Universal power law decay of spin polarization in double quantum dot

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(Received 22 May 2019; revised manuscript received 23 July 2019; published 5 August 2019)

We study the spin dynamics and spin noise in a double quantum dot, taking into account the interplay between hopping, exchange interaction, and the hyperfine interaction. At short timescales the spin relaxation is governed by the spin dephasing in the random nuclear fields. At long timescales the spin polarization obeys universal power law $1/t$ independent of the relation between all the parameters of the system. This effect is caused by the competition between the spin blockade effect and the hyperfine interaction. The spin noise spectrum of the system universally diverges as $\ln(1/\omega)$ at low frequencies.

DOI: 10.1103/PhysRevB.100.075409

I. INTRODUCTION

The most fascinating discoveries in solid-state physics in the 21st century are arguably related to the spin degree of freedom of electrons. Intense studies of the spin-related phenomena led to the formation of a new branch in solid-state physics: spintronics [1]. The spin-related phenomena are most pronounced in low-dimensional structures due to the enhanced role of the spin-orbit and hyperfine interactions [2–4]. From a practical point of view, the most promising for quantum information processing are zero-dimensional nanosystems, such as shallow impurities, color centers, and quantum dots (QDs).

There are two complementary approaches to study spin-related phenomena in QDs. The first one is based on optical spin orientation, manipulation, and detection and is usually applied to self-organized quantum dots [5–7]. The second one is based on electrical spin injection and detection in gate-defined quantum dots [8], which makes use of the external magnetic field. An interesting and promising system for the latter approach is a double quantum dot [9,10], which demonstrates the Pauli or spin blockade effect [11,12]. This effect was studied in detail theoretically [13–18], but the spin dynamics was investigated mainly in the presence of electric current and external magnetic field.

In this work we study manifestations of the spin blockade effect in the spin dynamics of a double quantum dot isolated from the environment in the absence of external magnetic field. Our theory can also be applied for an isolated pair of donors which are close to each other but far enough from the other donors.

The spin dynamics in quantum dots in zero magnetic field is largely driven by the hyperfine interaction with the host lattice nuclear spins [19]. In a double quantum dot the exchange interaction [20,21] and electron hopping [22,23] are also important and affect the spin dynamics. We stress that we consider only hopping between the QDs but not to the contacts or substrate [24,25]. The interplay between exchange interaction, hopping, and hyperfine interaction can hardly be investigated for large spin ensembles. But the double-quantum-dot system considered here allows for the exact solution and gives some hints about spin dynamics in larger spin systems.

In our study we focus on two effects: the spin relaxation and spin noise. The first one assumes the spin orientation and measurement of the spin polarization decay. The second one is based on the continuous measurement of the dynamics of spin fluctuations in thermal equilibrium [26,27]. We demonstrate that in both cases the spin dynamics essentially consists of the spin precession in a random nuclear field and a slow power law relaxation. The latter effect is a consequence of the interplay between the spin blockade and the hyperfine interaction.

We demonstrate that the measurement of the spin dynamics at short timescales or the high frequency spin noise spectra allows one to determine the parameters of the spin dynamics in the double QD. At the same time we find that the spin polarization decays as $1/t$ at long timescales for any relation between the hopping rates, the strength of the hyperfine interaction, and the exchange interaction. The revealed universality suggests that this effect can be easily observed experimentally. The practical importance of the power law spin relaxation is determined by the possibility to preserve parametrically more spin polarization at long timescales than for the usual exponential spin relaxation.

This paper is organized as follows. In the next section we present the model of the system under study. In Sec. III we present our approach to calculate the spin dynamics and spin noise spectrum. We show numerical results for the arbitrary relation between the system parameters and stress the

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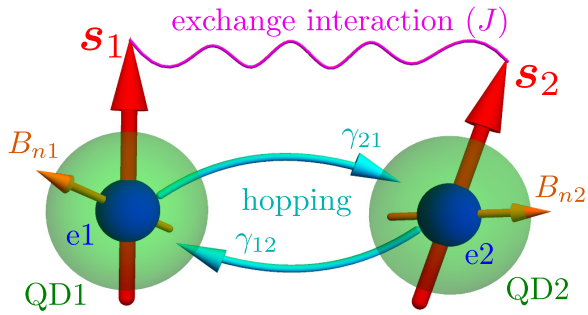

FIG. 1. Sketch of the double-QD system. The two electrons are represented by the blue balls, and their spins are shown by the red arrows. Orange arrows show the Overhauser field acting in each dot; the magenta wavy line denotes the exchange interaction. The cyan arrows show the possible hops of electrons between the QDs (green transparent balls).

universality of the power law $\propto 1/t$ spin relaxation. Then in Sec. IV we derive the analytical results in the limiting cases, which explain the numerical results. Further, in Sec. V we discuss the limits of applicability of our model, and finally, we summarize our findings in Sec. VI.

II. MODEL

We consider a double QD with two electrons, as shown in Fig. 1. We assume that the two electrons can be localized either in different QDs or in the same QD and can hop between the QDs. We take into account the exchange interaction between electrons and their hyperfine interaction with the host lattice nuclear spins. The Hilbert space of the system under study consists of six states: the two singlet states, when the two electrons are localized in the same QD, plus another singlet state and three triplet states, when the two electrons are localized in different QDs.

The Hamiltonian of the system has the form

$$\mathcal{H} = \sum_{i,\sigma} E_i n_i^\sigma + \sum_i U_i n_i^+ n_i^- + J\mathbf{s}_1\mathbf{s}_2 + \hbar\sum_i \mathbf{\Omega}_i\mathbf{s}_i. \quad (1)$$

Here in the first term E_i are the localization energies of electrons in the ith QD ($i = 1, 2$), and n_i^σ ($\sigma = \pm$) are the occupancies of the states, characterized by the spin index σ. The corresponding operators can be written using the Fermi creation (annihilation) operators $c_{i\sigma}^\dagger$ ($c_{i\sigma}$) as $n_i^\sigma = c_{i\sigma}^\dagger c_{i\sigma}$. The second term in Eq. (1) describes the on-site electron repulsion with the Hubbard energy U_i. The third term is the exchange interaction, characterized by the constant J, which also includes the tunneling-related contribution [8]. The spin operators can be expressed as

$$\boldsymbol{s}_i = \tfrac{1}{2}\boldsymbol{\sigma}_{\sigma\sigma'} c_{i\sigma}^\dagger c_{i\sigma'}, \quad (2)$$

where $\boldsymbol{\sigma} = (\sigma_x, \sigma_y, \sigma_z)$ is the vector composed of the Pauli matrices. Finally, the last term in Eq. (1) describes the hyperfine interaction, with $\mathbf{\Omega}_i$ being the spin precession frequency in the fluctuation of the Overhauser field of the host lattice nuclear spins. In this study we assume the number of host lattice nuclear spins in each QD is large, so that the Overhauser field can be considered static ("frozen") [28].

