Semi-Classical Model for the Dephasing of a Two-Electron Spin Qubit Coupled to a Coherently Evolving Nuclear Spin Bath

Semi-classical model for the dephasing of a two-electron spin qubit coupled to a coherently evolving nuclear spin bath

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We study electron spin decoherence in a two-electron double quantum dot due to the hyperfine interaction, under spin-echo conditions as studied in recent experiments. We develop a semi-classical model for the interaction between the electron and nuclear spins, in which the time-dependent Overhauser fields induced by the nuclear spins are treated as classical vector variables. Comparison of the model with experimentally-obtained echo signals allows us to quantify the contributions of various processes such as coherent Larmor precession and spin diffusion to the nuclear spin evolution.

I. INTRODUCTION

In recent years, electron spin qubits in solid-state quantum dots have emerged as promising candidates for the implementation of quantum information processing[1–8]. The confined electrons in these devices can be precisely manipulated using microwave frequency electromagnetic fields and/or nanosecond-scale pulses of nearby electrostatic gates, while maintaining spin coherence over much longer times. The main source of decoherence in such qubits is the hyperfine interaction between the electron spins and the nuclear spins of the host lattice[8–15]. Through this interaction, the nuclear spin bath produces a fluctuating effective Zeeman field on the electron spins. However, the timescale for evolution of this so-called "Overhauser field" is typically much longer than that required for manipulation of a single qubit. Therefore, dynamical decoupling techniques[16,17] based on fast control of the qubit can be employed to partially eliminate decoherence due to the interaction with the nuclear spins. Recent experiments confirm that such techniques can be used to extend qubit coherence times by a few orders of magnitude, up to approximately 200 μs[1,18].

Usually, interactions between a single electron spin and many weakly interacting spins (as described by the "central spin problem"[19]) can lead to complicated evolution of the joint quantum system[20–31]. Moreover, the nuclear spin bath may maintain coherence over a long time, which may in general result in coherent back-action on the electron spin. Indeed, using the Keldysh technique and a non-trivial re-summation of diagrams, the authors of Refs. 32,33 analyzed this problem, and predicted periodic collapses and revivals of the electron spin coherence over a specific range of external magnetic field strengths. This phenomenon was subsequently observed in two-electron spin echo measurements[18].

Despite the apparent complexity of the system, there are several reasons why one might expect to find a more intuitive semi-classical description of the electron spin dynamics. First, due to the small nuclear Zeeman energy, the initial state of the nuclear spin bath is well described by an infinite temperature (completely random) state. Second, the state of the nuclear spin bath is not measured in the experiment, and the experimental outcome is an average over many runs. We note that the semi-classical approximation of the nuclear spin system has been used to describe a variety of other interesting phenomena in quantum dots[21,28,30,31].

The aim of this paper is to demonstrate that a wide array of complex dynamical phenomena in two-electron spin echo measurements, such as those of Ref. 18, can be understood within the context of a semi-classical treatment of the nuclear spin bath. We first show that, within a simple model which neglects the effects of the Knight shift and the dipolar interaction between nuclear spins, the semi-classical treatment reproduces the expressions for the spin echo signal obtained in Refs. 32,33, where a summation of diagrams in a perturbative quantum mechanical treatment was used. We then present a more detailed microscopic model, which incorporates the Knight shift and the nuclear dipole-dipole interactions, as well as inhomogeneous hyperfine and Zeeman couplings. The semi-classical treatment for this model was sketched in the supplementary material of Ref. 18. Here we provide a systematic discussion of this semi-classical treatment, which relies on a low-order expansion of the qubit evolution in the inverse of the number of nuclear spins and in the hyperfine coupling. Using this approach, we identify the relevant physical processes which govern nuclear spin evolution and electron spin coherence, as measured by the spin-echo signal. The excellent agreement between the model and the experimental data that was shown in Ref. 18 lends additional justification to the approximations underlying the semi-classical approach.

The paper is organized as follows. In Sec. II we describe the two-electron-spin qubit system and outline the main physical processes which govern the qubit dynamics. In Sec. III we review the Hahn spin-echo sequence. In Sec. IV we describe the Hamiltonian of the two-electron-spin system and the nuclear spin bath. We then derive a simpler effective Hamiltonian through a perturbative treatment of the Overhauser field. In Sec. V we show that a semi-classical model based on this Hamiltonian reproduces the collapse and revival phenomenon. A more complete semi-classical approach is derived and justified in Sec. VI. There we include the effect of the dipolar coupling between nuclear spins and the back-action of the Knight field on the nuclear spin precession. Conclusions and discussion are presented in Sec. VII.

II. THE TWO-ELECTRON-SPIN QUBIT

We consider a qubit consisting of two electrons in a double quantum dot, in the regime where the two electrons are well separated with one electron occupying each dot. A uniform in-plane magnetic field $\mathbf{B}_{\rm ext} = B_{\rm ext}\hat{\mathbf{z}}$ is applied along the z-axis, with $B_{\rm ext} > 0$. The qubit Hilbert space is spanned by the singlet spin state, $|S\rangle = \frac{1}{\sqrt{2}}\left(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle\right)$, and the triplet state with zero net spin projection on the z-axis, $|T_0\rangle = \frac{1}{\sqrt{2}}\left(|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle\right)$. In this notation, the two arrows represent the orientations of the electron spins in each of the two dots, measured relative to the external field direction, $\hat{\mathbf{z}}$. Due to the large Zeeman splitting, this qubit subspace is energetically isolated from the two other two-electron triplet spin states $|T_+\rangle = |\uparrow\uparrow\rangle$ and $|T_-\rangle = |\downarrow\downarrow\rangle$, in which both electron spins point parallel or anti-parallel to the direction of the magnetic field.

The energy splitting between the two qubit states $|S\rangle$ and $|T_0\rangle$ can be controlled on a fast timescale by rapidly tuning nearby electrostatic gates which modify the confining potential for the quantum dot, and hence control the shape of the two-electron wave function. For tunings where the two electrons are held far apart in the ground state, i.e. where the electrons are separated into different dots, the states $|S\rangle$ and $|T_0\rangle$ are degenerate. However, when the potential is tuned to favor partial double occupation of one dot, the difference in orbital symmetry between $|S\rangle$ and $|T_0\rangle$ leads to an exchange energy splitting J between them.

In materials such as the commonly employed III-V compounds, the confined electron spins interact with a background of nuclear spins in the host lattice. This interaction is produced by the hyperfine coupling $H_{\rm HF} = \sum_{d,n} A_{d,n}\mathbf{I}_n\cdot\mathbf{S}_d$. Here the index n labels all nuclear spins, described by the operators $\{\mathbf{I}_n\}$, $d = L, R$ labels the electron spins in the left and right dots, described by the operators $\{\mathbf{S}_d\}$, and the coupling constants $\{A_{d,n}\}$ depend on the local electron spin density, as will be described in detail below. Defining the nuclear (Overhauser) field operator in dot d as $g^*\mu_B\mathbf{B}_{{\rm nuc},d} \equiv \sum_n A_{d,n}\mathbf{I}_n$, we write the "effective" electron spin Zeeman Hamiltonian as

$$H_{el} = g^*\mu_B\left(\mathbf{S}_L\cdot\mathbf{B}_{{\rm tot},L} + \mathbf{S}_R\cdot\mathbf{B}_{{\rm tot},R}\right), \qquad (1)$$

with

$$\mathbf{B}_{{\rm tot},d} = \mathbf{B}_{\rm ext} + \mathbf{B}_{{\rm nuc},d}. \qquad (2)$$

Here $g^* \approx -0.4$ is the electron effective g-factor in GaAs and μ_B is the Bohr magneton. Equations (1) and (2) describe the Zeeman coupling in a system of two isolated electrons, where each electron is subjected to an effective field which is the vector sum of a uniform static external magnetic field $\mathbf{B}_{\rm ext}$, and a local, operator-valued, Overhauser field $\mathbf{B}_{{\rm nuc},d}$.

We can gain extremely useful intuition about electron spin dynamics in this system by treating the operator-valued Overhauser fields $\mathbf{B}_{{\rm nuc},L}$ and $\mathbf{B}_{{\rm nuc},R}$ as classical (time-dependent) vector variables. In typical GaAs dots, the Overhauser field is produced by a large number of nuclear spins in each dot, $N_d \approx 10^6$. When all the nuclear spins are polarized, the resulting effective Overhauser field has a magnitude $|\mathbf{B}_{{\rm nuc},d}| \sim 5$ T. However, under experimental conditions, where thermal fluctuations randomize the directions of all nuclear spins, the typical values of $|\mathbf{B}_{{\rm nuc},d}|$ are reduced by a factor $\sqrt{N_d}$, and are of order 1 mT. For strong enough external fields, $B_{\rm ext} \gg |\mathbf{B}_{{\rm nuc},d}|$, the net fields $\mathbf{B}_{{\rm tot},L}$ and $\mathbf{B}_{{\rm tot},R}$ are nearly parallel to $\mathbf{B}_{\rm ext}$. Under these conditions, the two-dimensional qubit subspace is only slightly perturbed by the misalignment of local fields, and remains energetically isolated from the other two-electron spin states. To leading order in $\frac{|\mathbf{B}_{{\rm nuc},d}|}{B_{\rm ext}}$, the effect of the nuclear fields is simply to induce a Zeeman splitting between $|\downarrow\uparrow\rangle$ and $|\uparrow\downarrow\rangle$, proportional to the difference in z-projections of the effective fields in the two dots, $\Delta B^z_{\rm nuc} = B^z_{{\rm nuc},L} - B^z_{{\rm nuc},R}$.

If we define a Bloch sphere for the qubit, whose poles on the z axis are the states $|\downarrow\uparrow\rangle$ and $|\uparrow\downarrow\rangle$, then the field $\Delta B^z_{\rm nuc}$ points along the z axis. The states $|S\rangle$ and $|T_0\rangle$ lie on the x axis of this Bloch sphere. The splitting induced by $\Delta B^z_{\rm nuc}$ leads to oscillations between the qubit states $|S\rangle$ and $|T_0\rangle$, with a frequency proportional to $|\Delta B^z_{\rm nuc}|$. Such oscillations are polluted, however, by two sources of randomness. First, due to the fact that the nuclear state is random for an equilibrium nuclear spin bath, the magnitude of the initial nuclear field $|\Delta B^z_{\rm nuc}|$, and hence the initial frequency of oscillations, is unknown. Furthermore, due to internal dynamics of the bath itself, the nuclear fields $\mathbf{B}_{{\rm nuc},d}(t)$ evolve in time. The resulting "spectral diffusion" of the qubit oscillation frequency leads to dephasing of the qubit oscillations[34]. Using an electron-spin-echo pulse, as explained below in Sec. III, dephasing due to the unknown *mean value* of $\Delta B^z_{\rm nuc}(t)$ over some interval can be reversed. However, decoherence due to fluctuations of $\Delta B^z_{\rm nuc}(t)$ on a timescale comparable to or shorter than the period between echo pulses in general cannot be eliminated in this way.

