , the *z* component of the electron spin right before the pulse.

These conditions are inserted into the analytical solution for the spin-expectation values derived above:

$$\vec{S}(t) = \begin{pmatrix} 0 \\ -A_z \sin(\omega_{\rm L}t) + A_y \cos(\omega_{\rm L}t) - A_y e^{-\overline{\gamma}t} \\ A_y \sin(\omega_{\rm L}t) + A_z \cos(\omega_{\rm L}t) - A_y \frac{\overline{\gamma}}{\omega_{\rm L}} e^{-\overline{\gamma}t} \end{pmatrix}$$
(26)

with $\overline{\gamma} = 2\gamma$ and the prefactors

$$\begin{split} A_y &= \frac{\omega_{\rm L}\overline{\gamma}}{\overline{\gamma}^2 + \omega_{\rm L}^2} \frac{2S_z^{\rm bp} + 1}{4}, \\ A_z &= \frac{\overline{\gamma}^2}{\overline{\gamma}^2 + \omega_{\rm L}^2} \frac{2S_z^{\rm bp} + 1}{4} + \frac{2S_z^{\rm bp} - 1}{4} \end{split}$$

In general, the static approximation of the Overhauser field is not justified, since the effect of the Knight field on the nuclear spin is required for the energy conservation law in the absence of the laser pulses as well as the rearrangement of the Overhauser field distribution as a function of time. Since we are targeting the steady state of the electron spin under periodic laser pumping, we (i) refer to the Floquet periodicity condition for the *z*-component of the electron spin

$$S_z(T_{\rm R}) = S_z^{\rm bp} \tag{27}$$

and (ii) demand that the feedback of the Knight field to the nuclear spins vanishes in average over the course of one pulse repetition, i.e.,

$$\langle \vec{I}_k \rangle_{T_{\rm R}} = \langle a_k \vec{S} \times \vec{I}_k \rangle_{T_{\rm R}} = 0.$$
(28)

For an almost static nuclear spin vector \vec{I}_k , this translates into the vanishing of the average effect of the central spin onto each nuclear spin over the pulse period T_R ,

$$\langle \vec{S} \rangle_{T_{\rm R}} = \frac{1}{T_{\rm R}} \int_0^{T_{\rm R}} \vec{S}(t) dt = 0$$
⁽²⁹⁾

independent of the coupling constant a_k . Note that the electron spin lacks a *x* component after the pulse, and this component remains its zero value in a static effective magnetic field in *x* direction at all times.

Combining these two conditions with the analytic solution (26) reveals the $1/\omega_L$ dependence of the averaged Knight field, see Appendix B, and leads to the following equation:

$$\frac{\overline{\gamma}}{\omega_{\rm L}}(1 - \cos(\omega_{\rm L}T_{\rm R})) - \sin(\omega_{\rm L}T_{\rm R}) = 0, \qquad (30)$$

determining the set of Floquet values of the effective Larmor frequency $\omega_{\rm L}$ under these assumptions. Since the external magnetic field is fixed, the different values of $\omega_{\rm L}$ translate to different steady-state values of the Overhauser field in *x* direction.

One class of solutions for $\omega_{\rm L}$ fulfils the resonance conditions

$$\omega_{\rm L} T_{\rm R} = 2\pi n \qquad \text{with} \quad n \in \mathbb{Z} \tag{31}$$

that was already discussed by Greilich *et al.* [10]. They are only dependent on the external magnetic field and independent of the trion decay rate. A second class of solutions is determined by the transcendent equation

$$\omega_{\rm L}T_{\rm R} = 2\arctan\left(\frac{\omega_{\rm L}}{\overline{\gamma}}\right) + 2\pi n, \qquad n \in \mathbb{Z},$$
 (32)

where the ratio of the Larmor frequency to the decay rate $\overline{\gamma}$ generates an additional phase shift. Since $|\vec{b}_{ext}| \gg |\vec{b}_N|$ and the principal determination $\arctan \in (-\frac{\pi}{2}, \frac{\pi}{2})$ is monotonically increasing, 2 $\arctan(\frac{|\vec{b}_{ext}|}{\overline{\gamma}})$ serves as a good approximation. For large external magnetic fields ($\omega_L \gg \overline{\gamma}$), the second class of solutions leads to Larmor frequencies placed at odd resonance conditions, $\omega_L T_R = \pi (2n + 1)$, while for small magnetic fields these additional peaks are brought closer to the even resonances.

In the central spin dynamics, both Overhauser peak classes are combined. The first class of solutions, defined by the even resonance condition (31), always is connected with an electron spin that is aligned in negative z direction right before the pulse independent of the external magnetic field, $S_z^{bp,1} = -1$. Then, the π pulse has no effect on the electron spin dynamics and \vec{S}^{bp} is identical to \vec{S}^{ap} .

Though the Larmor frequency ω_L is strongly dependent on the magnetic field for the second class of solutions, the spin vector is always aligned in the positive z direction, $S_z^{bp,2} = 1/3$. The π pulse leads to a flip of the CS from $S_z^{bp,2} = 1/3$ to $S_z^{ap,2} = -1/3$. Note that these are the only two polarizations $S_z^{bp,2}$ where the effect of the laser pulse conserves the spin length and $|S_z^{bp,2}|$ is in a fixed point.

5. Mode locked electron spin

In order to set the stage for the analysis of the full numerical simulations, we discuss the potential impact of the resonance condition onto the central spin dynamics as well as the Overhauser field distribution. These Overhauser field distribution functions,

$$p(b_{\mathrm{N},i}) = \ll \delta \left(B_{N,i}^{\mu} - b_{\mathrm{N},i} \right) \gg , \qquad (33)$$

provide important statistical information about the nuclear spin system, where the symbol $\ll \cdots \gg$ denotes the configuration average, and i = x, y, z.

Prior to applying the periodic laser pulses, we assume the system to be in equilibrium and the high temperature limit to

be valid, since the thermal energy at ~ 6 K is much larger than the hyperfine interaction. Therefore the nuclear spins can be regarded as classical-spin vectors that are uniformly distributed on the unit sphere. By using the law of large numbers this leads to Gaussian distributed Overhauser fields $p(b_{N,i})$ in all spatial directions [5].

To investigate the influence of the periodic pulse sequence on the electron spin dynamics in a simplified toy model, we first combine the precondition of the Gaussian envelope with the resonance condition presented in the previous section. When the system reaches its steady state, we assume that each class of resonance conditions leads to δ peaks in $p(b_{N,x})$ inside a Gaussian distribution. Using Eq. (26), the solutions for central spin dynamics for different Larmor frequencies are superimposed and weighted according to the Gaussian envelope. Within the scope of this simple model we assume that both resonance conditions contribute equally to the combined dynamics.