The electron hopping, being an inelastic process, cannot be described solely by the electron Hamiltonian. One has to consider the total Hamiltonian

$$\mathcal{H}_{\text{tot}} = \mathcal{H} + \mathcal{H}_{ph} + \mathcal{V}, \quad (3)$$

which includes a phonon Hamiltonian $\mathcal{H}_{ph}$ and an electron phonon interaction $\mathcal{V}$. The phonon bath energy is given by

$$\mathcal{H}_{ph} = \hbar\sum_{\boldsymbol{q}} \Omega_q b_{\boldsymbol{q}}^\dagger b_{\boldsymbol{q}}, \quad (4)$$

where $\Omega_{\boldsymbol{q}}$ is the phonon frequency, corresponding to the wave vector $\boldsymbol{q}$, and $b_{\boldsymbol{q}}^\dagger$ ($b_{\boldsymbol{q}}$) is the phonon creation (annihilation) operator. We assume the phonon polarization index is included in $\boldsymbol{q}$. The electron-phonon interaction after the canonical (polaron) transformation [29,30] can be written as

$$\mathcal{V} = V\sum_{i,\sigma,\boldsymbol{q}} \gamma_q (e^{i\boldsymbol{q}\boldsymbol{R}_i} b_{\boldsymbol{q}} - e^{-i\boldsymbol{q}\boldsymbol{R}_i} b_{\boldsymbol{q}}^\dagger) c_{\bar{\imath}\sigma}^\dagger c_{\bar{\imath}\sigma} + \text{H.c.}, \quad (5)$$

where V is the hopping constant; $\gamma_q = v_q/(\hbar\Omega_q)$, with v_q being the electron-phonon interaction constant; $\boldsymbol{R}_{1,2}$ are the coordinates of the QDs; and the symbol $\bar{\imath}$ denotes the quantum dot other than i ($\bar{\imath} = 2$ if $i = 1$ and $\bar{\imath} = 1$ if $i = 2$). The justification of this approach is given in the Appendix.

The spin dynamics in the system can be described using the density matrix formalism. In the description of electron hopping we assume that the first two terms in the Hamiltonian (1) and the temperature exceed by far the two latter terms, so the states where two electrons are in the different QDs have nearly the same energy. In this case the off-diagonal matrix elements between the states with essentially different energies can be neglected. As a result the system is described by the 4×4 density matrix ρ in the basis of the four states of two electrons in different QDs and the two probabilities P_i to find the two electrons in QD i.

In the two lowest orders in the electron-phonon interaction the total density matrix of the electron system ρ_{tot} satisfies the equation

$$\dot{\rho}_{\text{tot}} = -\frac{i}{\hbar}[\mathcal{H}, \rho_{\text{tot}}] + \frac{\pi}{\hbar}\langle[2\mathcal{V}\rho_{\text{tot}}\mathcal{V} - \rho_{\text{tot}}\mathcal{V}^2 - \mathcal{V}^2\rho_{\text{tot}}]\delta(E_i - E_f)\rangle_{ph}, \quad (6)$$

where the first line describes the coherent spin dynamics and the second one describes the electron hopping. The angular brackets denote averaging of the phonon creation and annihilation operators over the phonon states. The energies E_i and E_f are the total energies of the system before and after the hop, respectively, including the phonon energy.

From Eq. (6) we find that the electron density matrix ρ obeys the master equation

$$\dot{\rho} = -\frac{i}{\hbar}[\mathcal{H}, \rho] + \frac{1}{2}\sum_i\sum_{\sigma,\sigma'}[2\Gamma_{i\bar{\imath}} c_{i\sigma}^\dagger c_{\bar{\imath}\sigma} P_{\bar{\imath}} c_{\bar{\imath}\sigma'}^\dagger c_{i\sigma'} - \gamma_{i\bar{\imath}}(\rho c_{\bar{\imath}\sigma'}^\dagger c_{i\sigma'} c_{i\sigma}^\dagger c_{\bar{\imath}\sigma} + c_{\bar{\imath}\sigma'}^\dagger c_{i\sigma'} c_{i\sigma}^\dagger c_{\bar{\imath}\sigma}\rho)], \quad (7)$$

where we introduce the rates $\gamma_{i\bar{\imath}}$ and $\Gamma_{i\bar{\imath}}$ describing the hopping from QD $\bar{\imath}$ to i when QD i is occupied or empty, respectively. On the right-hand side of this equation P_i should be treated as the corresponding operators multiplied by their

average values, similar to ρ. The hopping rate with the change of energy by ΔE is [4,31]

$$\gamma(\Delta E) = \frac{2\pi}{\hbar} V^2 2\gamma_{q_{\Delta E}}^2 D(|\Delta E|)[N_{\Delta E} + \theta(-\Delta E)], \tag{8}$$

where $q_{\Delta E}$ is the phonon wave vector corresponding to a phonon with energy $|\Delta E|$; $D(|\Delta E|)$ stands for the density of phonon states; $N_{\Delta E} = 1/[\exp(|\Delta E|/k_B T) - 1]$ is the occupancy of the corresponding state, with T being the temperature; and $\theta(x)$ is the Heaviside step function. The specific hopping rates between the QDs are given by

$$\gamma_{i\bar{i}} = \gamma(E_i - E_{\bar{i}} + U_i), \tag{9a}$$

$$\Gamma_{i\bar{i}} = \gamma(E_i - E_{\bar{i}} - U_{\bar{i}}). \tag{9b}$$

One can see that in the general case the relation $\Gamma_{i\bar{i}} \geqslant \gamma_{i\bar{i}}$ holds because of the electron Coulomb repulsion in the same QD.

In a way similar to Eq. (7) the probabilities P_i obey

$$\dot{P}_i = \frac{1}{2}\sum_{\sigma,\sigma'}[2\gamma_{i\bar{i}} c_{i\sigma}^\dagger c_{\bar{i}\sigma}\rho c_{\bar{i}\sigma'}^\dagger c_{i\sigma'} - \Gamma_{\bar{i}i}(P_i c_{i\sigma'}^\dagger c_{\bar{i}\sigma'} c_{\bar{i}\sigma}^\dagger c_{i\sigma} + c_{i\sigma'}^\dagger c_{\bar{i}\sigma'} c_{\bar{i}\sigma}^\dagger c_{i\sigma} P_i)]. \tag{10}$$

The electron conservation rule for this system can be written as

$$P_1 + P_2 + P_{12} = 1, \tag{11}$$

where $P_{12} = (n_1^+ + n_1^-)(n_2^+ + n_2^-) = \mathrm{Tr}\,\rho$ is the probability of finding the two electrons in different QDs.