For experiments involving weaker external magnetic fields and long enough evolution times, it is necessary to go beyond the leading order in $\frac{|\mathbf{B}_{{\rm nuc},d}|}{B_{\rm ext}}$. Here we find that the transverse components of the Overhauser field, $B^{x,y}_{{\rm nuc},d}(t)$, crucially affect the qubit evolution in two primary ways. First, these transverse field components contribute to qubit decoherence by causing leakage of the electron spin state into the "non-qubit" subspace spanned by the states $|\downarrow\downarrow\rangle$ and $|\uparrow\uparrow\rangle$. Second, the magnitude of the transverse part of the Overhauser field introduces a correction to the frequency of the $|S\rangle - |T_0\rangle$ oscillations described above. Note that while in general a spin-echo pulse cannot reverse dephasing due to time-dependent fluctuations in the local fields, a partial or full recovery is possible if these fields vary *periodically* in time. Such a periodic time dependence, produced by the relative Larmor precession of different nuclear species, is the origin of the collapse and revival phenomenon.

An added complication in this moderate field regime

arises from the fact that, when we treat the Overhauser fields more properly as quantum mechanical operators, $B^x_{\mathrm{nuc},d}(t)$, $B^y_{\mathrm{nuc},d}(t)$ and $B^z_{\mathrm{nuc},d}(t)$ do not commute. Consequently, at this order, the semi-classical approach to electron spin dephasing requires more justification. As we discuss below, such an approach is valid when the number of nuclear spins is large, and when the interaction between the electronic and nuclear spins is weak.

III. THE SPIN-ECHO SEQUENCE

With the above-described picture in mind, below we focus on the Hahn echo experiment in GaAs double quantum dots (see e.g. Ref. 18), where each electron spin interacts weakly with $N \approx 10^6$ nuclear spins. In such experiments, the two-electron state in the double quantum dot is initialized to the singlet ground state at large potential detuning, where both electrons reside in the right dot, $|(0,2)S\rangle$. Here the numbers in parentheses indicate the electron occupation numbers in the left and right dots, respectively, and the letter S indicates the two-electron (singlet) spin state. By rapidly tuning the potentials of nearby electrostatic gates, one of the electrons is transferred to the left dot within a time scale of approximately 1 ns. After this operation, the two separated electron spins evolve freely for a time $\frac{\tau}{2}$ under the influence of the net local fields $\mathbf{B}_{\mathrm{tot},d}$, given in Eq. (2). The gate potentials are then rapidly tuned to bring the electrons closer together. Here a substantial exchange energy splitting between $|S\rangle$ and $|T_0\rangle$ is maintained for a time corresponding to a "π-phase" duration, which effectively leads to the the swapping of the states $|\downarrow\uparrow\rangle \leftrightarrow |\uparrow\downarrow\rangle$. Then the gate voltages are rapidly tuned to separate the electrons, and the system is allowed to evolve over another interval of length $\frac{\tau}{2}$.

At the end of the cycle, a spin readout procedure is performed. The gates are rapidly tuned to a large positive potential bias, where the singlet ground state takes the "(0,2)" orbital configuration, while the orbital part of the triplet state remains of the "(1,1)" type due to Pauli exclusion. The charge configuration is then measured via a nearby charge sensitive detector. Due to the correlation between the orbital and spin degrees of freedom, the final spin state of the two electrons can be inferred from this charge measurement. Ignoring any imperfections of the measurement itself, we assume that the average of the charge detector signal taken over many runs depends linearly on the singlet return probability.

The detector signal is averaged over a timescale which is long compared with all correlation times of the nuclear spin bath. Therefore we equate the averaged singlet return probability for evolution duration τ, $P_S(\tau)$, with the average of single-run singlet return probabilities, taken over the equilibrium ensemble of initial nuclear spin configurations. Note that because the qubit is initialized in the singlet state, $P_S(\tau)$ approaches 1 for very short evolution times τ. For very long times, when coherence is lost and the qubit tends to an equal-probability classical mixture of the states $|S\rangle$ and $|T_0\rangle$, $P_S(\tau)$ approaches 1/2. Therefore, by convention, we define the echo signal as $2P_S(\tau) - 1$, which takes the value 1 for $P_S(\tau) = 1$ and 0 for $P_S(\tau) = 1/2$. In this sense, the echo signal is used as a measure of electron spin coherence.

In the recent experiment of Ref. 18, the echo signal was observed to decay monotonically on a timescale of approximately 30 μs in high external magnetic fields above 300 mT. At intermediate magnetic fields ($120 - 300$ mT) additional small fast oscillations were observed. At lower magnetic fields ($50 - 120$ mT) these oscillations evolved into a complete collapse of the echo signal at $\tau \approx 1\,\mu$s, followed by a pattern of revivals and collapses on a timescale $\tau \approx 10\,\mu$s. The collapse and revival pattern was attenuated by a decaying envelope over a timescale of $\tau \approx 30\,\mu$s.

Our theoretical analysis of the echo experiments rests on a separation of timescales in the dynamics of the system. First, if one omits the π-pulse from the experimental protocol, $P_S(\tau)$ for the "free induction decay" decays to the value of 1/2 on the timescale $T_2^* \approx 10$ ns[1,8]. This decay results from the uncertainty in the z component of the Overhauser field, $\Delta B^z_{\mathrm{nuc}}$, which varies from run to run. In contrast, in a spin-echo measurement, the influence of a random, static Overhauser field $\Delta B^z_{\mathrm{nuc}}$ on the final electron spin state is eliminated by the combination of the π pulse and the two equal-length free evolution periods. If the Overhauser field were truly static, the electron spins would return to the state $|S\rangle$ at the end of the evolution. In a perfect measurement, for such a static Overhauser field, one would then obtain $P_S(\tau) = 1$, or an echo signal of value 1. Due to the time dependence of $\mathbf{B}_{\mathrm{nuc},d}(t)$, however, the echo signal typically decays to zero on a timescale of tens of microseconds.

During the free evolution time while the electrons are separated, the system exhibits oscillations between $|S\rangle$ and $|T_0\rangle$. Our crucial finding is that, for $\frac{|\mathbf{B}_{\mathrm{nuc},d}|}{B_{\mathrm{ext}}} \ll 1$, the oscillations are well described in terms of the net accumulated phase determined by difference of magnitudes of the *total* effective fields on the two dots (we take $\hbar = 1$):

$$\Delta\Phi(t) = g^* \mu_B \int_0^t \Big[\, |\mathbf{B}_{\mathrm{tot},L}(t')| - |\mathbf{B}_{\mathrm{tot},R}(t')| \,\Big] dt'. \quad (3)$$

The magnitude of the total field is given by

$$|\mathbf{B}_{\mathrm{tot},d}(t)| = \sqrt{\Big(B_{\mathrm{ext}} + B^z_{\mathrm{nuc},d}\Big)^2 + |\mathbf{B}^{\perp}_{\mathrm{nuc},d}(t)|^2}, \quad (4)$$

where $|\mathbf{B}^{\perp}_{\mathrm{nuc},d}(t)|$ is the magnitude of the component of the Overhauser field in dot d which is perpendicular to the external magnetic field. For B_{ext} not too large, $|\mathbf{B}_{\mathrm{tot},d}(t)|$ includes a significant contribution from $|\mathbf{B}^{\perp}_{\mathrm{nuc},d}(t)|$. Note that the time dependence of $\mathbf{B}^{\perp}_{\mathrm{nuc},d}(t)$ is dominated by the relative Larmor precession of the three nuclear spins species, ^{69}Ga, ^{71}Ga and ^{75}As. Such relative precession leads to a time-dependent modulation of $|\mathbf{B}_{\mathrm{tot},d}|$ and causes a reduction of $P_S(\tau)$ on a timescale of microseconds. In addition, random fluctuations of

$B^z_{\mathrm{nuc},d}(t)$, which arise due to interactions between nuclear spins, lead to a reduction of $P_S(\tau)$ on the same timescale. To account for all the above-mentioned effects, which were observed in Ref. 18, we thus include a combination of deterministic (Larmor precession) and stochastic processes in the evolution of the Overhauser fields in our semi-classical approach.

IV. THE QUBIT HAMILTONIAN

We begin our quantitative investigation by constructing the quantum Hamiltonian that describes the spin echo experiment. Although the experiment consists of several temporal stages, we focus on the free evolution periods, during which the Overhauser fields exert their main influence on the qubit evolution. We consider all other stages of the experiment, i.e. the singlet state initialization at the first stage, the pi pulse, and the measurement of the final state, to be perfect.

During the free evolution period while the electrons are well-separated, the qubit state evolves according to the effective Zeeman Hamiltonian

$$H_{el} = g^* \mu_B \left(\mathbf{S}_L \cdot \mathbf{B}_{\mathrm{tot},L} + \mathbf{S}_R \cdot \mathbf{B}_{\mathrm{tot},R} \right), \tag{5}$$

presented above in Eq. (1). The total effective field in dot d, $\mathbf{B}_{\mathrm{tot},d}$, is formed by the vector sum of the uniform external field $\mathbf{B}_{\mathrm{ext}}$, and the Overhauser field

$$g^* \mu_B \mathbf{B}_{\mathrm{nuc},d} = \sum_n A_{d,n} \mathbf{I}_n, \quad A_{d,n} = \mathcal{A}_{\alpha(n)} |\psi_{d,n}|^2, \tag{6}$$

where n is a label which indexes all of the nuclei. The parameter $\mathcal{A}_{\alpha(n)}$ is the microscopic hyperfine coupling for nuclear spin species $\alpha(n)$, while the factor $|\psi_{d,n}|^2$ weights the hyperfine coupling to nuclear spin n in dot d by the local electron density, and satisfies the normalization condition $\sum_n |\psi_{d,n}|^2 = n_c$, where $n_c = 2$ is the number of nuclei per unit cell of the GaAs lattice. In regions where the electron density is substantial, the coupling to nuclei goes as $|\psi_{d,n}|^2 \sim \frac{1}{N_d}$, where we define $N_d \equiv n_c^2 / \sum_n |\psi_{d,n}|^4$ as the effective number of nuclei in dot d. For typical GaAs quantum dots, $N_d \approx (1-4) \cdot 10^6$. The index $\alpha = \{1, 2, 3\}$ runs over the three nuclear species ^{69}Ga, ^{71}Ga, and ^{75}As.