In order to relate the external field strength to the even resonance condition, we define K' as

$$K' = \frac{|\vec{b}_{\text{ext}}|T_{\text{R}}}{2\pi}.$$
(34)

For $K' \in \mathbb{Z}$, a free electron spin subject to an external magnetic field \vec{b}_{ext} fulfills the resonance condition. Off-resonance external magnetic fields can be quantified via a deviation ΔK from the next integer value, i.e., $K' = K + \Delta K$ with $K \in \mathbb{Z}$.

In the example shown in Fig. 1, we set K = 200, which corresponds to a field strength of about 2 T. For such a strong external field, the second class of resonance condition yields peak positions at about $\pi (2n + 1)/T_{\rm R}$. Note that the maximum length of the classical spin vector is 1.

The class of even resonance conditions leads to a central spin which is aligned fully in the negative z direction before the pulse. Hence the electron spin polarization can fully be transferred to the next pulse period since the π pulse does not have any affect, and the amplitude of the electron spin signal is maximal as shown in Fig. 1(a).

The electron spin configurations for the odd resonance conditions, however, are aligned in positive z direction. A full polarization of the electron spin, however, is not possible according to Eq. (25). This subclass also shows perfect revival as depicted in Fig. 1(b). The perfect revivals in each subclass at the end of the pulse period being a consequence of the resonance condition, is destroyed by the superposition of both since the spins point in opposite directions at the end of the period. When weighting both subclasses equally the revival is significantly reduced as demonstrated in Fig. 1(c). The revival can be completely suppressed when weighting the first and the second subclass in the ratio 1:3—not shown here.

III. RESULTS

A. Distributions of hyperfine couplings

While the short-time dynamics of the QD is governed by T^* and therefore independent of a particular a_k distribution the long-time dynamics is influenced by the probability density function $p(a_k)$ of the coupling constants. Several different distributions have been used for the CSM [6,7,13,20,33], ranging from the simple box model [21], which assumes



FIG. 1. Toy model for the central spin dynamics. (a) and (b) give the central spin dynamics for one class of resonance conditions for $T_{\rm R} = 13.5 T^*$. (c) combines the two classes with equal weight. The insets show the two oscillating electron spin components immediately before the next pulse.

equal coupling constants $a_k = a = 1/\sqrt{N}$, $\forall k$, to the more elaborate distributions of coupling constants $p(a_k)$ [4,6,7,20].

A simplified constant distribution has advantages concerning computation time whereas others provide a more realistic description of the hyperfine coupling. The coupling constants are proportional to the electronic probability of presence at the *k*th nuclear spin given by the envelope of the electron wave function $\psi(\vec{R}_k)$,

$$\psi(\vec{R}_k) \propto \exp\left(-\frac{1}{2}\left(\frac{r}{L_0}\right)^2\right)\Big|_{r=|\vec{R}_k|},$$
 (35)

at the location of the nucleus R_k . L_0 is the characteristic length scale of the QD and of the order of $L_0 \approx 5$ nm. For a spherical QD, a probability density function

$$p(a) = -\frac{\xi}{2r_0^{\xi}} \frac{1}{a} \left(\ln\left(\frac{a_{\max}}{a}\right) \right)^{\frac{\xi-2}{2}}$$
(36)

has been derived [20] where r_0 is the ratio between an artificial cut-off R and L_0 . a_{max} is the largest occurring coupling constant and contains information about the underlying material. ξ gives the dimension of the QD. For a spherical QD, $\xi = 3$, the coupling constants are defined by $a = a_{max} \exp(-r_0^2 x^{2/3})$ with the uniform random number $x \sim \mathcal{U}(0,1)$. With increasing cut-off ratio r_0 an increasing number of nuclei are located inside the sphere with radius $R = r_0 L_0$ around the quantum dot. For $r_0 \gg 1$, a large number of nuclei are included that couple exponentially weak to the localized electron spin. In order to generate a meaningful configuration of a_k , the larger r_0 , the large N must be to adequately represent this distribution. For N = 100 nuclear spins a cut-off ratio of $r_0 = 1.5$ was chosen.

Furthermore, an exponential distribution function used in the literature [6,27,34,35]

$$a_k = C e^{-(k-1)\lambda},\tag{37}$$

with k = 1, ..., N and $C = \sqrt{\frac{1 - \exp(-2\lambda)}{1 - \exp(-2\lambda N)}}$, represents a 2d QD, $\xi = 2$, with a harmonic potential [6]. λ determines the spread of the coupling constants.

B. Definitions of the parameters

The dynamics of the electron spin $\langle S_z \rangle$ and the distribution of the Overhauser field $p(b_{N,i})$ with i = x, y, z in a system subjected to periodic laser pulses is investigated. The parameters are chosen to correspond to the experimental setup [10].

Unless stated otherwise, these parameters will stay the same in the following sections where only one parameter is varied. We use a bath size of N = 100 nuclear spins and average over $N_{\rm C} = 10^5$ configurations. The length of the classical nuclear spin vector is $|\vec{I}_k| = 1$. This is also the maximal length for the electron spin vector $|\vec{S}_{\rm max}| = 1$. Therefore Eqs. (13) and (20) have been adjusted accordingly as discussed above.

For the theoretical simulations, we set the separation time between two instantaneous pulses $T_{\rm R} = 13.5 T^*$ for convenience while the experimental constrains lead to $T_{\rm R} = 13.2$ ns [10]. The trion decay rate is given by $\gamma = 10 \frac{1}{T^*}$. We have used the conversion factor $T^* \approx 1$ ns for simplicity to make contact with the experiments. The scope of this paper is to provide a basic understanding of the dynamics observed in periodically driven QDs and not the fitting of a specific experiment.

We convert $|\dot{b}_{ext}|$ in a dimensionless number K' defined in Eq. (34) to clearly signal a resonance condition of the external magnetic field. $b_{ext}(K = 200) \approx 93 (T^*)^{-1}$ is equivalent to $B_{ext} \approx 2$ T using the proper conversion constants. The modification of K' from an integer value to an arbitrary real number $(K + \Delta K)$ can be used to understand deviations from the resonance conditions which can also arise in a QD ensemble due to different g factors of individual QDs.

The strength of the nuclear Zeeman coupling is defined by the factor $\zeta = \frac{g_L \mu_N}{g_c \mu_B}$ between the nuclear and electron Zeeman energy as introduced in Eq. (3). The coupling of the nuclear spins to the external magnetic field can be explicitly neglected in the theoretical simulations by setting $\zeta = 0$.

