

Nonperturbative Master Equation Solution of Central Spin Dephasing Dynamics

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We solve the long-standing central spin problem for a general set of inhomogeneous bath couplings and a large class of initial bath states. We compute the time evolution of the coherence of a central spin coupled to a spin bath by resumming all orders of the time-convolutionless master equation, thus avoiding the need to assume weak coupling to the bath. The fully quantum, non-Markovian solution is obtained in the large-bath limit and is valid up to a time scale set by the largest coupling constant. Our result captures the full decoherence of an electron spin qubit coupled to a nuclear spin bath in a GaAs quantum dot for experimentally relevant parameters. In addition, our solution is quite compact and can readily be used to make quantitative predictions for the decoherence process and to guide the design of nuclear state preparation protocols.

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Since the central spin model was first introduced by Gaudin [1] several decades ago, it has appeared in diverse physical settings such as spin dynamics in disordered insulators [2], interacting electrons in metallic grains [3], and more recently in semiconductor spin qubits [4–13]. It has also been widely studied in the context of integrable systems, where close connections to BCS theory and related theories of pairing forces have been discovered [3,14]. In addition, several variants of the central spin model have served as toy models for comparing and contrasting various master equation formalisms [15–17].

This broad interest in the central spin model has led to a plethora of disparate approaches to computing its dynamics. Since its inception, it has been recognized as an integrable system which admits a solution via Bethe ansatz [1,3,8]. However, this yields only a very complicated ground state wave function, and progress in extracting dynamical information for the central spin has been limited; to date only modest bath sizes of up to 30 bath spins have been treated in this manner [8]. Sophisticated numerical recipes for calculating the central spin evolution have been developed [5,18,19], but success tends to require either small baths (tens of spins) or the assumption that the initial bath state is completely unpolarized. These requirements are too restrictive for many applications in the context of III–V semiconductor spin qubits, where the number of nuclear spins ranges from 10^4 to 10^6 , and polarized baths are employed to facilitate qubit operations and extend coherence times [20,21].

This state of affairs led to the development of several approximate analytical methods to compute the central spin evolution. Some of these methods employ an effective pure-dephasing Hamiltonian which is derived perturbatively from the central spin Hamiltonian using a canonical transformation [7,9,10]. This approach culminated with a nonperturbative solution of the effective Hamiltonian

dynamics [9,10] and has been successful in describing spin echo experiments in the case of unpolarized baths [22]. However, an uncontrolled approximation in the effective-Hamiltonian derivation makes it unclear when this approach is valid, and it has yet to be extended to more general bath states. Methods employing generalized master equations, on the other hand, offer a controlled approximation and naturally describe polarized bath states, but only perturbative treatments have been given so far [6,11,13,16,17], leading to solutions which are valid only outside the regime relevant for many semiconductor spin qubit experiments [22–25].

In this Letter, we solve the central spin problem using the time-convolutionless (TCL) master equation for a general set of inhomogeneous coupling constants and a large set of initial bath states, including both polarized and unpolarized baths. The TCL equation is an exact equation for the reduced density matrix of a system coupled to a bath; although this equation is time local, it incorporates the full bath dynamics [16,17,26]. With only a very modest condition on the magnetic field, we give a closed-form solution describing the evolution up to a time scale set by the largest bath coupling. We are therefore presenting an exact solution to the central spin model as it pertains to gated GaAs spin qubits since this temporal window contains the entire decay of the electron spin coherence in the low magnetic field regime where the central spin model gives a good description of the physics [27]. Although we will focus on the example of spin qubits, our results are potentially applicable to any Gaudin-type central spin problem. This result is also important to the general study of open quantum systems; we are not aware of other examples involving a large, nontrivial and highly non-Markovian bath where an all-orders resummation of a master equation expansion is performed.