To describe the evolution of the nuclear spin bath, we employ a Hamiltonian which includes Zeeman terms with a species- and site-dependent Larmor frequency for each nuclear spin, and the dipolar coupling between all pairs of nuclei:

$$H_{\mathrm{nuc}} = \sum_n \omega_n I^z_n + \sum_{n,n'} D^{ij}_{n,n'} I^i_n I^j_{n'}, \tag{7}$$

where n and n' run over all nuclei, and i and j label the Cartesian components of the nuclear spin operators.

We wish to identify the main sources of decoherence for the two electron spin qubit which arise from the combined evolution under the Hamiltonian in Eqs. (1) and (7). Naturally, we shall use $1/N_d$ as a small parameter. In addition, we note that the lowest external field used in the experiment in Ref. 18 (50 mT) was more than an order of magnitude bigger than the typical magnitude of the Overhauser field. Therefore we will proceed to study decoherence effects as an expansion in $\frac{|\mathbf{B}_{\mathrm{nuc},d}|}{B_{\mathrm{ext}}}$, and to leading order in $1/N_d$.

First, note that if one replaces the operators $\mathbf{B}_{\mathrm{nuc},L(R)}$ in Eq. (2) by classical vectors with magnitudes much smaller than B_{ext}, then, similar to the case when the Overhauser field was absent, the system described by Hamiltonian (1) possesses a two-dimensional subspace which is energetically well-separated from the remaining two levels. This new qubit subspace is spanned by the states $|\uparrow_{\hat{n}_L}\rangle \otimes |\downarrow_{\hat{n}_R}\rangle$ and $|\downarrow_{\hat{n}_L}\rangle \otimes |\uparrow_{\hat{n}_R}\rangle$, with eigenvalues $\pm\frac{1}{2}\mu_B g^* \left(|\mathbf{B}_{\mathrm{tot},L}| - |\mathbf{B}_{\mathrm{tot},R}|\right)$. Here the up and down arrows indicate the projections of the electron spins onto the quantization axes $\hat{n}_L$ and $\hat{n}_R$ parallel to the total fields $\mathbf{B}_{\mathrm{tot},L(R)}$ in each dot.

Deviations of the directions of $\hat{n}_L$ and $\hat{n}_R$ from the z-axis arise from the Overhauser field components perpendicular to the applied field, $\mathbf{B}^{\perp}_{\mathrm{nuc},d}$. Due to evolution of the nuclear spin bath (primarily due to Larmor precession of the nuclear spins), the fields $\mathbf{B}^{\perp}_{\mathrm{nuc},d}(t)$, and hence $\hat{n}_L$ and $\hat{n}_R$, are slowly modulated in time. However, because the frequencies of such modulations are typically two orders of magnitude smaller than the value of the energy gap between the two instantaneous eigenstates, $\Delta E = \mu_B g^* \left(|\mathbf{B}_{\mathrm{tot},L}| - |\mathbf{B}_{\mathrm{tot},R}|\right)$, we assume that each electron spin adiabatically follows its local, slowly varying, quantization axis $\hat{n}_L$ or $\hat{n}_R$. For small $\frac{|\mathbf{B}_{\mathrm{nuc},d}|}{B_{\mathrm{ext}}}$, the main effect of the nuclear field is thus to modulate the magnitude of the total field $|\mathbf{B}_{\mathrm{tot},d}(t)|$, Eq. (4), and hence to modify the dynamical phase $\int \Delta E(t) dt$ accumulated between the two eigenstates over the free evolution period. We therefore ignore changes in the directions of the quantization axes in each dot, and describe the evolution of the system by using the effective Hamiltonian

$$H_{el,z} = g^* \mu_B \left(S^z_L \left|B_{\mathrm{tot},L}\right| + S^z_R \left|B_{\mathrm{tot},R}\right| \right). \tag{8}$$

For given classical values of $\mathbf{B}_{\mathrm{tot},L}$ and $\mathbf{B}_{\mathrm{tot},R}$, this Hamiltonian preserves the instantaneous eigenvalues of the original Hamiltonian H_{el}, Eq. (1). Expanding $|\mathbf{B}_{\mathrm{tot},d}|$ in Eq. (4) in the small parameter $\frac{|\mathbf{B}_{\mathrm{nuc},d}|}{|B_{\mathrm{ext}}|}$, the Hamiltonian $H_{el,z}$ becomes

$$H_{el,z} \approx g^* \mu_B \sum_{d=L,R} \left(B^z_{\mathrm{nuc},d} + \frac{|\mathbf{B}^{\perp}_{\mathrm{nuc},d}|^2}{2|B_{\mathrm{ext}}|} \right) S^z_d. \tag{9}$$

The term proportional to $B^z_{\mathrm{nuc},d}$ is the zeroth-order contribution in $\frac{|\mathbf{B}_{\mathrm{nuc},d}|}{|B_{\mathrm{ext}}|}$, and may give rise to a magnetic-field-independent contribution to the electron spin decoherence. The term proportional to $\frac{|\mathbf{B}^{\perp}_{\mathrm{nuc},d}|^2}{|B_{\mathrm{ext}}|}$ is the first order correction in $\frac{|\mathbf{B}_{\mathrm{nuc},d}|}{|B_{\mathrm{ext}}|}$, and is responsible for the interesting collapse-and-revival behavior that we study below.

It should be noted that in writing Eqs. (8) and (9), we have ignored effects arising from the relative angle $\theta \sim \frac{|\mathbf{B}_{\mathrm{nuc},d}|}{B_{\mathrm{ext}}}$ between the quantization axes in the two dots, which enter at order $\left[\frac{|\mathbf{B}_{\mathrm{nuc},d}|}{B_{\mathrm{ext}}}\right]^2$. First, the misalignment of axes may cause unwanted transitions to the $|T_\pm\rangle$ states when initializing from the singlet state, or during free evolution under Hamiltonian H_{el}. However, these effects lead to a reduction of P_S on the order of θ^2. Second, we neglect any possible geometric phases which may accompany the dynamical phase accumulated while the electron spins adiabatically follow the local quantization axes in their separate dots. Such phases are proportional to the areas of closed loops swept out by $\hat{n}_L$ and $\hat{n}_R$ during their evolution, and for small θ are also proportional to θ^2.

Although we obtained Eq. (9) by treating the nuclear spin operators as classical vectors in a Taylor series approximation to Eq. (8), an identical expression for $H_{el,z}$ was formally derived in Ref. 32 from the full quantum Hamiltonian, Eq. (1), using a Schrieffer-Wolff transformation. The classical argument above thus provides a simple intuitive explanation for the formal perturbative derivation. Hereafter, unless otherwise specified, we treat the fields in Eq. (9) as quantum operators with appropriate commutation relations.

To complete the description of the problem, we now discuss the π-pulses employed in the Hahn echo sequence. These pulses are achieved by applying a time-dependent perturbation $H_\pi(t)$, which adds to the system's total Hamiltonian, $H = H_{el,z} + H_{\mathrm{nuc}} + H_\pi(t)$. We assume $H_\pi(t)$ is only nonzero over narrow intervals which are short compared to all timescales relevant for evolution under $H_{el,z}$ and H_{nuc}. Rather than specifying a detailed time-dependent protocol for $H_\pi(t)$, we define H_π implicitly in terms of its effect on the electron spin operators S_d^z:

$$\tilde{\mathcal{T}}\left[e^{i\int_0^t H_\pi(t')dt'}\right] S_d^z \, \mathcal{T}\left[e^{-i\int_0^t H_\pi(t'')dt''}\right] = c(t) S_d^z, \quad (10)$$

where $\mathcal{T}$ ($\tilde{\mathcal{T}}$) is the (reversed) time-ordering operator. In writing Eq. (10), we assume that $H_\pi(t)$ acts only within the two-dimensional qubit subspace. We shall consider perfect π-pulses, for which the "echo function" $c(t)$ switches between 1 and -1 over the short duration of the pulse. For simplicity we consider the pulses to be instantaneous, and for the Hahn echo sequence write

$$c(t) = \Theta\left(\tau/2 - t\right) - \Theta\left(t - \tau/2\right). \quad (11)$$

Using Eqs. (10) and (11) we switch to an interaction picture with respect to $H_\pi(t)$, where

$$S_d^z(t) \equiv c(t) S_d^z, \quad (12)$$

and where we employ the notation $S_d^z \equiv S_d^z\,(t=0)$. The interaction-picture time-dependent Hamiltonian $H_{el,z}(t)$ becomes

$$H_{el,z}(t) \approx g^* \mu_B \sum_{d=L,R} \left(B^z_{\mathrm{nuc},d} + \frac{|\mathbf{B}^\perp_{\mathrm{nuc},d}|^2}{2|B_{\mathrm{ext}}|} \right) c(t) S_d^z. \quad (13)$$

Equation (13), with Eq. (7), will serve as the starting point for our analysis of decoherence in the spin-echo experiment.

V. SEMI-CLASSICAL MODEL FOR THE REVIVALS

We now show that a simple semi-classical approach in which we treat the Overhauser field operators as classical vectors can reproduce the electron spin coherence collapse and revival effect predicted in Ref. 32 and observed in Ref. 18. In the next section, we will provide a systematic derivation and justification of this approach, starting from the full quantum description.

In this section, we treat the Overhauser field in each dot d in Eq. (13) as a sum of three classical vectors, $\mathbf{B}_{\mathrm{nuc},d}(t) = \sum_{\alpha=1}^{3} \mathbf{B}_{\alpha,d}(t)$, where α indexes the three nuclear spin species. We assume that the magnitudes of the species-dependent fields $\{|\mathbf{B}_{\alpha,d}|\}$, and their z components $\{B^z_{\alpha,d}\}$, are random but constant throughout the evolution. The time-dependence of $\mathbf{B}_{\mathrm{nuc},d}(t)$ within each run arises solely from the Larmor precession of the transverse nuclear spin components. Explicitly, we neglect the nuclear dipole-dipole interaction, and the influence of the Knight shift on the nuclear Larmor precession. We assume that all nuclei of the same species precess at a single Larmor angular velocity, $\omega_\alpha = \gamma_\alpha B_{\mathrm{ext}}$.