We begin with the so-called box model [21,33], i.e., we set all $a_k = a = 1/\sqrt{N}$ to reveal the basic properties of the dynamics before presenting data obtained by numerically

very expensive simulations. For nuclear spins coupling with individual a_k to the central spin, N + 1 coupled equations (10) have to be solved. By using the box model the equations for the nuclear spins collapse to a single EOM for the Overhauser field and the set of equations is reduced to two. We use the Runge-Kutta fourth-order method to solve the differential equations. The step width has to be adapted according to the strength of the external magnetic field to resolve the Larmor precession of the central spin. For an external field of K = 200 a step width of $\sim 0.001T^*$ has proven to be sufficient.

We start with a completely unpolarized system. At t = 0, the first pulse is applied. The distribution of the Overhauser field is measured immediately before the next pulse.

C. Benchmarking the semiclassical equation of motion

In order to benchmark the quality of the SCA [12,13,31] employed in this paper, we compare the spin correlation function $C_2(t) = \langle S_z(t)S_z \rangle$ in a finite magnetic field $b_x = 10$ obtained with the two classical approaches, the SCA and FOA, with the quantum mechanical results calculated using a Chebyshev expansion technique (CET) [36] and via exact diagonalization (ED) of the Hamiltonian. Our results are summarized in Fig. 2.

Note that the dephasing time scale is governed by the fluctuation of the Overhauser field $\langle \vec{B}_N^2 \rangle$ [5] where the average spin length enters. Since the quantum mechanical simulations have been performed with I = 1/2, we have absorbed the difference in the Overhauser fluctuations between $\langle \vec{I}_k^2 \rangle = |\vec{I}_k|^2 = 1$ of the classical simulation and $\langle \vec{I}_k^2 \rangle = I(I+1) = 3/4$ of the quantum simulations into rescaled coupling constants $a_k \rightarrow \tilde{a}_k = a_k \sqrt{3/4}$ for the SCA and the FOA.

While the SCA and the FOA provide identical results they also agree remarkably well with the two quantum mechanical data sets for N=10 (ED) and N = 20 (CET). The difference



FIG. 2. (a) Comparison between spin correlation function $C_2(t) = \langle S_z(t)S_z \rangle$ in a finite magnetic field $b_x = 10$ calculated using different quantum mechanical approaches, the CET with N = 20 and ED with N = 10, and the two classical approaches, SCA and FOA, with N = 100 spins and $N_C = 100\,000$. The CET data have been taken from Fig. 7 in Ref. [20]. (b) Difference between the CET and the SCA data (red solid line) and the ED and the SCA data (blue dashed line).

between classical and the quantum simulations depicted in Fig. 2(b) can be attributed to 1/N effects, which are suppressed using a large number of bath spins in a time-dependent density matrix renormalization group approach [13].

Since the spin length is conserved in the SCA and the feedback involves always a coupling constant, the differences can be absorbed into the definitions of the coupling constants or the reference time and energy scales, respectively. After establishing this quality of the SCA, we use the energy and time scales as defined in Sec. II A throughout the paper. For a classical spin length of 1, the coupling constants used are

$$a'_{k} = \frac{a_{k}}{2} = \frac{A_{k}/2}{\sqrt{\sum_{k=1}^{N} \frac{1}{4}A_{k}^{2}}} = \frac{A_{k}}{\sqrt{\sum_{k=1}^{N} A_{k}^{2}}} .$$
 (38)

D. Nonequilibrium Overhauser field distribution function: Nuclear self-focusing

1. Influence of the number of pulses

The electron spin dynamics is dominated by the precession around the strong external magnetic field. The electron spin component parallel to the external magnetic field remains at approximately zero since the laser pumping only generates a spin polarization in the *z* direction. The components perpendicular to the external magnetic field show the electron spin precession as demonstrated in Fig. 3. The first pulse at t = 0 depletes the $|\uparrow\rangle$ state of the previously unpolarized electron spin. Therefore the electron spin starts precessing from $\vec{S}(t=0) = \vec{S}^{ap} = -0.5 \vec{e}_z$. The trion decay leads to a steady increase in the electron spin polarization on a time scale of $0.1 - 0.2T^*$.

While coherent oscillations are observed on a very short time scale, defined by the inverse Larmor frequency, the hyperfine interaction leads to dephasing, which is governed by T^* , see Fig. 4. While the electron spin dephases completely after the first pulse, we observe a revival of electron spin polarization after the second pulse to an amplitude of $|\vec{S}| \approx$ 0.14 just before the next laser pulse arrives. After that, the central spin revival amplitude slowly grows with an increasing number of pulses.



FIG. 3. Precession of the electron spin after the first pulse. After the pulse the x and y component are zero and the z component is given by $(S_z^{bp} - 1)/2$. The analytical solution of the exponential trion decay has been added as dashed line.



FIG. 4. Dynamics of the *z* component of the electron spin during the time interval between two pulses for four different numbers of pulses calculated for the box model. The parameters for this simulation are: $\zeta = 1/800$ for the nuclear Zeeman strength and equal coupling constants $a_k = a \forall k$, a pulse repetition time of $T_R = 13.5T^*$, an external magnetic field of about 2 T (K = 200) which is applied in resonance $\Delta K = 0$. The pulses are instantaneous π pulses, and the trion decay rate is set to $\gamma = 10\frac{1}{T^*}$. N = 100 nuclear spins and $N_C = 100\,000$ classical configurations are taken into account.

The central spin dynamics is directly connected to the three distributions of the Overhauser field $p(b_{N,x})$, $p(b_{N,y})$, and $p(b_{N,z})$. The evolution of $p(b_{N,x})$ with the number of pulses is shown for the box model in Fig. 5. At t = 0, the Overhauser field is unpolarized, implying that all Overhauser field components follow a normal distribution $\mathcal{N}(0, (\langle \vec{l}_k^2 \rangle/3 = 1/3))$.

If the system is subjected to periodic pump pulses the distributions of the Overhauser field components perpendicular to the external magnetic field do not change from the initial Gaussian distribution. However, a new distribution emerges for $b_{N,x}$. Though the envelope of the distribution stays Gaussian, peaks begin to emerge at pronounced positions that become more distinct with time. We have identified two subsets of peaks. The distance between every other peak is given by the resonance condition, $\Delta b_{N,x} = 2\pi/T_R$.

Despite the strong approximations made in Sec. II B 4 on the resonance condition, the peak structure calculated in the fully numerical simulation of the EOM of the SCA, shown in Fig. 5, agrees remarkably well with the theoretical predictions for the resonance condition, which have been added as vertical dotted and dashed-dotted lines in the figure. We only observe deviations of 1%-2% and up to 9% at most.