We recall that we assume the nuclear fields $\boldsymbol{\Omega}_i$ are frozen. They are created by the nuclear spin fluctuations and are described by the Gaussian distribution function

$$\mathcal{F}(\boldsymbol{\Omega}_i) = \frac{1}{(\sqrt{\pi}\delta)^3} e^{-\Omega_i^2/\delta^2}, \tag{12}$$

with the parameter δ characterizing the dispersion. In order to obtain experimentally observable spin dynamics, the solution of the spin dynamics equations should be averaged over this distribution function. In the next section we demonstrate that this procedure ultimately leads to the spin decay $\propto 1/t$ at long timescales.

III. SPIN RELAXATION AND SPIN NOISE

The master equation (7) can be rewritten in the form of equations for the spin operators $\boldsymbol{s}_i$ and their correlation functions $s_i^\alpha s_j^\beta$, where α and β are the Cartesian indices. Their average values can be expressed through the density matrix as $\langle \boldsymbol{s}_i \rangle = \mathrm{Tr}(\boldsymbol{s}_i \rho)$ and $\langle s_i^\alpha s_j^\beta \rangle = \mathrm{Tr}(s_i^\alpha s_j^\beta \rho)$.

The electron spins obey

$$\frac{d\boldsymbol{s}_i}{dt} = \boldsymbol{\Omega}_i \times \boldsymbol{s}_i + \frac{J}{\hbar}\boldsymbol{s}_{\bar{i}} \times \boldsymbol{s}_i - \frac{\gamma}{2}(\boldsymbol{s}_i - \boldsymbol{s}_{\bar{i}}), \tag{13a}$$

where $\gamma = \gamma_{12} + \gamma_{21}$ is the total hopping rate for the singlet state of the two electrons in different QDs. The first term on the right-hand side in this equation describes the spin precession with the frequency $\boldsymbol{\Omega}_i$. Similarly, the second term describes the electron spin precession in the effective exchange magnetic field of another electron. Finally, the last term describes the electron hopping [32,33]. One can see that this term vanishes in the case of $\boldsymbol{s}_1 = \boldsymbol{s}_2$ due to the spin blockade. By contrast, the hopping rate equals γ when the two electrons are in the singlet state, i.e., $\boldsymbol{s}_1 = -\boldsymbol{s}_2$. In the general case $\boldsymbol{s}_1 - \boldsymbol{s}_2$ decays due to this term, while $\boldsymbol{s}_1 + \boldsymbol{s}_2$ does not. We recall that the relation $k_B T, U_{1,2} \gg J, \hbar\delta$ is assumed, so there is no spin polarization in the thermal equilibrium.

We stress that the term $\boldsymbol{s}_1 \times \boldsymbol{s}_2$ in Eq. (13a) does not simply reduce to the product of the two average values but should be treated as a vector composed of spin correlators. The spin correlation functions in general obey the equations

$$\frac{d}{dt}\left(s_1^\alpha s_2^\beta\right) = \varepsilon_{\alpha\gamma\delta}\Omega_1^\gamma s_1^\delta s_2^\beta + \varepsilon_{\beta\gamma\delta}\Omega_2^\gamma s_1^\alpha s_2^\delta + \frac{J}{4\hbar}\varepsilon_{\alpha\beta\gamma}\left(s_1^\gamma - s_2^\gamma\right) - \frac{\gamma}{2}\left(s_1^\alpha s_2^\beta - s_1^\beta s_2^\alpha\right) + \frac{\delta_{\alpha\beta}}{2}(\gamma P_s - \Gamma_{21}P_1 - \Gamma_{12}P_2), \tag{13b}$$

where $P_s = P_{12}/4 - \boldsymbol{s}_1\boldsymbol{s}_2$ is the occupancy of the singlet state in the two different QDs. The first two terms on the right-hand side of this expression describe the spin precession in the nuclear field. The third term is related to the exchange interaction and reduces to the first power of the spin operators for the spin-1/2 particles. The rest of the terms describe the hopping of electrons and deserve a longer discussion.

The hopping of two electrons to the same QD brings the system to the singlet state with the zero total angular momentum. At the same time, the hopping does not change the total angular momentum, so it is allowed only for the singlet spin state, in agreement with the Pauli exclusion principle. The correlators $s_1^\alpha s_2^\beta$ can be combined in the groups, which transform according to the representations D_2, D_1, and D_0 of the SU(3) group. The five correlators $s_1^z s_2^z - s_1^x s_2^x$, $s_1^z s_2^z - s_1^y s_2^y$, and $s_1^\alpha s_2^\beta + s_1^\beta s_2^\alpha$, with $\alpha \neq \beta$, belong to the D_2 representation and do not decay because they require the two electron spins to be parallel. The three combinations $s_1^\alpha s_2^\beta - s_1^\beta s_2^\alpha$ belong to the D_1 representation and decay with the rate γ, which is described by the second line of Eq. (13b). Finally, the correlator $\boldsymbol{s}_1\boldsymbol{s}_2$ belongs to the D_0 representation, and it couples to the scalar occupancies P_1, P_2, and P_{12}, which are described by the last line. This term consists of two contributions: hopping to the states, where the two electrons are localized in the same QD with the rate γ, and hopping from these states with the rates Γ_{12} and Γ_{21}.

The above equations describe the spin dynamics and should be accompanied by kinetic equations for the occupancies of the states. Taking into account Eq. (11), it is enough to write the two equations

$$\dot{P}_i = -2\Gamma_{\bar{i}i}P_i + 2\gamma_{i\bar{i}}P_s. \tag{14}$$

The set of 18 equations (11), (13), and (14) is equivalent to Eqs. (7) and (10) and completely describes the spin and charge dynamics.

Below we assume the high-temperature limit (the thermal energy is much larger than the Hubbard energies U_i), so that $\gamma_{12} = \Gamma_{21}$ and $\gamma_{21} = \Gamma_{12}$. This assumption requires that the size quantization energy is larger than the Hubbard energy, which is true for QDs smaller than the exciton Bohr radius. Moreover, we assume both these rates are equal to $\gamma/2$ for simplicity, but these assumptions do not qualitatively change

the results. Then it is convenient to introduce the parameter

$$X = P_1 + P_2 - 2P_s, \tag{15}$$

which describes the deviation of the occupancies from their steady-state values. This parameter simply obeys

$$\frac{dX}{dt} = 2(\mathbf{\Omega}_1 - \mathbf{\Omega}_2)(\boldsymbol{s}_1 \times \boldsymbol{s}_2) - 4\gamma X. \tag{16}$$

Note that the same parameter also describes the dynamics of the spin correlators in Eq. (13b). Thus, in this case one can consider only 16 equations: Eqs. (13) and (16). The spin dynamics can be calculated for the given initial conditions, and the double-QD system is characterized in total by the three parameters: J, δ, and γ.

To describe the spin relaxation we consider the initial conditions $\boldsymbol{s}_1(0) = \boldsymbol{s}_2(0) = \boldsymbol{e}_z/2$ (triplet state), where $\boldsymbol{e}_z$ is a unit vector along some z axis. These initial conditions correspond to the optical spin orientation, and they are opposite to what is realized in an electrically controlled double-QD system [16]. Figure 2(a) shows the evolution of the z component of the total spin $\boldsymbol{S} = \boldsymbol{s}_1 + \boldsymbol{s}_2$ in the most involved case, when all the parameters are of the same order $J/\hbar \sim \delta \sim \gamma$ (their values are given in the figure caption). The spin dynamics can be separated into two contributions; below we describe them separately.