The echo signal $P_S(\tau)$ is obtained by averaging over many experimental runs. Thus we must average the results of electron spin dynamics against the distribution of initial states of the nuclear spins. Due to the large number of nuclear spins, $N \approx 10^6$, the initial values of the components of each vector $\mathbf{B}_{\alpha,d}(t=0)$ are Gaussian distributed with zero mean and a standard deviation $\bar{b}_{\alpha,d}$ of order 1 mT (see calculations below).

The model in Eq. (13), under the assumptions above, is sufficient to produce the collapse and revival effect in $P_S(\tau)$, and further provides an intuitive semi-classical picture in which to understand the phenomenon. However, because we neglect the time dependence of $B^z_{\mathrm{nuc},d}(t)$, and the effects of the Knight shift and other dephasing mechanisms of the nuclear Larmor precession, this model does not capture the decaying envelope observed in the experiment of Ref. 18. These issues will be addressed in detail in Sec.VI.

We now calculate $P_S(\tau)$, using the singlet initial state $|\psi(t=0)\rangle = |S\rangle = \frac{1}{\sqrt{2}}\left(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle\right)$. For any given set of initial values of the (18 total) components of the six classical vectors $\{\mathbf{B}_{\alpha,d}(t=0)\}$, the Hamiltonian in Eq. (13) generates a pure quantum evolution which after an evo-

lution time $t = \tau$ yields

$$|\psi(\tau)\rangle = \frac{e^{-i\Delta\Phi(\tau)/2}}{\sqrt{2}}\left(|\uparrow\downarrow\rangle - e^{i\Delta\Phi(\tau)}|\downarrow\uparrow\rangle\right). \quad (14)$$

The relative phase $\Delta\Phi(\tau) = \Phi_L - \Phi_R$ is related to the difference of the magnitudes of the total effective fields, $|\mathbf{B}_{\mathrm{tot},d}(t)|$, in the two dots. Within the approximation of $|\mathbf{B}_{\mathrm{tot},d}(t)|$ used to write Eq.(13), we obtain:

$$\Phi_d(\tau) = \frac{g^*\mu_B}{2|B_{\mathrm{ext}}|}\int_0^\tau |\mathbf{B}^\perp_{\mathrm{nuc},d}(t)|^2 c(t)\,dt. \quad (15)$$

Note that the phase Φ_d is determined solely by the dynamics of a single isolated electron in dot d. Below we will use this fact to relate the decoherence of the two-electron singlet-triplet qubit to that of a single electron spin in a quantum dot.

For the final state $|\psi(\tau)\rangle$ in Eq. (14), the singlet return probability is given by

$$|\langle\, S\,|\,\psi(\tau)\,\rangle|^2 = \frac{1}{2} + \frac{1}{2}\cos(\Delta\Phi). \quad (16)$$

The ensemble-averaged singlet return probability $P_S(\tau)$ is found by averaging the result for a single run, Eq. (16), with respect to the distribution of initial magnitudes and directions of the six vectors $\{\mathbf{B}_{\alpha,d}(t=0)\}$. Note that because $\cos(\Delta\Phi) = \mathrm{Re}\left[e^{i\Phi_R}e^{-i\Phi_L}\right]$, and because the Overhauser field configurations in the two dots are independent, we can average over $e^{i\Phi_L}$ and $e^{i\Phi_R}$ independently,

$$P_s = \frac{1}{2} + \frac{1}{2}\mathrm{Re}\Big[\langle e^{i\Phi_R}\rangle\langle e^{-i\Phi_L}\rangle\Big]. \quad (17)$$

To perform the averaging, we calculate $\Phi_d(\tau)$ using Eq. (15), with $c(t)$ given by Eq. (11), and with the classical evolution for $\mathbf{B}^\perp_{\mathrm{nuc},d}(t)$ resulting from the free precession of the underlying nuclear spins. We write $|\mathbf{B}^\perp_{\mathrm{nuc},d}(t)|^2 = \sum_{\alpha,\beta} b_{\alpha,d}(t)b^*_{\beta,d}(t)$ in terms of the complex-valued fields $b_{\alpha,d} = B^x_{\alpha,d} + iB^y_{\alpha,d}$. In this decomposition, the time evolution of the Overhauser field due to Larmor precession is given by $b_{\alpha,d}(t) = b_{\alpha,d}(0)e^{i\omega_\alpha t}$. Each phase Φ_d is then given by

$$\begin{aligned}\Phi_d &= \frac{g^*\mu_B}{2|B_{\mathrm{ext}}|}\int_0^\tau c(t)\sum_{\alpha,\beta} b_{\alpha,d}(t)b^*_{\beta,d}(t)\,dt \quad (18)\\ &= \frac{2g^*\mu_B}{|B_{\mathrm{ext}}|}\sum_{\alpha,\beta} b_{\alpha,d}(0)b^*_{\beta,d}(0)\frac{e^{i\omega_{\alpha\beta}\tau/2}}{i\omega_{\alpha\beta}}\sin^2\left(\omega_{\alpha\beta}\tau/4\right),\end{aligned}$$

where $\omega_{\alpha\beta} = \omega_\alpha - \omega_\beta$.

Next we must integrate over all initial conditions, i.e. over the initial magnitudes and phases of the three fields $\{b_{\alpha,d}(0)\}$ in each dot d. For this purpose we express the initial conditions as $b_{\alpha,d}(0) = \overline{b}_{\alpha,d}z_\alpha$, where each $z_\alpha = x_\alpha + iy_\alpha$ is a dimensionless complex variable. The quantity $\overline{b}_{\alpha,d}$ is the root-mean-squared (rms) value of each component of the transverse field for species α in dot d,

$$g^*\mu_B\overline{b}_{\alpha,d} = \sqrt{a_\alpha\bar{n}_\alpha/N_d}\mathcal{A}_\alpha, \quad (19)$$

with $a_\alpha = \frac{2}{3}(I_\alpha+1)I_\alpha = \frac{5}{2}$. Here $\bar{n}_\alpha$ is the average number of nuclei of species α, per unit cell (see Ref. 32). This substitution gives

$$\Phi_d = \sum_{\alpha,\beta} T_{\alpha\beta,d}\frac{z_\alpha z^*_\beta}{2}, \quad (20)$$

with

$$T_{\alpha\beta,d} = -\frac{4g^*\mu_B\overline{b}_{\alpha,d}\overline{b}_{\beta,d}}{|B_{\mathrm{ext}}|}\frac{e^{i\omega_{\alpha\beta}\tau/2}}{i\omega_{\alpha\beta}}\sin^2\left(\omega_{\alpha\beta}\tau/4\right). \quad (21)$$

We now carry out an ensemble average over the initial conditions by treating all components of the $\{z_\alpha\}$ as independent Gaussian-distributed random variables with zero mean and unit variance, i.e. with probability density function $p(\{z_{\alpha'}\})\prod_{\alpha=1}^3 dx_\alpha dy_\alpha = \frac{1}{(2\pi)^3}\prod_{\alpha=1}^3\exp(-\frac{|z_\alpha|^2}{2})dx_\alpha dy_\alpha$:

$$\begin{aligned}\langle e^{-i\Phi_d}\rangle &= \int\prod_\alpha dx_\alpha dy_\alpha\, p(\{z_{\alpha'}\})\, e^{-\frac{i}{2}\sum_{\beta,\beta'}T_{\beta\beta',d}z^*_\beta z_{\beta'}}\\ &= \prod_\alpha\frac{1}{1+i\lambda_{\alpha,d}}. \quad (22)\end{aligned}$$

Here the parameters $\{\lambda_{\alpha,d}\}$ are the eigenvalues of the T-matrix for dot d.

The 3×3 Hermitian matrix T in Eq. (21) corresponds to that of Ref. 32. Because T is Hermitian and is similar to an antisymmetric matrix, it has one zero eigenvalue, $\lambda_{1,d} = 0$, and a pair of eigenvalues $\lambda_{2(3),d} = \pm\sqrt{\sum_{\alpha>\beta}|T_{\alpha\beta,d}|^2}$. Inserting these eigenvalues into Eq. (22), and using Eq. (21) for $T_{\alpha\beta,d}$, we obtain:

$$\langle e^{-i\Phi_d}\rangle = \left[1 + \sum_{\alpha>\beta}\left(\frac{4g^*\mu_B\overline{b}_{\alpha,d}\overline{b}_{\beta,d}}{|B_{\mathrm{ext}}|}\right)^2\frac{\sin^4\left(\omega_{\alpha\beta}\tau/4\right)}{\omega^2_{\alpha\beta}}\right]^{-1}. \quad (23)$$

Thus we see that the semi-classical model used in this section reproduces the result of Ref. 32 for the decay of the spin echo signal in a single quantum dot. The echo signal shows oscillations with amplitude $\left(\frac{4g^*\mu_B\overline{b}_{\alpha,d}\overline{b}_{\beta,d}}{|B_{\mathrm{ext}}|\omega_{\alpha\beta}}\right)^2$, which develop into the complete collapses and revivals at low magnetic fields. Note that the expression for the phase in Eq. (18) includes terms which are bilinear combinations of Gaussian variables, and so are not Gaussian themselves. Therefore the decay in the interval $\tau \ll 1/\omega_{\alpha\beta}$ behaves like an inverse polynomial, rather than the form $e^{-\mathrm{const}\cdot\tau^4}$ expected for spectral diffusion (see below and Refs. 35–37).

We now return to computing the echo signal in the double dot system, Eq. (17). First, note that $\langle e^{-i\Phi_d}\rangle$ in Eq. (23) is strictly real. This fact is a consequence of

antisymmetry of the echo function around the time $\tau/2$. Thus we can drop the "Re" from Eq. (17) and write the echo signal as

$$2P_S - 1 = \langle e^{-i\Phi_L}\rangle\langle e^{-i\Phi_R}\rangle. \tag{24}$$

The expressions in Eqs. (22) and (23), which describe the dephasing of a single electron spin in an isolated quantum dot, were derived previously from a fully quantum mechanical treatment in Ref. 32. A key element of the derivation in that work was the vanishing of the contribution of the commutator $[I_k^+, I_l^-]$ between nuclear spin operators in the low order perturbation expansion of the evolution operator. The vanishing commutator is indicative of classical behavior, and further motivates our classical treatment of the nuclear evolution.