FIG. 5. Panel (a) shows the influence of the number of pump pulses on the *x* component of the density distribution of the Overhauser field $b_{N,x}$ for K = 200. At the beginning of the pulse sequence $b_{N,x}$ is normally distributed. $b_{N,x} \sim \mathcal{N}(0,1/3)$. This scaled normal distribution is the envelope of the emerging density distribution. The vertical lines indicate the two subclasses of theoretical peak positions defined by the equations (31) and (32). (b) The corresponding *y* and *z* component of $p(\vec{b}_N)$ after 20 000 pump pulses.

2. Influence of the external magnetic field strength

The external magnetic field has two functions: (i) it induces a coherent oscillation of the spin polarization and (ii) it can also suppress dephasing stemming from the long-time fluctuations of the Overhauser field. It has been shown, that the accuracy of the FOA approximation [5] increases with increasing magnetic field [12,14,20]. Only in the theory of higher-order correlation functions, additional processes have to be included in order to make connection to the experiment [27].

The strength of the external magnetic field plays an important role in the development of the peak structure of the Overhauser field distribution. In this section, we examine the dependence on the external magnetic field as well as the resonance conditions (32) and (31). A low magnetic field allows for a fast build-up of the Overhauser field distribution due to the $1/\omega_{\rm L}$ dependency of the Knight field after integrating Eq. (10b).

The Overhauser field distribution $p(b_{N,x})$ is plotted for four different resonant magnetic field values K = 50, 100, 150, and 200 after 20 000 pulses in Fig. 6. The inset focusses on one peak at $b_{N,x} = 0$ belonging to the even resonance



FIG. 6. Overhauser field distribution along the external magnetic field direction for different strengths K of $\vec{b}_{ext} = \frac{2\pi}{T_R} K \vec{e}_x$. The inset shows the peak for the even resonance condition at $b_{N,x} = 0$ and the shifted peak given by the ratio $\omega_L/\overline{\gamma}$, see Eq. (32), after 20 000 pulses.

condition and one peak corresponding to Eq. (32). While the even resonance peaks located at positions independent on the external magnetic field values, the peaks following (32) are shifted away with increasing field strength *K*, as predicted by Eq. (32).

The peak positions of the two classes of peaks are well described by the analytical predictions. The additional features that become apparent in the simulations cannot be derived from the analytical results: the weight of each class of peaks in the combined Overhauser field distribution. For strong external magnetic fields, the peaks at even resonance are still sharp, while the second subclass of peaks have a less distinct shape.

3. Electron spin revival

As an experimentally accessible quantity through the modelocking amplitude, the electron spin revival merits a more indepth investigation. The electron spin dynamics is intertwined with the Overhauser field distribution. It determines the revival behavior since the superposition of configurations with different Larmor frequencies suppresses the growth of the central spin revival. That raises the question, which properties of $p(b_{N,x})$ influence the final revival amplitude.

The first class of peaks in the Overhauser field distribution is independent of the external magnetic field for all integer values of K. Since the period length of all even frequencies contributing to the electron-spin precession fit into $T_{\rm R}$ as integer, the central spin configurations are always aligned in negative z direction before the pulse.

For configurations characterized by the second resonance condition, the orientation of the electron spin prior to the next laser pulse depends on the external magnetic field strength: Contrary to the results of the simple toy model it can acquire a spin polarization in y direction just before the next laser pulse that does not influence the value of the spin-polarization after the pulse. The contribution of those configurations to the total signal is determined by the spectral weight of the peaks in the distribution function that cannot be obtained from the resonance condition.

Figure 7 shows the influence of the external magnetic field on the amplitude and the z component of the electron spin revival measured directly before the next pulse as function of



FIG. 7. Revival amplitude and S_z component of the electron spin for external magnetic fields $|\vec{b}_{ext}(K)| = 2\pi K/T_R$ applied in the *x* direction.

the pulse number $N_{\rm P}$. For external magnetic field strengths $K \leq 100$, the revival amplitude decreases in the long-time limit. Here the phase shift leads to a alignment of the central spin in the *y* direction before the pulse which does not influence the pumping process as is seen in Eq. (25).

For larger magnetic fields, i.e., K > 100, the electron spin polarization is aligned in z direction. Due to the mismatch in the probability weight of the resonance conditions the revival amplitude increases. The peaked nonequilibrium Overhauser field distribution, however, emerges slower for increasing magnetic fields due to the $1/\omega_L$ dependency of the averaged Knight field, see Eq. (29), leading to a slower increase of the revival. The electron spin polarization is not yet converged after 20 000 pulses as seen in the two left panels in Fig. 7.

E. Scaling behavior with the number of nuclear spins

In the experimental setup, the data are measured after an initial pulsing period which lasts from a few seconds up to 20 min [10]. For a laser repetition of ~13.5 ns this corresponds to 74×10^6 pulses per second. Such large time scales are impossible to achieve with our simulations even for the simplified box model. Therefore it is useful to derive and exploit a scaling relation associated with the number of nuclear spins in order to extrapolate the possible steady state of the system.

In Fig. 8, the time evolution of the Overhauser field distribution for different numbers of nuclear spins is shown: the larger the number of nuclear spins, the slower the build-up of $p(b_{N,x})$. The distribution $p(b_{N,x})$ is plotted for different combinations of N and pulse numbers $N_{\rm P}$ for a constant ratio $v_P = N_{\rm P}/N = 20, 200, 2000$: $p(b_{N,x})$ is universal and only depends on the ratio v_P .

This observed scaling behavior is attributed to the dependence of the Knight field on the strength of the coupling



FIG. 8. Time evolution of the Overhauser field distributions parallel to the external magnetic field for different numbers of nuclear spins. The three panels show $p(b_{N,x})$ for three different but fixed ratios $v_P = N_P/N = 20, 200, 2000, \text{ and } K = 200.$

constants, Eq. (10b), and to the influence of the Overhauser field on the central spin, Eq. (10a). Since the electron spin dynamics is fed back to the nuclear spins via the coupling constant $a_k = 1/\sqrt{N}$ the build-up scales with $a_k^2 \propto 1/N$. Consequently, the slower feedback of the electron-spin dynamics onto the $p(b_{N,x})$ with increasing number of nuclei must be compensated by an additional number of laser pulses.

Although we have only demonstrated this scaling property for the box model, we will show below that a qualitatively similar scaling behavior prevails for an arbitrary distribution function p(a) when T^* is used as a reference time scale independent of N. We will exploit this scaling law to perform simulations with as few nuclear spins as possible and extrapolate our results to the realistic number of nuclear spins in a QD. The results obtained for N = 10 nuclei and 20 000 pulses are therefore equivalent to those of 10^5 nuclei and 2×10^8 pulses, corresponding to approximately 2 sec in a typical experimental setup.