(i) The total spin quickly decays from 1 to less than 0.1 and then increases again at $t\delta \sim 5$. This time dependence is typical for the spin dephasing in a random Overhauser field [28,34,35]. Notably, the spin polarization does not decay to zero due to the conservation of the spin component parallel to the Overhauser field in each QD. The exchange interaction "exchanges" the electrons in the two QDs, so the direction of precession of the given electron spin changes. This, however, also does not lead to the complete spin relaxation [21]. Indeed, in the limit of very strong exchange interaction the hyperfine field does not mix the singlet and triplet states, so the component of the total spin $\boldsymbol{S}$ along the average Overhauser field $\mathbf{\Omega} = (\mathbf{\Omega}_1 + \mathbf{\Omega}_2)/2$ is conserved.

(ii) At long timescales the total spin slowly decays to zero due to the *hopping* of electrons between the QDs. In fact, this is a power law decay $S_z(t) \propto 1/t$, as shown by the red dashed line in Fig. 2(a). This asymptote is more clearly shown in the inset, where the longer timescales are shown in the bilogarithmic scale. We checked numerically that this law of spin relaxation is valid for an arbitrary relation between the parameters $J/\hbar$, γ, and δ. Moreover, this law will be derived analytically in a number of limiting cases in the next section. To understand the effect qualitatively we note that in the exceptional case $\mathbf{\Omega}_1 \parallel \boldsymbol{e}_z$ and $\mathbf{\Omega}_2 \parallel \boldsymbol{e}_z$ the total spin does not change, and the hopping is also forbidden [see Eq. (13a)]. So in this case the spin polarization (in our model) does not decay at all. In the more probable situation, when $\mathbf{\Omega}_1 \parallel \mathbf{\Omega}_2$, the component of the total spin along this direction does not decay either because of the spin blockade. Finally, in the general case of an arbitrary angle between $\mathbf{\Omega}_1$ and $\mathbf{\Omega}_2$ the smaller the angle is, the slower the spin polarization decays. Averaging over the Gaussian distribution of the Overhauser fields results in the power law decay $S_z(t) \propto 1/t$ at long timescales.

We note that for localized electron states in quantum wires the power law spin relaxation appears when neglecting the electron-electron interaction and is caused by the nearly isolated localized states, which have a very small tunneling rate [36]. Here, by contrast, we consider a finite hopping time and take into account the spin blockade effect.

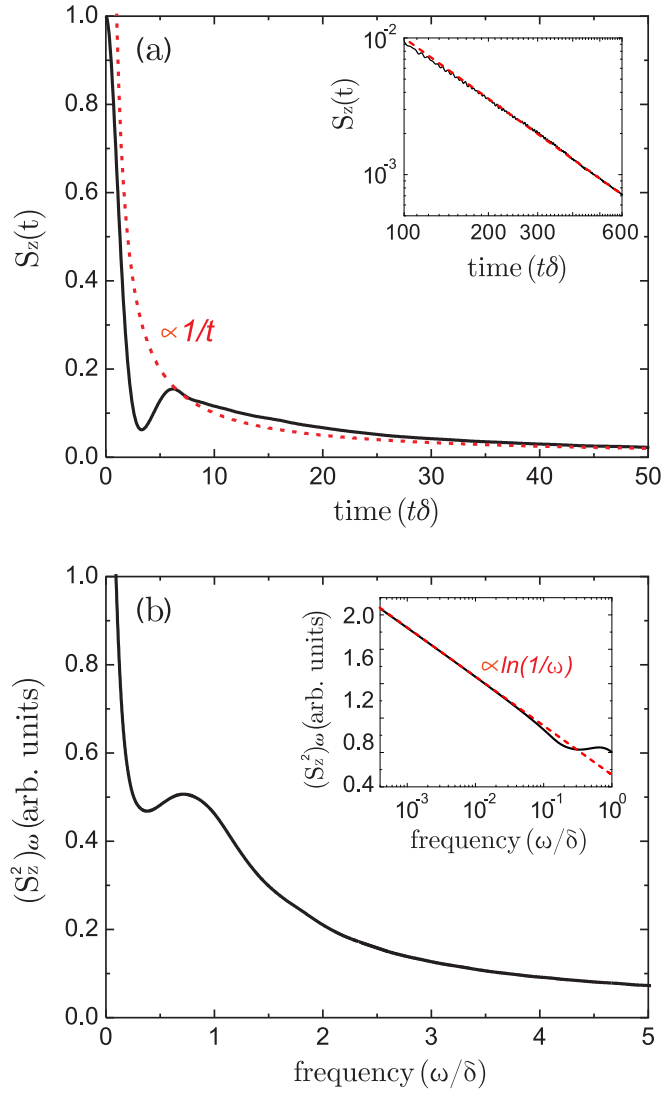

FIG. 2. (a) The spin relaxation for the initial conditions $\boldsymbol{s}_i(0) = \boldsymbol{e}_z/2$. The red dashed lines show the asymptote $\propto 1/t$. (b) The spin noise spectrum. The red dashed curve in the inset shows the asymptote $\propto \ln(1/\omega)$. The parameters of the calculation are $J = 0.9\hbar\delta$ and $\gamma = 1.1\delta$.

The slow spin decay can be conveniently revealed in the frequency domain. Experimentally, the spin dynamics at low frequencies can be studied by means of the spin noise spectroscopy [26]. This method is based on the measurement of the correlation functions of the spin fluctuations in the thermal equilibrium. The spin noise spectrum $(\delta S_z^2)_\omega$ is defined as a Fourier transform of the autocorrelation function

$$\left(\delta S_z^2\right)_\omega = \int_{-\infty}^{\infty} \langle \delta S_z(t) \delta S_z(t+\tau) \rangle \, e^{i\omega\tau} d\tau, \tag{17}$$

where the angular brackets denote averaging over t. In the equilibrium the spin polarization is absent, so $\langle \boldsymbol{S}(t) \rangle = 0$ and $\delta\boldsymbol{S} = \boldsymbol{S}$ in the system under study.

To calculate the correlation functions we note that the correlators at $\tau = 0$ can be simply found from the

steady-state solution of the equations of motion. One finds that the correlation functions of S_z with all the other operators in Eqs. (13) and (16) are zero except for

$$\langle S_z s_i^z \rangle = \frac{\langle P_{12} \rangle}{4}. \tag{18}$$

In the thermal equilibrium $\langle \boldsymbol{s}_1 \boldsymbol{s}_2 \rangle = 0$, so from Eqs. (14) and Eq. (11) we find that

$$\langle P_{12} \rangle = \left(1 + \frac{\gamma_{12}}{4\Gamma_{21}} + \frac{\gamma_{21}}{4\Gamma_{12}} \right)^{-1}. \tag{19}$$

In the case $\gamma_{i\bar{i}} = \Gamma_{i\bar{i}} = \gamma/2$ one has $\langle P_{12} \rangle = 2/3$, in agreement with Eq. (16). So for the total spin we obtain

$$\langle S_z^2 \rangle = \tfrac{1}{3}. \tag{20}$$

The correlators define the initial conditions for the time correlation functions.