The central spin model is comprised of a central spin coupled to a spin bath via a Heisenberg interaction. Assuming a nonzero external magnetic field, the Hamiltonian is

$$\hat{H} = \sum_k A_k \mathbf{S} \cdot \mathbf{I}_k + \Omega S^z + \sum_k \omega_k I_k^z, \quad (1)$$

with \mathbf{S} denoting the central spin operator, \mathbf{I}_k the bath spins, A_k the (hyperfine) couplings, and Ω and ω_k the central spin and bath spin Zeeman energies, respectively. In the context of semiconductor electron spin qubits, the A_k are determined by the shape of the electron wave function envelope, but we will leave the A_k completely arbitrary. We refer to $\sum_k A_k S^z I_k^z$ as the Overhauser term, and $V_{\text{ff}} = \frac{1}{2} \sum_k A_k (S^+ I_k^- + S^- I_k^+)$ as the flip-flop term, where $S^\pm = S^x \pm iS^y$, and similarly for I_k^\pm . The total interaction energy is $\mathcal{A} \equiv \sum_k A_k$, and the number of bath spins appreciably interacting with the central spin is $N \equiv \mathcal{A}^2 / (\sum_k A_k^2)$. Roughly speaking, the bath produces an effective (Overhauser) magnetic field, the magnitude of which is controlled by \mathcal{A} and the bath polarization, and about which the central spin precesses, while the A_k set the scale for the precession of individual bath spins about the central spin.

The TCL equation is an exact equation for the time evolution of the reduced density matrix of a system coupled to a bath [26]. Although this equation contains full memory of the bath dynamics, unlike equations such as the Nakajima-Zwanzig equation [6,11,13], it has the attractive feature that it is a time-local ordinary differential equation. Working in an interaction picture defined with respect to $\hat{H}_0 = \sum_k A_k S^z I_k^z + \Omega S^z + \sum_k \omega_k I_k^z$ and denoting the total density matrix in the interaction picture by $\rho(t)$, the TCL equation has the form

$$\frac{d}{dt} P\rho(t) = \sum_{n=1}^{\infty} \mathcal{K}_n(t) P\rho(t). \quad (2)$$

The operator P projects the full density matrix onto the reduced density matrix of the system; its precise definition will be given shortly. Equation (2) is defined in terms of a perturbative expansion in V_{ff} which is nominally controlled by the quantity \mathcal{A}/Ω . Ultimately our solution will not require this expansion to be convergent since we will sum the entire series, so \mathcal{A}/Ω need not be small. The n th-order kernel $\mathcal{K}_n(t)$ encapsulates full bath effects arising from n th-order flip-flop processes in which the central spin flips n times with one or more bath spins. It can be expressed as an integral of ordered cumulants involving P and the interaction picture Liouville operator L (defined by $\dot{\rho} = -iL\rho$); for example, the second-order kernel is $\mathcal{K}_2(t) = -\int_0^t dt' PL(t)L(t')P$. The rules for constructing the higher order kernels can be found in Ref. [26].

We will assume that the initial density matrix separates into system and bath components, $\rho(0) = \rho_S(0) \otimes \rho_B(0)$, and we will use a set of ‘‘correlated projectors’’ [16], in

which case the action of P on a matrix M is $PM = \sum_\alpha \text{Tr}_B\{\Pi_\alpha M\} \otimes \frac{1}{\mathcal{N}_\alpha} \Pi_\alpha$, where the Π_α are a set of bath projectors satisfying $\Pi_\alpha \Pi_\beta = \delta_{\alpha\beta} \Pi_\beta$ and $\sum_\alpha \Pi_\alpha = 1$, and $\mathcal{N}_\alpha = \text{Tr}_B\{\Pi_\alpha\}$. The Π_α allow us to write the reduced density matrix of the system as a sum of independent degrees of freedom: $\rho_S = \sum_\alpha \text{Tr}_B\{\Pi_\alpha \rho\} \equiv \sum_\alpha \rho_S^{(\alpha)}$. The form of the TCL equation we use requires $P\rho(0) = \rho(0)$, which in turn implies that the choice of the Π_α will constrain the possible initial bath states. An appropriate choice of the Π_α can either simplify or vastly improve the convergence of the TCL equation depending on the symmetries of the Hamiltonian and initial bath state [13,16]; for now we leave the Π_α completely general.