To conclude this section, we show that the classical treatment provides an intuitive explanation for the collapses and revivals. The total effective electron Zeeman field in each dot d, $|\mathbf{B}_{\rm ext} + \mathbf{B}_{{\rm nuc},d}|$, depends on the square of the transverse Overhauser field, $|\mathbf{B}^{\perp}_{{\rm nuc},d}|^2$. The terms in the expansion $|\mathbf{B}^{\perp}_{{\rm nuc},d}|^2 = \sum_{\alpha\alpha'}\left(B^x_{\alpha,d}B^x_{\alpha',d} + B^y_{\alpha,d}B^y_{\alpha',d}\right)$ which involve nuclear spins of two different species, $\alpha \neq \alpha'$, oscillate at the relative Larmor angular velocity $\omega_\alpha - \omega_{\alpha'}$. As a result, the magnitude of the total field, $|\mathbf{B}_{{\rm tot},d}|$, and hence the splitting between electron spin energy levels, oscillates as a function of time. These oscillations determine the phase accumulation during each run of the experiment. In this simple semi-classical treatment the nuclear spin evolution is not affected by the electron spin, so that the phase between electron spin components remains well-defined during each run. However, averaging over the ensemble of nuclear spin initial states amounts to averaging over the phases and amplitudes of these oscillations, and causes the collapse of the echo signal. However, if the free evolution time $\tau/2$ is simultaneously an integer multiple of each of the three relative Larmor periods, then the contribution of those oscillations vanishes independently of the initial nuclear state, and a revival of electron spin coherence is observed. Note that it is a fortunate coincidence in GaAs that the Larmor frequencies of the three species are nearly equidistant, which, to a good approximation, allows the commensurability condition to be easily fulfilled simultaneously for all three pairs of nuclear species.

VI. DERIVATION OF THE SEMI-CLASSICAL APPROACH

In this section we present a systematic derivation of the semi-classical treatment of electron spin dynamics presented above. Keeping in mind the discussion surrounding Eq. (9), we now restore the quantum nature of the electron spin operators and begin with the interaction-picture quantum Hamiltonian $H = H_{el,z}(t) + H_{\rm nuc}$, Eq. (13). We also now reintroduce the nuclear dipolar interaction, see Eq. (7), and allow for local variations of the nuclear Larmor frequencies, which were omitted from the simple model in section V. At each step in the derivation, we will justify the approximations needed to arrive at the semi-classical model with arguments from first principles, or with the help of experimental data. Along the way, we will also incorporate several important features of the dynamics, such as the time dependence of $B^z_{\rm nuc}(t)$, back-action of the electron spin evolution on the nuclear state, and nuclear spin dephasing, which were ignored in the heuristic treatment in the previous section.

A. Quantum expression for the echo signal

Our aim is to derive an expression for the echo signal at the end of the full Hahn echo sequence, via the calculation of the singlet return probability

$$P_S(\tau) = \frac{1}{Z_\infty}{\rm Tr}_{\rm nuc}\Big[\,\langle S|\,U^\dagger(\tau)\,|S\rangle\,\langle S|\,U(\tau)\,|S\rangle\,\Big], \tag{25}$$

where $U(\tau) = \mathcal{T}e^{-i\int_0^\tau (H_{el,z}(t)+H_{\rm nuc})dt}$ is the interaction picture evolution operator of the joint electron-nuclear spin system, with $\mathcal{T}$ representing the time-ordering operator. Here we have assumed that the electron spins are initialized to the singlet state $|S\rangle$. The trace in Eq. (25) is taken over all nuclear spin states, with $Z_\infty = {\rm Tr}_{\rm nuc}[\hat{1}]$ representing the partition function for an infinite temperature (completely random) nuclear spin state.

To separate the time evolution due to the static part of $B^z_{\rm nuc}$ (which dominates the evolution for $t < \tau$, but has no influence on $U(t = \tau)$ due to the spin-echo), we introduce the zero-order Hamiltonian

$$H_0(t) = g^*\mu_B \sum_{d=L,R} B^z_{{\rm nuc},d}\, c(t) S^z_d + \sum_k \omega_k I^z_k, \tag{26}$$

and a corresponding zero-order evolution operator $U_0(t) = e^{-i\int_0^t H_0(t')dt'}$. Note that due to antisymmetry of the echo function, $\int_0^\tau c(t')dt' = 0$, the evolution operator $U_0(t = \tau)$ at the end of the full sequence does not depend on the electron spin operators and simply rotates all nuclear spins about the z-axis.

The full evolution operator $U(\tau)$ can be rewritten as

$$U(\tau) = U_0(\tau)\cdot\mathcal{T}e^{-i\int_0^\tau dt\sum_d[H_{\perp,d}(t)S^z_d c(t) + H_{D,d}(t)]}, \tag{27}$$

where the time-dependent operators in the exponent are given by $H_{\perp,d}(t) = U_0^\dagger(t)\left[g^*\mu_B\frac{|\mathbf{B}^{\perp}_{{\rm nuc},d}|^2}{2|B_{\rm ext}|}\right]U_0(t)$ and $H_{D,d}(t) = U_0^\dagger(t)\Big[\sum_{n,n'\in d} D^{ij}_{n,n'} I^i_n I^j_{n'}\Big]U_0(t)$, see Eq. (7). These terms describe the evolution of the Overhauser fields due to Larmor precession and the dipole-dipole interaction between nuclear spins in dot d, respectively. We assume that the two dots are well separated, such that the inter-dot dipolar coupling can be neglected. This

approximation is not essential for the derivation ahead, however, and the existence of a small inter-dot coupling would not significantly affect the final result.

In order to evaluate Eq. (25), we decompose the evolution operator $U(\tau)$ into four separate pieces. Because operators describing spins in different dots commute, the exponentials in Eq. (27) can be factored by dot index d. Therefore the evolution operator can be written as $U(\tau) = U_L(\tau)U_R(\tau)$, where $U_{L(R)}(\tau)$ only involves spin operators in the left (right) dot. In addition, because the z-projection of electron spin in each dot is conserved by $U(\tau)$, see Eq. (27), we can separate the evolution by introducing projectors onto the product states $|\uparrow\downarrow\rangle$ and $|\downarrow\uparrow\rangle$:

$$U(\tau) = U_{L+}(\tau)U_{R-}(\tau)\,|\uparrow\downarrow\rangle\langle\uparrow\downarrow| + U_{L-}(\tau)U_{R+}(\tau)\,|\downarrow\uparrow\rangle\langle\downarrow\uparrow|\,, \tag{28}$$

where $U_{d\sigma}$ is a unitary operator acting only on the nuclear spins in dot d, with the electron spin taken to be in the S_d^z eigenstate $|\uparrow\rangle$ for $\sigma = +$, or $|\downarrow\rangle$ for $\sigma = -$. Inserting Eq. (28) into Eq. (25), and performing the trace over nuclear spin states separately for the two dots, we find that the echo signal is given by [*c.f.* Eq. (24)]

$$2P_S - 1 = \mathrm{Re}\,(C_L \cdot C_R)\,, \tag{29}$$

where

$$C_d = \frac{1}{Z_{\infty,d}}\mathrm{Tr}_{\mathrm{nuc}}\left[U_{d+}^\dagger(\tau)U_{d-}(\tau)\right],\ d = L, R \tag{30}$$

The echo signal, Eq. (29), is a product of two similar averages which are taken over the different sets of nuclear spins in the two dots. In the remainder of this section we focus on the behavior of the average within a single dot, which we denote as $C = \frac{1}{Z_\infty}\mathrm{Tr}_{\mathrm{nuc}}\left[U_+^\dagger(\tau)U_-(\tau)\right]$. This allows us to simplify all formulas by suppressing the dot index d. At the end of subsection VI C we will return to the two-electron double-dot echo signal, including the combined effects of dephasing in each of the two dots.

B. The semi-classical approximation of separating the dynamics of the spin diffusion and transverse Overhauser field

In this subsection we introduce a semi-classical treatment in which the evolution operators in Eq.(29) are factorized into contributions depending separately on $H_\perp(t)$ and $H_D(t)$. We start by using Eq. (27) and the fact that $U_0(\tau)$ involves only nuclear spin operators to write the evolution operator in C as

$$\begin{aligned} U_+^\dagger(\tau)U_-(\tau) &= \widetilde{\mathcal{T}}\left[e^{i\int_0^\tau dt\left[\frac{1}{2}H_\perp^+(t)c(t)+H_D^+(t)\right]}\right] \\ &\times \mathcal{T}\left[e^{-i\int_0^\tau dt'\left[-\frac{1}{2}H_\perp^-(t')c(t')+H_D^-(t')\right]}\right], \end{aligned} \tag{31}$$

where the superscripts + and − indicate projection onto the $|\uparrow\rangle$ and $|\downarrow\rangle$ electron spin states. The projection is needed because $H_\perp(t)$ and $H_D(t)$ depend on $U_0(t)$, which for $t \neq \tau$ depends on S^z.

Intuitively, the two terms involving $H_\perp(t)$ and $H_D(t)$ lead to suppression of the echo signal, each through a different physical mechanism. In the spirit of the semi-classical approximation in Sec. V, where the Overhauser field operators were treated as classical vector variables, $H_\perp(t)$ causes dephasing through the time-dependence of $|\mathbf{B}_{\mathrm{nuc}}^\perp(t)|$ generated by nuclear Larmor precession, while $H_D(t)$ causes decoherence through fluctuations of $B_{\mathrm{nuc}}^z(t)$ generated by dipolar-interaction-mediated spin diffusion.

The semi-classical approach relies on three main approximations, which involve neglecting various commutators of the form $[B_{\mathrm{nuc}}^i(t), B_{\mathrm{nuc}}^j(t')]$. When the number of nuclei N_d is large, these commutators scale as $|\mathbf{B}_{\mathrm{nuc}}(t)|/N_d$. Below we provide a physical motivation for each such approximation, and discuss the associated range of validity.

First, we neglect the commutator between $H_\perp(t)$ and $H_D(t')$, and factor the exponentials appearing in Eq. (31):

$$U_\sigma(\tau) \approx U_{\perp\sigma}(\tau)U_{D\sigma}(\tau), \tag{32}$$

with

$$\begin{aligned} U_{\perp\sigma}(\tau) &= \mathcal{T}e^{-\frac{i}{2}\int_0^\tau \sigma H_\perp^\sigma(t)c(t)dt}, \\ U_{D\sigma}(\tau) &= \mathcal{T}e^{-i\int_0^\tau H_D^\sigma(t)dt}. \end{aligned} \tag{33}$$

The commutator $[H_\perp(t), H_D(t')]$ which was set to zero in writing Eq. (32) leads to nuclear spin dephasing, i.e. decay of the correlator $\langle I_k^i(t)I_k^j(t')\rangle$, due to dipolar flip-flop events. In our model, we account for this effect phenomenologically by introducing random site-to-site small variations of the nuclear Larmor frequencies (see Sec. VI C). In doing so, we assume that intrinsic and extrinsic dephasing of nuclear spins affect the electron spin dynamics in the same way.