Then the set of the correlators of $\delta S_z(t)$ with the other operators taken at time $t + \tau$ obeys the same equations of motion, Eqs. (13) and (16), for $\tau > 0$ [37]. Moreover, the spin autocorrelation function is an even function of τ, which allows us to find $\langle \delta S_z(t) \delta S_z(t+\tau) \rangle$ and the spin noise spectrum $(\delta S_z^2)_\omega$ after Eq. (17). We note that the spin noise spectrum can also be calculated directly in the frequency domain by replacing the time derivatives in the equations of motion with the multipliers $-i\omega$ [38].

The spin noise spectrum is shown in Fig. 2(b) for the same system parameters as in Fig. 2(a). It again consists of two contributions. (i) The first is a peak at frequency $\omega \sim \delta$, which corresponds to the spin precession in the Overhauser field [39,40]. Its shape reproduces the distribution function of the absolute values of the Overhauser field [39,41]. (ii) The other is a peak at zero frequency, which corresponds to the slow spin decay at long times. This peak corresponds to the divergence $(\delta S_z^2)_\omega \propto \ln(1/\omega)$ at $\omega \to 0$, in agreement with the asymptote $\langle \delta S_z(t) \delta S_z(t+\tau) \rangle \propto 1/\tau$ in the time domain. The logarithmic asymptote for the spin noise spectrum is shown in the inset in Fig. 2(b).

Thus, the spin relaxation and the spin noise spectrum essentially describe the same spin dynamics in the time and frequency domains, respectively.

IV. LIMITING CASES

The main result of the previous section is the very slow power law decay $\propto 1/t$ of the spin polarization despite all the necessary ingredients for the spin relaxation in the model. This result corresponds to the divergence of the spin noise spectrum at zero frequency $\propto \ln(1/\omega)$. In this section we derive these asymptotes in limiting cases when one of the system parameters, δ, $J/\hbar$, or γ, is much larger than the other two.

A. Strong hyperfine interaction

The limit $\delta \gg J/\hbar, \gamma$ corresponds to the two nearly independent QDs, where the spins $\boldsymbol{s}_{1,2}$ precess around the corresponding nuclear fields $\boldsymbol{\Omega}_{1,2}$. As a result of this precession the initial spin polarization on average decays three times on the timescale $\sim 1/\delta$ [34]. One third of spin polarization on average is parallel to the static fluctuation of the Overhauser field and does not decay at this timescale. The exchange interaction only slightly changes the eigenfunctions and does not lead to the complete spin relaxation. By contrast, the hopping, being an incoherent process, leads to the complete decay of the spin polarization. As a result the exchange interaction in this limit can be neglected, while the hopping cannot.

The spin dynamics in this limit can be described by Eqs. (13) with $J = 0$:

$$\dot{\boldsymbol{s}}_i = \boldsymbol{\Omega}_i \times \boldsymbol{s}_i - \frac{\gamma}{2} \cdot (\boldsymbol{s}_i - \boldsymbol{s}_{\bar{i}}). \tag{21}$$

The last term of this expression was discussed after Eq. (13a). One can separate the spin components parallel and perpendicular to the nuclear field as

$$s_{i\parallel} = \boldsymbol{n}_i \boldsymbol{s}_i, \quad \boldsymbol{s}_{i\perp} = \boldsymbol{s}_i - \boldsymbol{n}_i s_{i\parallel}, \tag{22}$$

where $\boldsymbol{n}_i = \boldsymbol{\Omega}_i/\Omega_i$. These components approximately obey [32]

$$\dot{\boldsymbol{s}}_{i\perp} = \boldsymbol{\Omega}_i \times \boldsymbol{s}_{i,\perp} - \frac{\gamma}{2} \boldsymbol{s}_{i,\perp}, \tag{23a}$$

$$\dot{s}_{i\parallel} = -\frac{\gamma}{2} (s_{i\parallel} - s_{\bar{i}\parallel} \cos\theta), \tag{23b}$$

where $\cos\theta = \boldsymbol{n}_1 \boldsymbol{n}_2$ and we neglected the mixing between quickly oscillating components of $\boldsymbol{s}_{i\perp}$ and slowly varying $s_{i,\parallel}$. The solution of these equations gives

$$\begin{aligned} \boldsymbol{s}_1(t) + \boldsymbol{s}_2(t) = {} & \sum_i [\boldsymbol{s}_{i\perp}(0) \cos(\Omega_i t) + \boldsymbol{n}_i \times \boldsymbol{s}_i(0) \sin(\Omega_i t)] \\ & + \frac{\boldsymbol{n}_1 + \boldsymbol{n}_2}{2} [s_{1\parallel}(0) + s_{2\parallel}(0)] e^{-t\gamma(1-\cos\theta)/2} \\ & + \frac{\boldsymbol{n}_1 - \boldsymbol{n}_2}{2} [s_{1\parallel}(0) - s_{2\parallel}(0)] e^{-t\gamma(1+\cos\theta)/2}. \end{aligned} \tag{24}$$

This expression should be averaged over the distribution of $\boldsymbol{\Omega}_i$ [see Eq. (12)]:

$$\begin{aligned} & \langle \boldsymbol{s}_1(t) + \boldsymbol{s}_2(t) \rangle \\ & = [\boldsymbol{s}_1(0) + \boldsymbol{s}_2(0)] \frac{2}{3} \left\{ \left[1 - \frac{(\delta t)^2}{2} \right] e^{-(\delta t)^2/4} + \frac{e^{-\gamma t} + \gamma t - 1}{(\gamma t)^2} \right\}. \end{aligned} \tag{25}$$

This expression is shown by the red dashed curve in Fig. 3(a) and agrees with the numerical calculations, shown by the black solid curve. At $t \gg 1/\gamma$ this expression yields the power law decay [42]

$$\langle \boldsymbol{s}_1(t) + \boldsymbol{s}_2(t) \rangle = \frac{2}{3\gamma t}. \tag{26}$$

This expression is shown by the red dashed line in the inset in Fig. 3(a).