The amplitude of the electron spin revival for different numbers of nuclear spins is depicted in Fig. 9 versus $v_P =$



FIG. 9. Revival amplitude of the electron spin for different numbers of nuclear spins. Parameters as in Fig. 8.

 N_P/N following the same scaling law. Since the steady state is approached but has not been reached even for N = 10 and 20 000 pulses, we conjecture that we would need another factor 10–100 more pulses to achieve final convergence. This would translate to reaching the steady state after approximately half a minute to several minutes of pulsing, which is in the same order of magnitude as in the experiments [10].

F. Influence of an external magnetic field off resonance

For a given applied external magnetic field and a fixed laser repetition time $T_{\rm R}$, an individual QD may not fulfill the resonance condition due to its electron $g_{\rm e}$ factor leading to a noninteger value of K' in (34). We have introduced the parameter ΔK to represent the distance of K' to the closest integer value K in order to measure the distance from the integer resonance condition.



FIG. 10. (a) Density distribution of the *x* component of the Overhauser field b_N for deviations from the resonance ΔK , recorded for K = 200 after the 20 000th pulse. (b) Shifted distribution $\overline{b}_{N,x}$ by $\frac{2\pi\Delta K}{T_p}$. The dashed lines: the corresponding shifted Gaussian envelope.



FIG. 11. Dynamics of the *z* component of the central spin for deviations from the resonance ΔK after the 20 000th pulse. (Top) Self-focusing of the electron spin immediately before and after the pulse. (Middle) Envelope of the ensemble average. (Bottom) Frequency shifts at intermediate times.

 $p(b_{N,x})$ is shown for different ΔK in Fig. 10. In all distributions, the distance between every other peak remains constant, and the envelope follows a Gaussian distribution with a mean value of zero and a variance of 1/3. Depending on the magnitude of ΔK , however, the peak positions shift to adjust for the two resonance conditions for the Overhauser field. After accommodating displacement induced by the off-resonance external magnetic field into the Overhauser field by plotting $p(b_{N,x})$ vs $\overline{b}_{N,x} = b_{N,x} + 2\pi \Delta K/T_{\rm R}$ the peak positions coincide. The peak heights, however, are asymmetric due to the shifted Gaussian envelope as illustrated in the lower panel of Fig. 10.

These shifted resonance positions are understood in terms of the resonance conditions (31) and (32) where the effective Larmor frequency enters rather then the external magnetic field. Consequently, our calculations back the conjectured notion [10] of a self-focusing central spin dynamics by the dynamical redistribution of $p(b_{N,x})$ due to the periodic laser pumping. This is illustrated in Fig. 11 where the averaged electron spin response is plotted for two different off-resonant external magnetic fields in comparison with a resonant field. The top panel demonstrates the congruent dynamics immediately after and before the pulse. Only at intermediate times, small dephasing between the response of different QDs are observable, as shown in the bottom two panels of Fig. 11.

G. Single QD vs QD ensemble

The different QDs in an ensemble not only differ in their g factors but also in their hyperfine constants a_k . Since it has been established [5] that the key quantity for describing



FIG. 12. Dynamics of the *z* component of the electron spin for different scaling factors η of the time scale T^* . (a) Dynamics of $S_z(t)$ after the 20 000th pulse. (b) The modulus of the revival of the electron spin vs the number of pulses. The external magnetic field is given by K = 200.

the decoherence induced by the hyperfine interaction is given by T^* , we parameterize the individual difference of a QD to a fictitious reference QD characterized by T^* via a scaling factor η

$$T_{\eta}^* = \eta T^*. \tag{39}$$

 η depends on the different growth processes and the distribution of radii of the QD. Here we investigate only small variations from $\eta = 0.9$ to 1.1: larger η implies a slower dephasing of the central spin. The difference in the central spin dynamics for three different T_{η}^{*} is depicted in Fig. 12(a). T_{η}^{*} determines the characteristic time scale of the initial decoherence as well as of the revival since it defines the width of the Gaussian envelope function of the central spin dynamics.

Figure 13 shows that the variation of η does not affect the peak positions of the distribution. We can conclude that the subset of QDs resonantly pumped by the laser pulse leads to an in-phase interference of the central spin dynamics. Therefore the results obtained by the simulation of a single QD help understanding the dynamics of the whole QD ensemble.

The peak height, however, increases with decreasing η as expected from the feedback mechanism of the Overhauser field and the Knight field: the smaller η , the larger the hyperfine coupling, the faster the build-up of the distribution function. Figure 12(b) also illustrates this effect of T_{η}^{*} onto the time evolution of the revival amplitude of the central spin. Since we already discussed the influence of the number of nuclear spins



FIG. 13. Density distribution of the *x* component of the Overhauser field b_N after 20 000 pulses for different scaling factors η of the time scale T^* .

N onto the time evolution, we can plot the amplitude versus N_P/η^2 to accommodate the leading effect of η . The plots demonstrate the scaling, confirming the underlying feedback mechanisms between electron and nuclear spin system via the Overhauser field and Knight field. However, deviations are observable for $\eta = 1.1$. We attribute that to the fact that the ratio T_R/T_{η}^* changes in comparison to Sec. III D 1 where this ratio was kept constant.

H. Influence of nuclear Zeeman effect

While in experiments, the nuclear Zeeman effect cannot be switched off, we performed numerical simulations for the different ratios $\zeta = \frac{g_k \mu_N}{g_e \mu_B} = 0$, 1/800, and 1/500. As discussed above, $\zeta = 1/800$ corresponds to the typical experimental situation of a GaAs based QD and has been used in all previous calculations of this paper. $\zeta = 1/500$ is the highest realistic ratio given by the *g* factor for ⁷¹Ga with $g_k = 1.7$.

We found a striking difference in the revival amplitude for $\zeta = 0$ in comparison to $\zeta > 0$ as shown in Fig. 14. The data for $\zeta = 1/800$ included here have already been plotted in Fig. 7. While the build-up of the revival amplitude increases slightly by artificially doubling of the nuclear Zeeman term, the $\zeta = 0$ result shows a fundamentally different behavior. Initially, the revival spin polarization is identical for all cases, since it is of purely electronic origin. After some 100 pulses, the feedback of the electron spin polarization on the nuclear spin system



FIG. 14. Evolution of the electron spin amplitude for different ζ . The data for $\zeta = 1/800$ are taken from Fig. 4. Parameters as in Fig. 7.



FIG. 15. Density distribution of the *x* component of the Overhauser field b_N for different ratios $\zeta = \frac{g_k \mu_N}{g_e \mu_B}$.

becomes relevant. For $\zeta = 0$, the revival amplitude rapidly decreases and is stabilized at a rather low value of 0.06.