In addition, the commutators neglected above may lead to enhanced nuclear spin diffusion through the combination of dipole-dipole and hyperfine-mediated spin flips. The validity of this approximation thus depends on material and system parameters. However, it can be checked by comparison with experimental data, as discussed in more detail in section. VII.

The second approximation is to split the average of the product of operators involving $U_{D\sigma}$ and $U_{\perp\sigma}$ into a product of averages involving $U_{D\sigma}$ and $U_{\perp\sigma}$ separately:

$$\frac{1}{Z_\infty}\mathrm{Tr}_{\mathrm{nuc}}\left[U_{D+}^\dagger U_{\perp+}^\dagger U_{D-}U_{\perp-}\right] \approx \frac{1}{Z_\infty}\mathrm{Tr}_{\mathrm{nuc}}\left[U_{D+}^\dagger U_{D-}\right]\cdot\frac{1}{Z_\infty}\mathrm{Tr}_{\mathrm{nuc}}\left[U_{\perp+}^\dagger U_{\perp-}\right]. \tag{34}$$

The correlations which are neglected by splitting this average are related to correlations of the longitudinal and transverse components of the Overhauser fields, $\mathbf{B}_{\mathrm{nuc}}^z(t)$ and $\mathbf{B}_{\mathrm{nuc}}^\perp(t)$. These correlations are contributed by expectation values of at least four operators of the same nuclear spin n, e.g. $(I_n^x)^2(I_n^z)^2$. On the other hand, the leading contribution to the split-average comes from the expectation values of only two operators of the same nuclear spin, e.g. $(I_n^z)^2$, and therefore is roughly N_d times bigger, due to the fact that it contains a sum over at least N_d times as many terms.

C. The semi-classical approximation of averaging over all nuclear spin states

Through Eq. (34), the dephasing in a single dot can be approximately expressed as a product of two separate averages, one involving $U_{D\sigma}$ and the other involving $U_{\perp\sigma}$. The average over $U_{D\sigma}$ can be associated with decoherence due to spectral diffusion caused by dipole-dipole mediated nuclear spin diffusion. This process was analyzed in Refs. 35–37, and was shown to result in an echo decay factor $\exp(-(\tau/\tau_{SD})^4)$. Using this result and Eq. (34), we rewrite Eq. (30) as

$$C(\tau) \approx e^{-(\tau/\tau_{SD})^4}\frac{1}{Z_\infty}\mathrm{Tr}_{\mathrm{nuc}}\left[U_{\perp+}^\dagger(\tau)U_{\perp-}(\tau)\right], \tag{35}$$

The spectral diffusion time τ_{SD} depends on the details of the quantum dots, and weakly depends on the magnetic field strength. According to Refs. 35,36 τ_{SD} is of order 10 microseconds, which is comparable to the duration of the experiment. Thus, the decay factor is likely to be important for a detailed fit to the experiments, and will be dominant at high magnetic fields. However, as we show below, at low enough magnetic field the dephasing associated with $U_{\perp\sigma}$, i.e. with the evolution of the transverse components of the Overhauser fields, becomes dominant.

We now turn to calculate the the dephasing associated with $U_{\perp\sigma}$ semi-classically. Making use of the large number of nuclei involved, $N_d \gtrsim 10^6$, the trace in Eq. (35) can be cast into a form which closely resembles the classical average described in Sec. V. We first combine together large groups of nuclei with similar couplings to form a set of "giant" collective spins. Then, by evaluating the trace in a basis of coherent states with well defined orientations of these giant spins, we find that their induced Overhauser fields effectively act as classical variables like those introduced "by hand" in Sec. V. However, there are two important differences in this more refined treatment. First, whereas in Sec. V all nuclear spins within each species were forced to precess at a fixed Larmor frequency, we now account for the electron-spin-dependent shift (i.e. the Knight shift) of the nuclear precession rate which is inherent in Eqs. (9) and (26). Second, we now account for inhomogeneity in the system, both in the Knight field terms and in the Larmor frequencies of all nuclear spins. Site-to-site variations of the Larmor frequencies ω_n, see Eq. (7), phenomenologically account for nuclear spin dephasing due to, e.g. local quadrupole moments. We assume that the inhomogeneities are weak, such that the differences between the mean Larmor angular velocities of the three species, $\omega_\alpha = \gamma_\alpha B_{\mathrm{ext}}$, are much larger than the widths of the distributions for each one.

To account for inhomogeneities in the system, we divide the nuclei in each dot into K groups labeled by the index $k = 1,\ldots,K$, with each group k containing nuclei of the same species $\alpha(k)$. These $\tilde{N}_k$ are picked from the N_d nuclei in dot d such that all members of the group have nearly the same hyperfine coupling A_k and feel nearly the same phenomenological local shift in magnetic field, δB_k. The Larmor angular velocity for nuclear spins in group k is given by $\omega_{\alpha(k)}+\delta\omega_k+\frac{1}{2}\sigma c(t)A_k$ where $\delta\omega_k = \gamma_{\alpha(k)}\delta B_k$. Note that the sign of the Knight shift $\pm\frac{1}{2}A_k$ depends on the state of the electron spin in the dot at time t, and is therefore proportional to $\sigma c(t)$.

To calculate C, the single dot contribution to the echo signal, we need to choose a basis for the nuclear spin Hilbert space. For each group k, we form a collective spin from all of the nuclear spins in the group, and consider "giant spin" states of well-defined total angular momentum I_k and orientation $\hat{\mathbf{n}}_k = (\sin\theta_k\cos\varphi_k, \sin\theta_k\sin\varphi_k, \cos\theta_k)$: $|\mathbf{I}_k|^2|I_k,\hat{\mathbf{n}}_k\rangle = I_k(I_k+1)|I_k,\hat{\mathbf{n}}_k\rangle$, $(\hat{\mathbf{n}}_k\cdot\mathbf{I}_k)|I_k,\hat{\mathbf{n}}_k\rangle = I_k|I_k,\hat{\mathbf{n}}_k\rangle$, where θ_k and φ_k are the polar and azimuthal angles of giant spin k, respectively. A wave function in the Hilbert space of all nuclear spins is written as $|\Psi\rangle = \bigotimes_{k=1}^K |I_k,\hat{\mathbf{n}}_k\rangle$. The trace is then performed by summing over all possible assignments of the values $\{I_k\}$ with appropriate weights[38], and integrating over all possible directions $\{\hat{\mathbf{n}}_k\}$. The length I_k of each giant spin k can vary from 0 to $\frac{3}{2}\tilde{N}_k$, but the vast majority of giant spin states have lengths of order $I_k \sim \sqrt{\tilde{N}_k}$. The relative quantum uncertainty in the transverse components of the giant spin coherent states, $\Delta I_k^i/I_k^i$, scales as $1/\sqrt{I_k} = 1/\sqrt[4]{\tilde{N}_k}$. As we shell see below, for analyzing the behavior near the revivals peaks it is sufficient to divide the nuclei into about $K \sim 10$ groups, which leaves as many as $\tilde{N}_k \approx 10^5$ nuclei in each group.

In the coherent state basis, the semi-classical approximation converts the expectation values

$\langle\Psi|\, U^{\dagger}_{\perp d+}(\tau)U_{\perp d-}(\tau)\,|\Psi\rangle$, which arise in the evaluation of the trace in Eq. (35), into "classical" expressions

$$\langle\Psi|\, U^{\dagger}_{\perp +}(\tau)U_{\perp -}(\tau)\,|\Psi\rangle \simeq e^{-i\int_0^\tau \frac{g^*\mu_B}{4B_{\rm ext}}|\mathbf{B}^{\perp +}_{{\rm nuc},d}(t)|^2_{cl}c(t)dt}e^{-i\int_0^\tau \frac{g^*\mu_B}{4B_{\rm ext}}|\mathbf{B}^{\perp -}_{{\rm nuc},d}(t)|^2_{cl}c(t)dt}, \tag{36}$$

where $|\mathbf{B}^{\perp +}_{{\rm nuc},d}(t)|^2_{cl}$ is determined by the expectation values $\overline{I_k^{\sigma x(y)}(t)} = \langle\, I_k, \hat{\mathbf{n}}_k \,|\, I_k^{\sigma x(y)}(t)\,|\, I_k, \hat{\mathbf{n}}_k\,\rangle$ of the giant spin components:

$$\frac{g^*\mu_B}{4B_{\rm ext}}|\mathbf{B}^{\perp\sigma}_{{\rm nuc},d}(t)|^2_{cl} \equiv \frac{1}{4g^*\mu_B B_{\rm ext}}\sum_{k,k'} A_k A_{k'}\left(\overline{I_k^{\sigma x}(t)}\cdot\overline{I_{k'}^{\sigma x}(t)} + \overline{I_k^{\sigma y}(t)}\cdot\overline{I_{k'}^{\sigma y}(t)}\right).$$

The superscript σ indicates that the time dependence of the operators is determined by the evolution with respect to H_0, Eq. (26), which depends on the electron spin state σ.

The validity of the approximation, Eq. (36), is discussed in Sec. VI E below. After making the approximation, however, the trace reduces to an integration over all possible expectation values $\overline{I_k^{\sigma x(y)}(t)}$ of the components of the K giant spins. We perform the integrations using a similar method to that used in Section V. First we write the classical field $|\mathbf{B}^{\perp\sigma}_{\rm nuc}(t)|^2_{cl} = \sum_{k,l} b_k^\sigma(t) b_l^{\sigma *}(t)$ as a sum of products of complex variables, where $b_k^\sigma = A_k\left(\overline{I_k^{\sigma x}(t)} + i\overline{I_k^{\sigma y}(t)}\right)$. Each $b_k^\sigma(t)$ evolves according to $b_k^\sigma(t) = b_k(0)e^{i[\omega_{\alpha(k)}t + \delta\omega_k t + \sigma A_k\int_0^t c(t')dt']}$. Using the approximation in Eq. (36) to evaluate the trace in Eq. (35), we have

$$C\,(\tau) \approx C_{sc}\,(\tau) = e^{-(\tau/\tau_{SD})^4}\left\langle e^{i\Phi}\right\rangle, \tag{37}$$

where the angled brackets indicate a "classical" averaging over the complex random variables $b_k^\sigma(0)$, and the phase Φ is given by the integrals in the exponents in Eq. (36)

$$\begin{aligned}\Phi\,(\tau) &= \frac{g^*\mu_B}{4|B_{\rm ext}|}\sum_{\sigma=\pm 1}\int_0^\tau dt\sum_{k,l} b_k^\sigma(t) b_l^{\sigma *}(t)\\ &= \frac{g^*\mu_B}{4|B_{\rm ext}|}\sum_{k,l} b_k(0)b_l(0)\sum_{\sigma=\pm 1}\int_0^\tau dt\, c(t)\exp\left[i(\omega_{kl}+\delta\omega_{kl})t + i\sigma A_{kl}\int_0^t dt' c(t')\right],\end{aligned} \tag{38}$$

with $\omega_{kl} = \omega_{\alpha(k)} - \omega_{\alpha(l)}$, $\delta\omega_{kl} = \delta\omega_k - \delta\omega_l$, and $A_{kl} = A_k - A_l$.