Since the spin correlation functions also obey an equation like Eq. (21), the spin noise spectrum can be found simply as a Fourier transform of Eq. (25) with $\boldsymbol{s}_1(0) + \boldsymbol{s}_2(0) = \boldsymbol{e}_z/3$ [see Eq. (20)]:

$$\left(S_z^2\right)_\omega = \frac{1}{\delta} f\left(\frac{\omega}{\delta}\right) + \frac{1}{\gamma} g\left(\frac{\omega}{\gamma}\right), \tag{27}$$

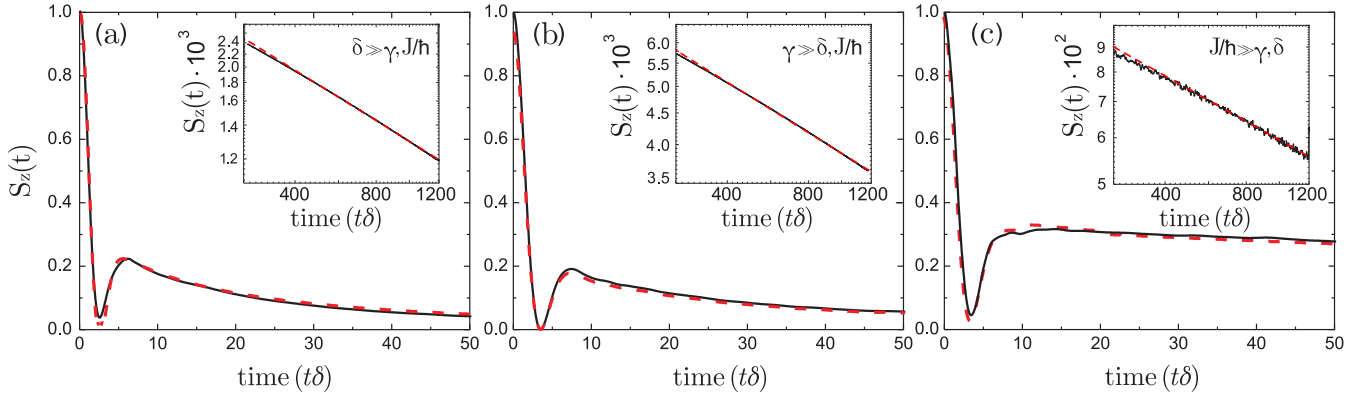

FIG. 3. Relaxation of the spin polarization $S_z(t)$ calculated numerically (black solid curves) and analytically (red dashed curves) for the three limiting cases (a) $J = 0.04\hbar\delta$ and $\gamma = 0.2\delta$ [Eq. (25)], (b) $J = 0.2\hbar\delta$ and $\gamma = 5\delta$ [Eq. (34)], and (c) $J = 5\hbar\delta$ and $\gamma = 0.2\delta$ [Eq. (41)]. The initial conditions are $\boldsymbol{s}_{1,2} = \boldsymbol{e}_z/2$. The insets show the power law decay $\propto 1/t$ for the same parameters.

where we introduce the functions

$$f(x) = \frac{8}{9}\sqrt{\pi}x^2 e^{-x^2}, \tag{28}$$

$$g(x) = \tfrac{2}{9}[\pi|x| + \ln(1+1/x^2) - 2(1 + x\,\mathrm{arctg}\,x)]. \tag{29}$$

The analytical expression for the spin noise spectrum in this limit is shown in Fig. 4 by the blue dashed curve and agrees with the numerical calculations (blue solid curve). At low frequencies the spin noise spectrum diverges as

$$\left(S_z^2\right)_\omega = \frac{4}{9\gamma}\ln\left(\frac{\gamma}{\omega}\right), \tag{30}$$

as expected.

B. Fast hopping between QDs

In the limit $\gamma \gg \delta, J/\hbar$ one could expect that the spin polarization quickly decays to zero because of the fast hops of electrons into one QD, where the total spin is zero. This, however, does not happen because of the spin blockade: when the two spins are parallel to each other, the electrons do not hop.

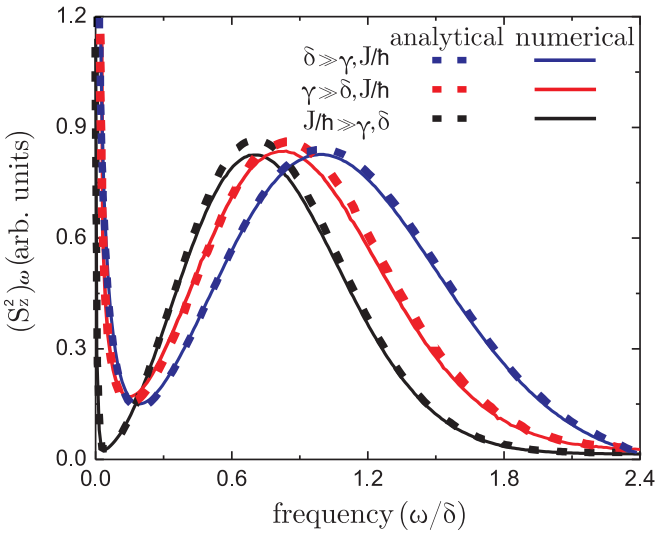

FIG. 4. Spin noise spectra calculated numerically (solid curves) and analytically (dashed curves) for the same parameters as in Fig. 3(a) (blue curves), Fig. 3(b) (red curves), and Fig. 3(c) (black curves); see Eqs. (27), (36), and (43), respectively.

It is convenient to rewrite Eqs. (13a) as

$$\dot{\boldsymbol{S}} = \boldsymbol{\Omega}\times\boldsymbol{S} + \Delta\boldsymbol{\Omega}\times\Delta\boldsymbol{S}, \tag{31a}$$

$$\Delta\dot{\boldsymbol{S}} = \Delta\boldsymbol{\Omega}\times\boldsymbol{S} - \gamma\Delta\boldsymbol{S} - 2\frac{J}{\hbar}\boldsymbol{s}_1\times\boldsymbol{s}_2, \tag{31b}$$

where $\Delta\boldsymbol{S} = \boldsymbol{s}_1 - \boldsymbol{s}_2$, $\boldsymbol{\Omega} = (\boldsymbol{\Omega}_1 + \boldsymbol{\Omega}_2)/2$, and $\Delta\boldsymbol{\Omega} = (\boldsymbol{\Omega}_1 - \boldsymbol{\Omega}_2)/2$. In the lowest order in $J/(\hbar\gamma)$ the term with $\boldsymbol{s}_1\times\boldsymbol{s}_2$ can be neglected, while $\Delta\boldsymbol{S}$ in the second equation quickly relaxes to the value

$$\Delta\boldsymbol{S} = \frac{\Delta\boldsymbol{\Omega}\times\boldsymbol{S}}{\gamma}. \tag{32}$$

From Eq. (31a) one can see that $\boldsymbol{S}$ precesses around $\boldsymbol{\Omega}$, while its projection on $\boldsymbol{\Omega}$ decays due to the second term. Therefore, one can solve separately the equations for these two components and find

$$\langle\boldsymbol{S}(t)\rangle = \boldsymbol{S}(0)\Big\langle \sin^2(\theta)\cos(\Omega t) + \cos^2(\theta)\exp\left(-\frac{\Delta\Omega^2\sin^2(\theta')}{\gamma}t\right)\Big\rangle, \tag{33}$$

where θ is the angle between $\boldsymbol{\Omega}$ and $\boldsymbol{S}(0)$ and θ' is the angle between $\Delta\boldsymbol{\Omega}$ and $\boldsymbol{\Omega}$.