We present the corresponding Overhauser field distribution $p(b_{N,x})$ in Fig. 15. While the shape of the envelope remains Gaussian and the distance as well as the position of the peaks stays the same, the weights of these peaks differ significantly. Only marginal differences are observed for the two finite ζ values. For $\zeta = 0$, the weights have shifted almost completely to the subset of peaks connected to the resonance condition (32) corresponding to an additional phase shift of $\Delta\omega_{\rm L}T_{\rm R} = \Delta\phi = 2 \arctan(\omega_{\rm L}/\overline{\gamma})$, accumulated during the laser repetition time $T_{\rm R}$, compared to the integer resonance condition (31). Our findings perfectly agree with a recent fully quantum-mechanical investigation of the mode locking [22] in the absence of the nuclear Zeeman effect.

In order to gain some better understanding of this surprising decay, we used the distribution $p(b_{N,x})$ as a guide and resort to the toy model presented in Sec. II B 5. Peaks are found in $p(b_{N,x})$ fullfiling both resonance conditions, (31) and (32). Assuming a ratio of 1 : 3 between Gaussian envelope function corresponding to the peaks defined by (31) and respectively the peaks defined by (32), allows one to superimpose the results for the toymodel depicted in Figs. 1(a) and 1(b) with these modified spectral weights. This leads to a finite spin-polarization after the laser pulse, which completely destructively interferes before the next laser pulse as depicted in Fig. 16.

Therefore the rapid decrease of the revival amplitude for $\zeta = 0$ plotted in Fig. 14 to a small finite value is related to the strong weight imbalance between the two subsets of peaks. A slightly different broadening and deviations from the trial ratio 1 : 3 are responsible for a small but finite revival amplitude.

We emphasize that the toy model phenomenologically explains the low revival amplitude but not the deeper reason for the strong weight imbalance between the peak heights of the two subclasses. It has been conjectured, that the imbalance between the peak weights of the two resonance conditions might be attributed to the nuclear spin precession. In our SCA, we do not see any indication of the reported quantum mechanical effects [23]. No indication for a transfer of weight between both resonance conditions when altering ζ or K were observed in the results obtained by our approach.



FIG. 16. The toy model for a ratio of 1:3 between the peaks of the resonance subclasses shows no electron spin revival.

I. Influence of the distribution function p(a)

In this section, we extend our investigation to the influence of different distributions p(a) for the coupling constants a_k on $p(b_{N,x})$. The distribution used for the data labeled $\xi = 3$ is defined by Eq. (36), while for $\xi = 2$ the exponential distribution of the coupling constants in a two-dimensional QD is given by $a_k \propto \exp(-\lambda(k-1))$ with k = 1..N and $\lambda = (r_0^2/N) = 0.1$ [35,37,38], see Eq. (37).

Figure 17 shows $p(b_{N,x})$ for these two nonconstant p(a) and a fixed number of laser pulses in comparison with the box model, where $a_k = 1/\sqrt{N}$. The distribution of the Overhauser fields still features the two classes of peaks inside the Gaussian envelope. The differences can be seen in the speed of the Overhauser field build-up. Distributions with nonequal coupling constants lead to a faster development of the Overhauser field distribution.

This corresponds to a faster build-up of the revival for nonconstant p(a), see Fig. 18. Since the coupling constant enter quadratically into the change of the Overhauser field,

$$\frac{d}{dt}\vec{b}_N = \sum_k a_k^2 \vec{S} \times \vec{I}_k + \zeta \vec{b}_{\text{ext}} \times \vec{b}_N , \qquad (40)$$

its time dependency is dominated by the larger coupling constants for fixed T^* . A nonconstant distribution is therefore



FIG. 17. Density distribution of the *x* component of the Overhauser field $b_{\rm N}$ after 20 000 pulses for different distributions of the coupling constants. The external magnetic field is given by K = 200. N = 100 and a cutoff radius $r_0 = 1.5$ in (36) for $\xi = 3$. For a $(\xi = 2)$ -dimensional QD, $\lambda = 0.1$ in Eq. (37), i.e., $r_0 \approx 3.16$.





FIG. 18. Revival of the central spin amplitude for different distributions of the coupling constants. Parameters as in Fig. 17.

equivalent on a reduced number of nuclear spins in the box model plus distribution specific corrections, cf. Sec. III E.

1. N dependent scaling behavior for distributed coupling constants

In order for a potential speed-up the numerics, the scaling behavior with the number of nuclear spins for nonconstant p(a)is very important. Contrary to the box model, each spin must be simulated individually hence the run time is proportional to $N \cdot N_P$, and the validity of the scaling argument is even more desirable. To test its applicability the distribution given by Eq. (36) with $\xi = 3$ and $r_0 = 1.5$ was chosen.

The results for $p(b_{N,x})$ using the same distribution function p(a) is shown in Fig. 19 for the combinations $(N, N_P) = (100, 20\,000)$, (10, 2000), (10, 4000). Clear deviations from the $v_P = N_P/N$ scaling established only for the box model are noticeable: $p(b_{N,x})$ for $(100, 20\,000)$ almost coincides with the results for the combination (10, 4000). While in the box model, all nuclear spins rotate synchronized, in general, different nuclear spins have different precession speeds.

We have demonstrated that v_P has to be replaced by a distribution dependent scaling variable $x_P = v_P f(p(a), N)$ where the deviation from the box model scaling has to be included in the unknown correction f(p(a), N) depending on the distribution function p(a) as well as the total number of samples taken. We can estimate the ratio f(p(a), 100)/f(p(a), 10) = 2 for the single data point provided by Fig. 19: we need a larger number of pulses compared to number of nuclei to achieve the same scaling behavior exhibited in the box model. This implies that $f(p(a), N_A) < f(p(a), N_B)$ if $N_A < N_B$. This shows that computation time in the full classical model can be reduced by a smaller system size not only because the argument presented in Sec. III E still holds but also because, in contrast to the box model, less nuclear spin EOM are required to be solved.

IV. EXPERIMENTAL STUDIES OF THE MODE SPECTRUM

Experimental access to the precessional mode spectrum can be gained through Faraday rotation measurements, in which the impact of the periodic pump pulses onto the electron spins in quantum dots is traced by a linearly polarized probe pulse whose polarization change is measured after transmission through the sample. The spin precession dynamics about a perpendicular magnetic field is determined by varying the delay between pump and probe, from which the precessional mode



FIG. 19. Scaling behavior for N = 100 and 10 with coupling constants which are distributed according to Eq. (36) with $\xi = 3$ and $r_0 = 1.5$ in an external magnetic field of K = 200. (a) shows that the combination $(N, N_P) = (100, 20000)$ coincides with (10,4000), whereas (10,2000), which was equivalent to the first tuple in the box model now corresponds to lower pulse count. (b) shows the Overhauser field distribution for $(N, N_P) = (10, 20000)$.

spectrum can be retrieved by taking the Fourier transform. Further, by choosing a delay between pump and probe that is only slightly shorter than the pulse separation time, we can measure directly the mode-locking amplitude due to the spin revival.