Assuming that $\tilde{N}_k$ is large for every group k, we perform the average over all initial coherent nuclear spin states by writing $b_k(0) = \overline{b}_k z_k$ in terms of a collection of independent, complex Gaussian random variables $\{z_k\}$ with unit variance. Here $g^*\mu_B\overline{b}_k = \sqrt{\widetilde{N}_k a_{\alpha(k)}/2A_k}$ is the rms value of each component of the Overhauser field associated with group k in the dot, similar to the parameters $\{\overline{b}_\alpha\}$ appearing in Eq. (21). In this representation, Eq. (38) becomes

$$\Phi\,(\tau) = \sum_{k,l} T_{kl}\frac{z_k z_l^*}{2}, \tag{39}$$

with

$$\begin{aligned}T_{kl}\,(\tau) &= \frac{ig^*\mu_B\overline{b}_l\overline{b}_k(\omega_{kl}+\delta\omega_{kl})}{2|B_{\rm ext}|}\\ &\times\ 4\frac{\cos(A_{kl}\tau/2) - \cos\left[(\omega_{kl}+\delta\omega_{kl})\tau/2\right]}{(\omega_{kl}+\delta\omega_{kl})^2 - A_{kl}^2}.\end{aligned} \tag{40}$$

Performing the Gaussian average over $\{z_k\}$, we obtain

$$\begin{aligned}\langle e^{-i\Phi}\rangle &= \int\prod_{k'} dx_{k'}dy_{k'}p(|z_{k'}|^2)\exp\left(-i\sum_{k,l}T_{kl}\frac{z_k^* z_l}{2}\right)\\ &= \prod_m \frac{1}{1+i\lambda_m\,(\tau)},\end{aligned} \tag{41}$$

where the parameters $\{\lambda_m\}$ are the eigenvalues of the $M\times M$ Hermitian matrix with elements $\{T_{kl}\}$.

Finally, we calculate the spin-echo signal of the two-electron double quantum dot singlet-triplet qubit, incorporating the dephasing due to both two dots, Eq. (29), using Eqs. (37) and (41) for each dot $d = L, R$:

$$2P_s - 1 = e^{-(\tau/\tilde{\tau}_{SD})^4} \prod_{d=L,R} \left(\prod_m \frac{1}{1 + i\lambda_{m,d}} \right). \quad (42)$$

Here $\tilde{\tau}_{SD}$ is the effective spectral diffusion timescale for the two electron system, $\tilde{\tau}_{SD}^{-4} = \tau_{SD,L}^{-4} + \tau_{SD,R}^{-4}$. For non-identical dots, the eigenvalues $\lambda_{m,d}$ are generally different due to the differing number of nuclei N_d, and their associated distributions of coupling constants $\{A_k\}$ and $\{\omega_k\}$. All of these parameters affect the grouping of spins into "giant spins," and the matrix elements T_{lk}, calculated according to Eq. (40).

Equation (42) is the main result of the paper. This result shows that, within the semi-classical approach, the echo signal $2P_S - 1$ in the Hahn echo experiment can be understood in terms of a decay envelope arising from spectral diffusion, along with an additional factor which arises from the relative precession of different nuclear species comprising the transverse Overhauser field. At low magnetic fields, this precession term leads to the collapse and revival effect.

D. Stability of the revivals peaks against system inhomogeneities

In this subsection we investigate how modifications of nuclear precession induced by the small spatial variations of the Knight fields and nuclear Zeeman couplings affect the electron spin echo signal, Eq. (42). In particular, we focus on the stability of the revival peaks.

Note that a necessary condition for the revivals to appear is the clear separation of Larmor frequencies, such that $\omega_{kl} \gg \delta\omega_{kl}, A_{kl}$ for $\alpha(k) \neq \alpha(l)$. For $\delta\omega_{kl} = A_{kl} = 0$, i.e. for a homogeneous nuclear system with no intrinsic nuclear spin dephasing, Eq. (41) reduces to Eq. (23), where the right hand side exhibits revivals for values of the free evolution time τ satisfying $\omega_{\alpha\beta}\tau \approx 4\pi n$, for any integer n. In this case the overall decay of the spin echo signal results solely from the spectral diffusion factor $e^{-(\tau/\tilde{\tau}_{SD})^4}$ in Eq. (42).

Small but nonzero frequency differences $\delta\omega_{kl}$ and A_{kl} can give rise to nuclear spin dephasing and cause additional decay of the revival peak envelope as a function of τ. We now analyze this decay and discuss its physical origin. Near the revival peaks $\omega_{kl}\tau \approx 4\pi n$, and for early times τ satisfying $\delta\omega_{kl}\tau, A_{kl}\tau \ll 1$, Eq. (40) simplifies to

$$T_{kl}|_{\text{peak}} \approx \frac{ig^*\mu_B \overline{b}_l \overline{b}_k (\omega_{kl} + \delta\omega_{kl})}{4|B_{\text{ext}}|} \frac{(A_{kl}\tau)^2 - (\delta\omega_{kl}\tau)^2}{(\omega_{kl} + \delta\omega_{kl})^2 - A_{kl}^2}. \quad (43)$$

For homo-nuclear terms with $\omega_{kl} = 0$ (i.e. for groups k and l comprised of the same nuclear species), we find

$$T_{kl}|_{\text{peak}} \approx \frac{ig^*\mu_B \overline{b}_l \overline{b}_k \delta\omega_{kl}}{4|B_{\text{ext}}|}\tau^2, \quad \alpha(k) = \alpha(l), \quad (44)$$

while hetero-nuclear terms are given by

$$T_{kl}|_{\text{peak}} \approx \frac{ig^*\mu_B \overline{b}_l \overline{b}_k}{4|B_{\text{ext}}|} \frac{A_{kl}^2 - \delta\omega_{kl}^2}{\omega_{kl}}\tau^2, \ \alpha(k) \neq \alpha(l). \quad (45)$$

Note that the hetero-nuclear terms and suppressed by the small ratios of $\frac{\delta\omega_{kl}}{\omega_{kl}}$ or $\frac{A_{kl}}{\omega_{kl}}$. Hence, for $\delta\omega_{kl}\tau, A_{kl}\tau \ll 1$, near the revival peaks the T-matrix is approximately block-diagonal with respect to the three species. Up to second order in $\omega_{kl}\tau, A_{kl}\tau$, we find that each block has a single pair of nonzero eigenvalues given by

$$\lambda_\alpha|_{\text{peak}} = \pm \frac{g^*\mu_B a_\alpha n_\alpha \mathcal{A}_\alpha^2 \sqrt{\langle \delta\omega_\alpha^2 \rangle}}{4N_d|B_{\text{ext}}|}\tau^2, \quad (46)$$

where $\sqrt{\langle \delta\omega_\alpha^2 \rangle}$ is the rms spread of Larmor angular frequencies of species α. Thus, we obtain a simple expression for the echo envelope decay at the revival peaks:

$$\langle e^{i\Phi} \rangle|_{\text{peak}} = \prod_\alpha \left[1 + \left(\frac{g^*\mu_B a_\alpha n_\alpha \mathcal{A}_\alpha^2}{4N_d|B_{\text{ext}}|} \right)^2 \langle \delta\omega_\alpha^2 \rangle \tau^4 \right]^{-1}. \quad (47)$$

Physically, Eq. (47) describes decay of the revival envelope with timescale $\tau_\perp^{-4} = \left(\frac{g^*\mu_B a_\alpha n_\alpha \mathcal{A}_\alpha^2}{4N_d|B_{\text{ext}}|} \right)^2 \langle \delta\omega_\alpha^2 \rangle$, which arises primarily from the intra-species spread of the Larmor frequencies, $\sqrt{\langle \delta\omega_\alpha^2 \rangle}$. The effect of the off-diagonal matrix elements between different species is negligible. Additionally, the effect of the Knight field is also negligible, due to the fact that the Knight field reverses its sign halfway through the evolution when the electron spin is flipped by the π-pulse of the spin-echo sequence.

E. Estimate of quantum corrections to the semi-classical results

The semi-classical treatment is expected to be valid in the limit of a large number of participating nuclei, N_d. To better understand the validity of the approximation for finite N_d, in this section we provide a heuristic estimate for the deviation of the semi-classical expression for the single electron coherence function, C_{cs}, Eq. (37), from the quantum expression for C, given in Eq. (35). We define the quantum error as $C_{cs} - C$.

We are interested in particular in the quantum error associated with the semi-classical approximation to the dynamics induced by the hyperfine and Zeeman couplings. Therefore we ignore the nuclear spin-diffusion contribution to the decoherence, which is caused by the dipolar interaction. This is done by setting $\tau_{SD} = \infty$ in Eqs. (35) and (37). We analyze the scaling of the quantum error as N_d is increased. However, while changing N_d, we wish to

keep fixed the rms values of the Overhauser field components, which determine the timescale for the decoherence of the electrons spins. Given Eq. (6), the rms value of the Overhauser field for each species scales as $\mathcal{A}_\alpha/\sqrt{N_d}$, therefore we require $\mathcal{A}_\alpha \propto \sqrt{N_d}$.

For large N_d, the leading contribution to the quantum error comes from the fact that for a given initial nuclear spin state $|\Psi_i\rangle = \bigotimes_k |I_k, \hat{\mathbf{n}}_k\rangle$, the overlap of the final nuclear spin states for different initial electron spin states, $|\Psi_{f+}\rangle \equiv U_{\perp+}(\tau)|\Psi\rangle$ for $\sigma = +$ and $|\Psi_{f-}\rangle \equiv U_{\perp-}(\tau)|\Psi\rangle$ for $\sigma = -$, is not unity, $|\langle \Psi_{f+} | \Psi_{f-} \rangle| < 1$. In other words, for every initial coherent state, the final state includes quantum correlations between the electron and nuclear spins, which are not captured by the semi-classical treatment. Thus, the electron-state-dependent modification to the nuclear evolution contributes an additional suppression of C which is not accounted for in Eq. (36).