The frequencies $\boldsymbol{\Omega}$ and $\Delta\boldsymbol{\Omega}$ are normally distributed, similar to Eq. (12), but with δ being $\sqrt{2}$ times smaller. This allows us to find

$$\langle\boldsymbol{S}(t)\rangle = \boldsymbol{S}(0)\frac{2}{3}\left[\left(1-\frac{(\delta t)^2}{4}\right)e^{-(\delta t)^2/8} + \frac{\gamma}{2\gamma+\delta^2 t}\right]. \tag{34}$$

This expression is plotted in Fig. 3(b). One can see that it is very similar to Fig. 3(a) despite the opposite relation between the parameters. At long timescales the spin polarization decays as

$$\langle\boldsymbol{S}(t)\rangle = \boldsymbol{S}(0)\frac{2\gamma}{3\delta^2 t}, \tag{35}$$

in agreement with the general result of the previous section.

Like in Sec. IV A, the spin noise spectrum can be derived by simply performing the Fourier transform of Eq. (34):

$$\left(S_z^2\right)_\omega = \frac{\sqrt{2}}{\delta} f\left(\frac{\sqrt{2}\omega}{\delta}\right) + \frac{\gamma}{\delta^2} h\left(\frac{2\gamma\omega}{\delta^2}\right), \tag{36}$$

where we introduced

$$h(x) = \tfrac{2}{9}\{\sin(|x|)[\pi - 2\,\mathrm{Si}(|x|)] - 2\cos(x)\,\mathrm{Ci}(x)\}, \tag{37}$$

with $\mathrm{Si}(x)$ and $\mathrm{Ci}(x)$ being the sine and cosine integral functions, respectively. This expression is shown by the red dashed curve in Fig. 4 and agrees with numerical calculations. At low frequencies one finds

$$\left(S_z^2\right)_\omega = \frac{4\gamma}{9\delta^2} \ln\left(\frac{\delta^2}{\gamma\omega}\right), \tag{38}$$

so the spectrum again diverges logarithmically.

C. Strong exchange interaction

In the limit $J/\hbar \gg \delta, \gamma$ the spins strongly couple into the triplet and singlet. The hyperfine interaction weakly mixes these states, while the electron hopping is possible only in the singlet state because of the Pauli spin blockade.

The equations for spin dynamics in this limit can be obtained from Eqs. (13) in the lowest order in $\hbar\boldsymbol{\Omega}_{1,2}/J$. Note that the terms containing γ again cannot be neglected because they lead to the complete spin decay at long timescales. As a result we obtain

$$\dot{\boldsymbol{S}} = \boldsymbol{\Omega} \times \boldsymbol{S} + \Delta\boldsymbol{\Omega} \times \Delta\boldsymbol{S}, \tag{39a}$$

$$\Delta\dot{\boldsymbol{S}} = \Delta\boldsymbol{\Omega} \times \boldsymbol{S} - \gamma\Delta\boldsymbol{S} - 2\frac{J}{\hbar}\boldsymbol{s}_1 \times \boldsymbol{s}_2, \tag{39b}$$

$$(\boldsymbol{s}_1 \times \boldsymbol{s}_2)^{\cdot} = \frac{J}{2\hbar}\Delta\boldsymbol{S} - \gamma\boldsymbol{s}_1 \times \boldsymbol{s}_2. \tag{39c}$$

One can see that $\Delta\boldsymbol{S}$ and $\boldsymbol{s}_1 \times \boldsymbol{s}_2$ decay much faster than $\boldsymbol{S}$, so the time derivatives in the second and third equations can be set to zero. This gives

$$\boldsymbol{s}_1 \times \boldsymbol{s}_2 = \frac{J}{2\hbar\gamma}\Delta\boldsymbol{S}, \tag{40a}$$

$$\Delta\boldsymbol{S} = \frac{\hbar^2\gamma}{J^2}\Delta\boldsymbol{\Omega} \times \boldsymbol{S}. \tag{40b}$$

Substituting the last expression in Eq. (39a) and averaging the solution over the nuclear fields, we find

$$\langle \boldsymbol{S}(t)\rangle = \boldsymbol{S}(0)\frac{2}{3}\left[\left(1 - \frac{(\delta t)^2}{4}\right)e^{-(\delta t)^2/8} + \frac{J^2}{2J^2 + \hbar^2\gamma\delta^2 t}\right]. \tag{41}$$

This expression is shown in Fig. 3(c), and one can see that the spin polarization in this case decays particularly slowly. Indeed, at long timescales one finds

$$\langle \boldsymbol{S}(t)\rangle = \boldsymbol{S}(0)\frac{2J^2}{3\hbar^2\gamma\delta^2 t}, \tag{42}$$

so the prefactor of $1/t$ is parametrically large in the limit under study, $J/\hbar \gg \delta, \gamma$.

The spin noise spectrum in this limit again can be calculated like in Sec. IV A, and the result reads

$$\left(S_z^2\right)_\omega = \frac{\sqrt{2}}{\delta} f\left(\frac{\sqrt{2}\omega}{\delta}\right) + \frac{J^2}{\hbar^2\gamma\delta^2} h\left(\frac{2J^2\omega}{\hbar^2\gamma\delta^2}\right). \tag{43}$$

This expression is shown by the black dashed curve in Fig. 4 and again agrees with the numerical calculations. At low frequencies one finds

$$\left(S_z^2\right)_\omega = \frac{4J^2}{9\hbar^2\gamma\delta^2} \ln\left(\frac{\hbar^2\gamma\delta^2}{J^2\omega}\right), \tag{44}$$

which shows once again that the spin correlations decay particularly slowly in this limit.

V. DISCUSSION

Despite the universality of the power law spin relaxation, the spin dynamics at short timescales depends on the relation between the system parameters. This is most clearly seen for the spin noise spectra in Fig. 4. In particular, the maximum shifts from the frequency δ to $\delta/\sqrt{2}$ with an increase of the exchange interaction constant [21]. This effect can be observed for the gate-defined double QD, where the exchange interaction can be tuned electrically.

At long timescales we demonstrated that the spin polarization decays as $\propto 1/t$ for any relation between the system parameters. Let us discuss the applicability limits of our model. In typical GaAs-based self-assembled QDs the short timescale is defined by $1/\delta \sim 1$ ns [43,44]. At longer timescales a few mechanisms of the spin relaxation can come into play, which can limit the applicability of our model.

At zero magnetic field the on-site electron spin flip-flops because the electron-phonon and spin-orbit interactions have very low rates because of, e.g., a zero phonon density of states with zero energy. Indeed, according to Refs. [45–50], the spin relaxation caused by the direct spin-phonon coupling [51,52] or spin admixture mechanisms [51,53] should exceed 1 s at a magnetic field smaller than 1 T.