Corresponding studies on InGaAs/GaAs quantum dot ensembles so far had revealed only modes which fulfill the condition that the precession frequency is an integer of the laser repetition rate according to Eq. (31) and dominate the spectrum (we call them integer modes in the following for brevity). Indications for modes that do not fulfill this condition but can be associated with modes fulfilling Eq. (32) had not been observed.

We have carefully repeated Faraday rotation studies in order to find indications for the additional modes predicted by Eq. (32). Details about the experiments can be found in Refs. [3,10]. The challenge in these experiments is to scan a large enough temporal range in time to obtain sufficient resolution in frequency space. This is complicated by the variation of the electron *g*-factor in the studied dot ensembles, which leads to a fast dephasing of the signal and a corresponding broadening of the precession modes after Fourier transformation. Further, also a more complex form



FIG. 20. (a) Faraday rotation measurements as function of delay between pump and probe for magnetic fields applied in the Voigt-configuration varied between 0 and 6 T. (b) The amplitude of the mode-locked signal before the pump pulse as derived from these measurements as function of the external magnetic field.

of the hyperfine coupling or additional interactions such as dipole-dipole couplings may lead to a more complex behavior of the experimental data.

As suggested by the theory, some indications for the additional modes may be found from the amplitude of the mode-locking signal right before the next pump pulse where the spin revival occurs. The modes that fulfill the integer spin revolution criterion (31) add constructively to this amplitude. On the other hand, the modes associated with Eq. (32) can add to the amplitude if their frequency is not too different from the integer modes. However, if they are located around the middle between these modes, their orientation is opposite to the one of the modes fulfilling Eq. (31). These modes then destructively contribute to the total amplitude of the mode-locked signal.

The interplay of these two types of modes can be varied through varying the magnetic field strength. Corresponding magnetic field measurements from B = 0 up to 6 T are shown in Fig. 20(a), where we focus on the amplitude of the modelocked signal right before the pump. Clearly the amplitude

of this signal shows a nonlinear dependence with increasing magnetic field as confirmed by the magnetic field dependence of the mode-locked signal amplitude shown in Fig. 20(b). This strong variation may be related to the calculation results in Fig. 6, which indicate that the signal amplitude does not show a simple variation with magnetic field, but a much more complex behavior, even though the results there are not fully converged.

From the data in Fig. 20 one can in particular see that the mode-locked amplitude becomes particularly weak at about 4 T. This may correspond to a situation where the modes according to Eq. (31) and those according to Eq. (32)almost compensate each other. For other magnetic fields the noninteger modes after Eq. (32) influence the mode-locking amplitude apparently much weaker.

To get a more direct proof of these modes we have irradiated the quantum dot sample for an extended period of pump pulses and have switched off then the pump, to monitor the free evolution of the spin ensemble. The ensemble dynamics then shows revivals that occur periodically with a separation equal to the separation between the laser pulses in the previously applied pump protocol. To obtain sufficient resolution, we have recorded the Faraday rotation signal over several of these echoes as long as they show significant amplitudes.

Figure 21 shows corresponding Faraday rotation traces (top panel, recorded at 2 and 4 T) and the corresponding Fourier transforms (bottom panel). Indeed, the spectrum at 2 T is dominated by the integer spin revolution modes. However, at 4 T, side modes appear, whose frequencies do not fulfill the criterion of Eq. (31), which have not been reported before. We want to highlight that these modes are prominent around the field strengths where the mode-locked spin amplitude shows a minimum, providing a consistent phenomenology. This is a clear signature that indeed not only the integer precession modes after Eq. (31) appear, but also additional modes contribute to the time-periodic steady state.

The goal of this experimental augment is the demonstration that the precessional mode spectrum is more complex than being just given by Eq. (31), rather than to claim quantitative agreement with the calculations of spectral positions and amplitudes of the additional modes according to Eq. (32). Such agreement cannot be expected, not only because of the ensemble study but also because of the much larger number of nuclei of about 10^5 in each dot in combination with a more complex distribution of hyperfine couplings.

Additional interactions such as the electric quadrupolar interaction [14,25,26] as well nuclear dipole-dipole interactions [4] neglected in the simulations are also expected to lead to a broadening of the peaks in the Overhauser distributions and, therefore, to a reduction in the steady-state revival amplitude. While the experiments clearly reach the steady state, the theoretical revival amplitude has not been converged even after 20 000 pulses, as can be seen from Fig. 7.

The experimental data presented in Fig. 20 clearly demonstrate a nonmonotonic dependency of the mode locking amplitude on the external magnetic field. The discussion of the toy-model in the Secs. II B 5 and III H suggests that an almost complete vanishing of the mode locking amplitude might originate in the different amplitude ratios for the even and the odd resonance revival contributions.



FIG. 21. (a) Faraday rotation measurements of the spin dynamics recorded after pumping the system by periodic pulses with 13.2 ns separation. The pumping is then switched off and the Faraday rotation is recorded over longer time scales in which periodic revivals occur due to the previous periodicity of the pumping. (b) Taking the Fourier transform of the traces provides sufficient accuracy to resolve the precession modes. In particular, at B = 4 T, additional modes can be seen which are an indication for the additional modes according to Eq. (32). The curves are vertically shifted for clarity.

V. SUMMARY AND CONCLUSION

We have derived a semiclassical description of the system, also encompassing the trion decay, for the simulation of a periodically pulsed QD. Using the FOA, we derived two classes of steady-state resonance conditions: one depends only on the repetition rate of the pulses, $\omega_L T_R = 2\pi n$ and the other is also influenced by the trion decay rate via $\omega_L T_R = 2 \arctan(\omega_L/\gamma) + 2\pi n$. By the means of a simple toy model, we have analytically shown how the Overhauser field distribution and the electron spin dynamics, especially the revival of the electron spin immediately before the next pulse, are connected in the limit of large external magnetic fields.

Nuclear self-focussing was demonstrated in the build-up of the Overhauser field distribution as well as in the revival of the central spin signal employing the full semiclassical simulation of the model for equal coupling constants. The theoretical predictions of the peak positions also hold for nonconstant Overhauser fields with only a small margin of error. For large external magnetic fields $b_{\text{ext}}(K > 100)$, the peaks are placed at integer multiples of π and the electron spin revival increases over time, while the behavior for smaller external magnetic fields exhibits peaks shifted by the arctan and an electron spin revival decrease with an increasing number of pulses.