Up to leading order in the hyperfine coupling, the nuclear spin evolution is dominated by Larmor precession due to the combination of the external magnetic field and the component of the Knight field parallel to the external field axis, z. Both contributions are included in H_0 in Eq. (26). However, these two effects alone will result in a perfect overlap of the final states, $|\Psi_{f+}\rangle = |\Psi_{f-}\rangle$. This is because the Larmor precession about the external field is independent of the electron spin state, while the precession due to the Knight field is perfectly reversed halfway through the evolution due to the echo pulse which flips the electron spin. Thus both effects were fully accounted for in the semi-classical treatment above.

The quantum error results from the higher order terms in the hyperfine coupling, in particular from the transverse components of the Knight field, i.e. from the terms $S_d^+ I_n^- + S_d^- I_n^+$ in the system's Hamiltonian, Eq. (5). These terms contribute to $\sigma H_\perp^\sigma(t) = \sigma \frac{g^* \mu_B |\mathbf{B}^{\perp\sigma}_{\mathrm{nuc},d}(t)|^2}{4|B_{\mathrm{ext}}|}$ in the reduced Hamiltonian, Eq. (13). Although the transverse components of the Knight field are also reversed after the echo pulse, the fact that they do not commute with H_0 means that difference in the final states $|\Psi_{f+}\rangle$ and $|\Psi_{f-}\rangle$ may survive the echo.

Below we first focus our discussion on a theoretical model which, in the semi-classical treatment, produces perfect revivals in the echo signal due to exact commensuration between Larmor periods of different nuclear species. We separate the discussion into two cases, in which the free evolution time τ is either exactly on, or is away from, a revival peak. Then, at the end of the section we consider the quantum error at a revival peak which is not perfect, even within the semi-classical approximation, due to a lack of commensuration between nuclear Larmor periods.

For the initial state $|\Psi_i\rangle$ given above, which is a product of "giant spin" coherent states in each of the K groups of nuclear spins, we argue that, to leading order in N_d, the final states $|\Psi_{f\sigma}\rangle$ remain approximately tensor products of coherent states,

$$|\Psi_{f\sigma}\rangle \approx \bigotimes_k |I_k, \hat{\mathbf{n}}_{k,f\sigma}\rangle \tag{48}$$

Within this picture, $\sigma H_\perp^\sigma(t)$ causes the giant spin coherent states to rotate to new directions $\{\hat{\mathbf{n}}_{k,f\sigma}\}$, which depend on the electron spin state $\sigma = +$ or $-$. Other quantum effects such as coherent spin state squeezing due to the $(I_k^x)^2 + (I_k^y)^2$ terms in $H_\perp^\sigma(t)$, which may stretch the coherent states anisotropically, or a build-up of quantum correlations between different giant spins, are also expected. However, these effects enter only at higher order in $1/I_k$, because they are seeded by the small quantum fluctuations of the spin components in the initial coherent state.

Using the argument above, we find that away from the revival peaks the angle between the directions of the two final states of each giant spin k, for $\sigma = +$ or $-$, scales as $1/I_k$. Why is this so? First, according to Eq. (5), the Knight field acting on the nuclear spins, $A_{d,n}\mathbf{S}_d$, is smaller than the Overhauser field acting on the electron by a factor $1/\sqrt{N_d} \sim 1/I_k$. Second, assuming that the timescale of the electron spin coherence collapse is comparable to the timescale between revivals, the contribution of $H_\perp(t)$ to the Overhauser field causes electron spin precession through an angle of order 2π. Over the same time interval, the corresponding part of the Knight field will cause the nuclear spins to rotate through an angle which is $1/I_k$ times smaller.

The overlap $|\langle \Psi_{f+} | \Psi_{f-} \rangle|$ can thus be approximated to leading order by a product of overlaps between pairs of coherent states $\prod_k |\langle I_k, \hat{\mathbf{n}}_{k,f+} | I_k, \hat{\mathbf{n}}_{k,f-} \rangle|$, which are misaligned by an angle that scales down as $1/I_k$. Furthermore, because I_k is large, each coherent state $|I_k, \hat{\mathbf{n}}_{kf\pm}\rangle$ is characterized by a Gaussian phase space distribution with angular width of order $1/\sqrt{I_k}$. The reduction of the overlap, $1 - |\langle I_k, \hat{\mathbf{n}}_{k,f+} | I_k, \hat{\mathbf{n}}_{k,f-} \rangle|$, therefore scales as $1/I_k \sim \sqrt{\frac{K}{N_d}}$. We therefore find that the overall reduction of the total overlap, $1 - |\langle \Psi_{f+} | \Psi_{f-} \rangle|$, scales down at least as $\frac{K}{I_k} \sim \sqrt{K^3/N_d}$, as does the quantum error.

However, exactly at a revival peak, where the condition of perfect nuclear precession commensurability is met, this leading contribution vanishes. Here, for a given giant spin k, treating all other spins $k' \neq k$ as classical vectors, the precession and squeezing induced by $H_\perp^\sigma(t)$ is perfectly reversed after the π-pulse. Thus the quantum error at a revival peak is a higher order effect in $1/\sqrt{N_d}$. Numerical simulations with just two giant spins indicate that the quantum error at the revival peaks may scale down even faster then $1/N_d$, but a more detailed analysis is a subject for further study.

In more realistic cases, when the commensurability condition of all nuclear species cannot be exactly met, the quantum error near the quasi-revival peak scales like $f(\tau)\sqrt{K^3/N_d}$, with the pre-factor $f(\tau)$ becoming small as τ approaches the approximate commensuration point. At the quasi-revival peak, $f(\tau)$ is then dominated by the

classical effect of imperfect commensuration, captured by $1 - C_{sc}(\tau)$. We estimate the error in the case of GaAs quantum dots with $N_d = (2-4) \times 10^6$. For this estimate, we take the minimal number of groups of nuclear spins K which, within the semi-classical treatment, accurately produces the echo signal behavior near the revivals peaks, without changing significantly upon further refinement to more groups. For that purpose, note that only the spread of the Larmor frequencies for each nuclear species, $\langle \delta\omega_\alpha^2 \rangle$, enters the expression for $1 - C_{sc}(\tau)$ in Eq. (47). Consequently, it is sufficient to divide each of the three species into two collective groups. Grouping the nuclei into $K \approx 6$ collective giant spins gives $\sqrt{K^3/N_d} \lesssim 1/90$. Therefore the quantum contribution to the imperfect revival is smaller than the semi-classical contribution by more than an order of magnitude.

VII. DISCUSSION

Equation (42), together with Eq. (40), was used to fit the experimental echo signal data in Ref. 18. As stated in Sec. VI C, in general the matrix T_{kl} and its eigenvalues $\{\lambda_m\}$ are different for each dot due to variations in dot size and local environment. In particular, the distribution of hyperfine couplings, $\{A_k\}$, and the number of nuclei, N_d, depend on the distribution of electron density in the dot. Furthermore, the distribution of $\{\delta\omega_k\}$ depends on, e.g. local electric field gradients which couple to the nuclear quadrupole moments. However, in practice we found that choosing the same parameters for T_{kl} in the two dots produced a very good fit to the experimental data. Allowing, for example, different values of N_d for the two dots did not significantly improve the fits.

The spread of the Larmor frequencies $\sqrt{\langle \delta\omega_\alpha^2 \rangle}$ that was found from fitting to the experimental results[18] was equivalent to 3 Gauss effective spread of magnetic field, somewhat bigger than the values of NMR line-widths, typically about 1 Gauss equivalent spread, reported for bulk GaAs in the literature[39,40]. In addition to a random contribution to the local magnetic field coming from the dipolar interaction with neighboring nuclei, the nuclear spins in a GaAs quantum dot experience a quadrupolar splitting due to electric field gradients originating from the confined electrons, which we estimate to be at the order of a few Gauss (see Ref. 18, supplementary materials). These quadrupolar shifts may be responsible for the difference between the observed value and the NMR line-width. In the echo experiments which have been performed so far there is no way to distinguish between different origins of the apparent spread of nuclear Larmor frequencies (i.e. between decay due to nuclear spin flip-flop, due to inhomogeneous broadening, or due to quadrupolar effects).

Our model does not directly include the dipole-dipole-induced temporal decay of *local* nuclear spin correlations $\langle I_n^i(0) I_n^j(t) \rangle$. However, such decay is accounted for phenomenologically by a time-independent spread of site-dependent Larmor precession frequencies. Because the current Hahn echo experiments can't distinguish between these intrinsic and extrinsic nuclear spin dephasing processes, our approximation of accounting for these effects by a random static, disordered, Zeeman field is reasonable. According to Eq. (47), the affect of this random field is to cause an additional decay to the echo signal with a timescale $\tau_\perp$. This decay becomes dominant at low magnetic fields, as $\tau_\perp$ becomes shorter then the decay time associated with the spectral diffusion, τ_{SD}.

In addition, note that the spectral diffusion decay time τ_{SD} was found experimentally to be independent of the magnetic field strength. Other experiments[41] have also suggested that the external magnetic field does not significantly influence spin-diffusion at the relevant field range, above 20 mT. This provides further justification for neglecting the commutators $[H_\perp, H_d]$ in section VI, which, *a priori*, could introduce such a dependence.

In conclusion, we have provided physical justification for treating the Overhauser fields in a GaAs double quantum dot system as classical vector variables, based on the relative smallness of the commutation relations due to the large number of spins in each dot. For the simple model in section V, in which we ignored the nuclear dipole-dipole interaction and the Knight shift of the nuclear Larmor frequencies, we demonstrated the equivalence of the semi-classical treatment and the quantum diagrammatic summation of Ref. 32. The semi-classical treatment quantitatively captures the observed phenomena of monotonic decay of the echo signal in strong magnetic fields, and the collapses and revivals of the echo signal in weaker magnetic fields. The overall effect of the Knight field on the spin-echo is found to be negligible. This fact is parameter dependent. However note that the Knight field is reversed by the echo pulse. As a result, If one treats the Knight field contribution to the phase in Eqs. (39) and (43) as a perturbation, it vanishes in the leading order in τ, near the revivals peak. This is unlike the contribution of competing process, i.e. the spread of the Larmor frequencies. This means that, for the typical parameters in GaAs quantum dots, after averaging over all nuclear spin states, both the collapse-revivals effect and the overall envelope decay can be understood simply as arising from averaging over the initial conditions of the Larmor precession of the nuclear spins.

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