Since we want to compute the off-diagonal component of $\rho_S(t)$ (coherence function), we multiply Eq. (2) by S^+ and trace over both system and bath to obtain

$$\dot{\rho}_{S,-+}(t) = \text{Tr}_S\{S^+ \dot{\rho}_S(t)\} = \sum_{n=1}^{\infty} \text{Tr}\{S^+ \mathcal{K}_n(t) P\rho(t)\}. \quad (3)$$

The summand on the right-hand side of this equation is comprised of integrals of terms with the general structure $\text{Tr}\{S^+ \mathcal{L}_1 P \mathcal{L}_2 P \dots \mathcal{L}_r P \rho(t)\}$, where \mathcal{L}_i represents a string of Liouville operators $L(t_{i_1})L(t_{i_2})\dots$. If one assumes that Π_α is such that bath correlators of the type $\text{Tr}_B\{\Pi_\alpha I_{\ell_1}^\pm I_{\ell_2}^\pm \dots\}$ vanish unless they contain equal numbers of raising and lowering operators, then it is straightforward to show [29] that in the case of the central spin model (1), such terms factorize:

$$\begin{aligned} & \text{Tr}\{S^+ \mathcal{L}_1 P \mathcal{L}_2 P \dots \mathcal{L}_r P \rho(t)\} \\ &= \sum_\alpha \frac{1}{\mathcal{N}_\alpha^r} \rho_{S,-+}^{(\alpha)}(t) \prod_{i=1}^r \text{Tr}\{S^+ \mathcal{L}_i S^- \Pi_\alpha\}. \end{aligned} \quad (4)$$

Since every term on the right-hand side of Eq. (3) factorizes in this way, we can expand the left-hand side as $\dot{\rho}_{S,-+}(t) = \sum_\alpha \dot{\rho}_{S,-+}^{(\alpha)}(t)$ and separately equate each term of the α -sum. The resulting set of equations is readily solved:

$$\rho_{S,-+}^{(\alpha)}(t) = \rho_{S,-+}^{(\alpha)}(0) \exp\left\{\sum_n \mathcal{G}_n^{(\alpha)}(t)\right\}, \quad (5)$$

where $\mathcal{G}_n^{(\alpha)}(t) \equiv \int_0^t dt' \text{Tr}\{S^+ \mathcal{K}_n(t') S^- \Pi_\alpha\}$.

To calculate $\mathcal{G}_n^{(\alpha)}(t)$ we must first compute correlators of the type $\text{Tr}\{S^+ \mathcal{L} S^- \Pi_\alpha\}$ where \mathcal{L} is an arbitrarily long string of Liouville operators. Restricting ourselves for simplicity to the case $\omega_k = \omega$, we find [29]

$$\begin{aligned} & \text{Tr}\{S^+ L(t_{b_1}) \dots L(t_{b_q}) S^- \Pi_\alpha\} \\ & \approx \frac{1}{4^q} e^{-i\Omega_\alpha \sum_{i=1}^{2q} (-1)^{b_i} t_{b_i}} \sum_{k=0}^q \binom{q}{k} \text{Tr}_B\{(h^+ h^-)^k \Pi_\alpha (h^- h^+)^{q-k}\}. \end{aligned} \quad (6)$$

We have defined the operators $h^j \equiv \sum_\ell A_\ell I_\ell^j$ and frequency $\Omega_\alpha \equiv \Omega - \omega + \text{Tr}_B\{h^z \Pi_\alpha\} / \mathcal{N}_\alpha$, and we have assumed that $[h^z, \Pi_\alpha] = 0$. The latter condition ensures that the correlator vanishes for odd numbers of Liouville operators, which in turn implies that $\mathcal{G}_n^{(\alpha)}$ vanishes for odd n . There are two approximations being made in Eq. (6). The first approximation assumes $t \ll 1/A_{\max}$, where $A_{\max} \sim \mathcal{A}/N$ is the largest coupling, and this leads to the time dependence appearing only as a phase factor in Eq. (6). For spin qubits in GaAs with $N = 10^6$, $1/A_{\max}$ can be on the order of 10 μs , long enough to capture the full decay of the electron spin coherence for experimentally relevant values of the magnetic field [22–25]. For comparison, we can also consider spin qubits in Si [30], in which case $1/A_{\max}$ is on the order of 250 μs .