It has been shown that larger numbers of pulses are accessible in the box model at the same computational effort by reducing the system size and exploiting the scaling properties defined by the variable $v_P = N_P/N$. This scaling argument is used to make conjectures about the steady state for realistic numbers of nuclear spins after several seconds of pulsing that is not directly accessible to our numerical simulations.

We have investigated the QD ensemble features by including the effects of g-factor variations as well as the change of the characteristic time scale T^* from QD to QD. We have demonstrated that the electron spin dynamics shortly after and shortly before each pulse is essentially independent of the individual properties of each QD, and the steady state is determined by a Floquet condition. The Overhauser field distribution displayed self-focussing by shifting the peak positions to accommodate the resonance conditions. This was reflected by the congruent central spin dynamics immediately before and after the pulse. The different hyperfine coupling constants in each QD lead to a rescaling of the characteristic time scale T^* . Larger hyperfine couplings do not only cause a shorter dephasing time but also induce a faster build-up of the Overhauser field distribution and the electron spin revival. At the end, the Floquet condition imposes the self-focussing superposition of the dynamics of different QDs and a congruent central spin behavior. Therefore the investigation of the dynamics in a single QD can be used to gain an understanding of the ensemble properties.

The different isotopes of the QD are modeled by the ratio ζ between the nuclear Zeeman energy and the electron Zeeman energy. While realistic, nonzero values of ζ lead to a similar behavior in the Overhauser field and the central spin dynamics, $\zeta = 0$ stands out. The peaks of the class of odd resonance condition are pronounced and only a minuscule electron spin revival is observed similar to what has been reported for a fully quantum mechanical treatment of the problem for a small number of nuclei [22].

For nonequal coupling constants, the computation time increases drastically since all EOM for each individual spins have to be solved in order to achieve reliable results for the long-time asymptotic. The basic features such as the position of the Overhauser peaks or the increase of electron spin revival remain untouched. The build-up speed of the peak amplitudes, however, as well as the spin revival amplitude is different for different distributions of hyperfine fine couplings for the same number of pulses. While a reduced number of nuclear spins still leads to a faster convergence to the steady state the scaling behavior is not as pronounced as it is for the box model: the reduction of the number of nuclei in the simulation is less efficient.

One of the main findings of the calculations is the claim of the existence of additional precession modes besides those described by Eq. (31). Only those had been reported in experimental studies so far. By designing experiments with proper resolution in frequency space, we could indeed resolve additional modes which may be related with those fulfilling Eq. (32). These modes should lead to a reduction of the spin revival, which has been confirmed for the magnetic field strengths where they appear most prominently in the spectra. On the other hand, at field strengths where they hardly are observable the spin mode-locking amplitude is large. It will be an effort for future activities to provide a quantitative comparison of experimental data with model calculations. This will require elaborating tools (spectroscopy on refined samples) by which the precession spectra can be measured with even higher resolution in combination with calculations that are extended towards the steady state and in which further relevant interactions are included.

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APPENDIX A: UNITARY TRANSFORMATION OF DENSITY OPERATOR VIA AN IDEAL π LASER PULSE

The density operator of the electronic subsystem including the trion is transformed according to

$$\rho^{\rm ap} = \hat{T} \rho^{\rm bp} \hat{T}^{\dagger}, \tag{A1}$$

where \hat{T} is a unitary operator accounting for the laser pulse. Under resonance conditions, one finds (5)

$$\hat{T} = \mathbf{i}|\uparrow\downarrow\uparrow\rangle\langle\uparrow|+\mathbf{i}|\uparrow\rangle\langle\uparrow\downarrow\uparrow|+|\downarrow\rangle\langle\downarrow| \tag{A2}$$

and its the matrix representation

$$\underline{\underline{T}} = \begin{pmatrix} 0 & 0 & i \\ 0 & 1 & 0 \\ i & 0 & 0 \end{pmatrix}$$
(A3)

for an ideal π pulse.

Starting from the initial density matrix for the central spin in a single classical configuration,

$$\rho_{\mu}^{\rm bp} = \begin{pmatrix} \rho_{\uparrow\uparrow} & \rho_{\uparrow\downarrow} & \rho_{\uparrow T} \\ \rho_{\downarrow\uparrow} & \rho_{\downarrow\downarrow} & \rho_{\downarrow T} \\ \rho_{T\uparrow} & \rho_{T\downarrow} & \rho_{TT} \end{pmatrix}, \tag{A4}$$

we arrive at

$$\rho_{\mu}^{\rm ap} = \begin{pmatrix} \rho_{TT} & i\rho_{T\downarrow} & \rho_{T\uparrow} \\ -i\rho_{\downarrow T} & \rho_{\downarrow\downarrow} & -i\rho_{\downarrow\uparrow} \\ \rho_{\uparrow T} & i\rho_{\uparrow\downarrow} & \rho_{\uparrow\uparrow} \end{pmatrix}.$$
(A5)

Assuming that the trion was completely decayed, this matrix reduces to

$$\rho_{\mu}^{\rm ap} = \begin{pmatrix} 0 & 0 & 0\\ 0 & \rho_{\downarrow\downarrow} & -i\rho_{\downarrow\uparrow}\\ 0 & i\rho_{\uparrow\downarrow} & \rho_{\uparrow\uparrow} \end{pmatrix} = \begin{pmatrix} 0 & 0 & 0\\ 0 & \frac{1}{2} - S_z & S_y - iS_x\\ 0 & S_y + iS_x & S_z + \frac{1}{2} \end{pmatrix},$$
(A6)

so that the initial electron spin is away aligned in z direction after the pulse $\vec{S}(0) = 1/2((S_z - 1/2))\vec{e}_z$.

APPENDIX B: INTERIM RESULTS FOR ANALYTICAL STEADY-STATE SOLUTION

 S_z^{bp} can be derived from the steady-state condition $S_z^{\text{bp}} = S_z(T_{\text{R}})$,

$$S_{z}^{\text{bp}} = \frac{1}{2A} (\overline{\gamma} \omega \sin(\omega T_{\text{R}}) - \omega^{2} \cos(\omega T_{\text{R}})), \qquad (B1)$$

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with

$$A = (\omega^2 + \overline{\gamma}^2)(2 - \cos(\omega T_{\rm R})) - \overline{\gamma}\omega\sin(\omega T_{\rm R}) - \overline{\gamma}^2\cos(\omega T_{\rm R}).$$
(B2)

Then the z component of the time averaged central spin is given by the expression

$$\langle S_z \rangle_{T_{\rm R}} = \frac{1}{2AT_{\rm R}} [\overline{\gamma}(1 - \cos(\omega T_{\rm R})) - \omega \sin(\omega T_{\rm R})].$$
 (B3)

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