The spin-orbit interaction during the hops leads to the spin rotations [54,55], which is not taken into account by our model. However, this effect can be simply accounted for by the rotation of the coordinate frames for the two QDs in spin space [20]. Still, the small random deviations of the electron hopping trajectory from the semiclassical one [56] cannot be compensated in the same way.

Another possible effect disregarded in this work is the hopping to a third remote localized state (QD) in the vicinity of the double QD [57]. This state can be either empty or occupied by an electron. In the former case the small tunneling rate to the empty state quenches the $1/t$ asymptote. In the latter case the spin blockade effect can also take place. Our estimations show that the configurations where the Overhauser fields in all three QDs are nearly parallel lead to the asymptote $1/t^2$ at timescales longer than the tunneling time to the third QD. Moreover, one can expect that the spin polarization of N electrons in N QDs decays as $1/t^{N-1}$, but this conjecture calls for a separate study.

The most probable limitations of our model are the external excitation of the system, e.g., by optical pulses [58,59], and the nuclear spin dynamics. The nuclear spin precession can be caused either by the strain in the QDs and the quadrupole interaction or by the Knight field created by electrons. These effects take place at the microsecond timescales [60,61] and quench $\propto 1/t$ asymptotic behavior. Nevertheless, we assume that our theory will correctly describe the spin dynamics in a double QD on submicrosecond timescales. In particular, from a few nanoseconds to a few microseconds, the spin relaxation should be described by the power law, which is the three orders of magnitude in the time and frequency domains.

VI. CONCLUSION

To summarize, we studied the spin dynamics in a double QD, taking into account the interplay between the hyperfine interaction, exchange interaction, and electron hopping. We demonstrated numerically that for an arbitrary relation between the system parameters the spin relaxation consists of the partial spin dephasing in a random nuclear field and a universal power law decay $\propto 1/t$ at large timescales. The spin noise spectrum of the system similarly consists of the two contributions and diverges as $\propto \ln(1/\omega)$ at low frequencies. We proved our results analytically in the limits when one of the system parameters exceeds the others by far. Due to the universality, we believe that these unusual asymptotes can be easily observed experimentally.

ACKNOWLEDGMENTS

We gratefully acknowledge the fruitful discussions with M. M. Glazov. All numerical calculations were performed under financial support from the Russian Science Foundation (RSF Grant No. 18-72-10002). D.S.S. was partially supported by the RF President Grant No. MK-1576.2019.2, Russian Science Foundation Grant No. 19-12-00051, and the Basis Foundation.

APPENDIX: ALTERNATIVE DERIVATION OF THE HOPPING RATES

Here we justify our approach based on the Hamiltonian of the electron-phonon interaction (5) [32]. To do this we start from the usual Hamiltonian

$$\mathcal{H}_{\rm tot}^{(0)} = \mathcal{H}_0 + \mathcal{H}_{ph} + \mathcal{H}_{e-ph}. \tag{A1}$$

Here the electron Hamiltonian is

$$\mathcal{H}_0 = \sum_{i,\sigma} E_i n_i^\sigma + \sum_i U_i n_i^+ n_i^- + J\mathbf{s}_1\mathbf{s}_2 + V\sum_\sigma (c_{1\sigma}^\dagger c_{2\sigma} + c_{2\sigma}^\dagger c_{1\sigma}), \tag{A2}$$

where we neglect the hyperfine interaction. The phonon Hamiltonian is given by Eq. (4). The electron-phonon interaction Hamiltonian is

$$\mathcal{V} = \sum_{i,\sigma,q} \upsilon_q (e^{i\boldsymbol{q}\boldsymbol{R}_i} b_{\boldsymbol{q}} + e^{-i\boldsymbol{q}\boldsymbol{R}_i} b_{\boldsymbol{q}}^\dagger) n_{i\sigma}. \tag{A3}$$

It is convenient to analyze the eigenstates of the Hamiltonian $\mathcal{H}_0$ in the basis of states $|T_m\rangle$, $|S\rangle$, $|S_i\rangle$, where $m = 0, \pm$ denotes the projection of the total spin of the triplet state on the z axis, $|S\rangle$ is the singlet state of the two electrons in different QDs, and $|S_i\rangle$ are the singlet states of the two electrons in QD i. In this basis one has

$$\begin{aligned}
\mathcal{H}_0|T\rangle &= \left(E_1 + E_2 + \frac{1}{4}J\right)|T\rangle,\\
\mathcal{H}_0|S\rangle &= \left(E_1 + E_2 - \frac{3}{4}J\right)|S\rangle + \sqrt{2}V(|S_1\rangle + |S_2\rangle),\\
\mathcal{H}_0|S_i\rangle &= (2E_i + U_i)|S_i\rangle + \sqrt{2}V|S\rangle.
\end{aligned} \tag{A4}$$

In the first order in V the eigenstates of $\mathcal{H}_0$ are $|T_m\rangle$ and

$$\begin{aligned}
|\tilde{S}\rangle &= |S\rangle - \frac{\sqrt{2}V}{E_{12}}|S_1\rangle - \frac{\sqrt{2}V}{E_{21}}|S_2\rangle,\\
|\tilde{S}_i\rangle &= |S_i\rangle + \frac{\sqrt{2}V}{E_{i\bar{i}}}|S\rangle,
\end{aligned} \tag{A5}$$

where $E_{i\bar{i}} = U_{\rm i} + E_{\rm i} - E_{\bar{i}}$ and we neglect J compared with $E_{i\bar{i}}$. The matrix elements of $n_i = n_i^+ + n_i^-$ between these states are

$$\langle\tilde{S}_i|n_i|\tilde{S}\rangle = -\frac{\sqrt{2}V}{E_{12}}, \quad \langle\tilde{S}_i|n_{\bar{i}}|\tilde{S}\rangle = \frac{\sqrt{2}V}{E_{12}}. \tag{A6}$$

Then from Fermi's golden rule we find the hopping rates from state $|\tilde{S}\rangle$ to $|\tilde{S}_i\rangle$,

$$\frac{2\pi}{\hbar}\frac{4V^2}{E_{i\bar{i}}^2}\upsilon_q^2 N_{E_{i\bar{i}}} D(E_{i\bar{i}}) = 2\gamma_{i\bar{i}}, \tag{A7}$$

which agree with Eqs. (8) and (14) since $\Delta E = E_{i\bar{i}}$. In the same way we find that the hopping rates from $|\tilde{S}_i\rangle$ to $|\tilde{S}\rangle$ equal $2\Gamma_{\bar{i}i}$. Thus, the hopping Hamiltonian (5) yields the correct hopping rates.

We note also that in the second order of the perturbation theory the energy correction to state $|S\rangle$ is $-2V^2/E_{12} - 2V^2/E_{21}$, which can be included in the exchange interaction constant J, as is usually assumed.

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