If this were the only approximation, then the right-hand side of Eq. (6) would have an additional sum over permutations of the t_{b_i} , but only a certain subset of these permutations were kept in Eq. (6). Retaining only this subset amounts to keeping the leading order terms in the $\Omega_\alpha t \gg 1$ limit at each order of the TCL expansion. To illustrate the nature of this RPA-like approximation, we consider its effect on the lowest-order terms. If we kept all permutations, then the second-order term would have the form $\mathcal{G}_2^{(\alpha)}(t) \sim \delta_\alpha^2 u(t, \Omega_\alpha)$ while the fourth-order terms stemming from the $q = 2, k = 0, 2$ cases in (6) would have the form $\mathcal{G}_4^{(\alpha)}(t) \sim \delta_\alpha^4 v(t, \Omega_\alpha)$ where $\delta_\alpha \equiv \frac{\mathcal{A}}{\sqrt{N}\Omega_\alpha}$ and $u(t, \Omega_\alpha) = i\Omega_\alpha t + e^{-i\Omega_\alpha t} - 1$, $v(t, \Omega_\alpha) = -\Omega_\alpha^2 t^2 - 4i\Omega_\alpha t - (6 + 2i\Omega_\alpha t)e^{-i\Omega_\alpha t} + 6$. The RPA-like approximation amounts to taking $u \rightarrow i\Omega_\alpha t$ and $v \rightarrow -\Omega_\alpha^2 t^2$. Self-consistency of the approximation requires $\delta_\alpha \ll 1$, which imposes a lower bound on the magnetic field (corresponding to a few mT for gated dots in GaAs). The necessity for $\delta_\alpha \ll 1$ can be seen from u and v by noting that we can only neglect the linear (in t) term in v if it is small compared to the linear term in u . Later on, we will see that this approximation captures the envelope of the coherence function, and we will also find that we can relax this approximation to a large degree by keeping the full form of $\mathcal{G}_2^{(\alpha)}(t)$.

With Eq. (6) in hand, it is straightforward to assemble these correlators into the function $\mathcal{G}_n^{(\alpha)}(t)$ using the rules for constructing the TCL ordered cumulants [29]:

$$\mathcal{G}_n^{(\alpha)}(t) = \left(\frac{it}{4\Omega_\alpha}\right)^{n/2} \sum_{\{q_i\} \in \mathcal{P}(n/2)} \frac{1}{\prod_{i=1}^r q_i!} \frac{(-1)^{r+1}}{r \mathcal{N}_\alpha^r} \times \prod_{i=1}^r \sum_{k=0}^{q_i} \binom{q_i}{k} \text{Tr}_B\{(h^+ h^-)^k \Pi_\alpha (h^- h^+)^{q_i-k}\}. \quad (7)$$

In this expression, $\{q_i\} \in \mathcal{P}(n/2)$ means that $\{q_i\}$ is an ordered integer partition of $n/2$, with r being the number of q_i comprising the partition [31]. To evaluate

this expression, we make perhaps the simplest choice for the projectors: $\Pi_\alpha = \Pi_\chi = |\chi\rangle\langle\chi|$, where $|\chi\rangle \equiv \bigotimes_k |I_k, m_k^\chi\rangle$ is a product of eigenstates of the I_k^z [$I_k(I_k + 1)$] and m_k^χ are eigenvalues of I_k^z and I_k^z . In this case, $\mathcal{N}_\alpha = 1$. This particular choice is well suited to applications pertaining to spin qubits [6,11,13]. With an initial bath density matrix of the form $\rho_B(0) = \sum_\chi \rho_{\chi\chi} |\chi\rangle\langle\chi|$, it is possible to perform the various sums in (7) despite their complexity [29], and we find in the limit of large N that the coherence function $W(t) \equiv \tilde{\rho}_{S,-+}(t)/\tilde{\rho}_{S,-+}(0)$ in the Schrödinger picture is

$$W(t) = \sum_\chi \frac{\rho_{\chi\chi} (d_\chi^+ - d_\chi^-) e^{i(\Omega + h_\chi^z)t}}{d_\chi^+ e^{-i(d_\chi^+ - d_\chi^-)/4\Omega_\chi} - d_\chi^- e^{i(d_\chi^+ - d_\chi^-)/4\Omega_\chi}}, \quad (8)$$

where $h_\chi^z \equiv \text{Tr}_B\{\Pi_\chi h^z\}$ is the Overhauser field associated with the state $|\chi\rangle$, $d_\chi^\pm \equiv \langle\chi|h^\mp h^\pm|\chi\rangle = \sum_\ell A_\ell^2 [I_\ell(I_\ell + 1) - m_\ell^\chi(m_\ell^\chi \pm 1)]$ quantifies transverse fluctuations of this field, and $\Omega_\chi \equiv \Omega - \omega + h_\chi^z$ is the difference between the effective Zeeman energy of the central spin ($\Omega + h_\chi^z$) and the bath spin Zeeman energy (ω). Equation (8) is the main result of this Letter; it describes the envelope of the coherence function of the central spin for an arbitrary set of couplings A_k and for a large set of initial bath states up to time $t \lesssim 1/A_{\max}$.

As a first example, we consider the case of a uniformly polarized bath [32] with all bath spins having the same total angular momentum I , and for which h_χ^z is the same for all χ [6,11]. Writing $h_\chi^z = \mathcal{A}Ip$ and $\Omega_p = \Omega - \omega + \mathcal{A}Ip$, where $p = \frac{1}{N} \sum_\chi \rho_{\chi\chi} \sum_k m_k^\chi$ is the average polarization of the bath, with $p = 0$ denoting an unpolarized bath and $|p| = 1$ maximal polarization, we find

$$W(t) = \frac{p e^{i(\Omega + \mathcal{A}Ip)t}}{p \cos(\frac{2Ip t}{\tau_p}) - i p_\perp^2 \sin(\frac{2Ip t}{\tau_p})}, \quad (9)$$

with $p_\perp^2 \equiv I + 1 - \frac{1}{N} \sum_\chi \rho_{\chi\chi} \sum_k (m_k^\chi)^2$ (for $I = 1/2$, $p_\perp^2 = 1$), and $\tau_p \equiv 4N\Omega_p/\mathcal{A}^2$. Setting $p = 0$ in (9) yields the zero-polarization result, $W(t) = 1/(1 - 2iIp_\perp^2 t/\tau_p)$, obtained in previous works [9,10,13] using less rigorous

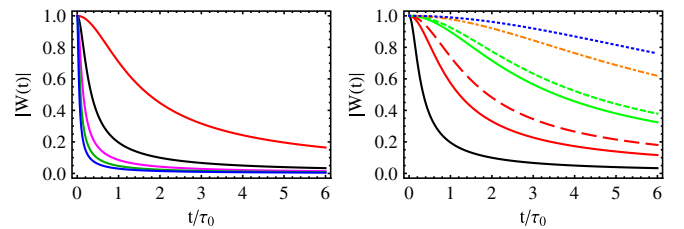


FIG. 1 (color online). Coherence function [Eq. (9)]. Left panel: changing bath total spin I with $p = 0$ and $p_\perp^2 = \frac{2}{3}(I + 1)$. From top to bottom, I takes the values $\frac{1}{2}, \frac{3}{2}, \frac{5}{2}, \frac{7}{2}, \frac{9}{2}$. Right panel: changing bath polarization p with $I = \frac{3}{2}$, $\mathcal{A}/\Omega = 30$, and $p_\perp^2 = \frac{5}{3}$. From top to bottom, p takes the values $\frac{3}{4}, \frac{1}{2}, \frac{1}{4}, -\frac{1}{4}, \frac{1}{10}, -\frac{1}{10}, 0$.

methods. The left panel of Fig. 1 shows that the decoherence rate increases with increasing I due to a corresponding increase in the number of bath degrees of freedom. For electron spin qubits in a GaAs nuclear spin bath ($I = 3/2$) where the magnetic field is typically on the order of 100 mT [23] so that $\mathcal{A}/\Omega \approx 30$ and $\tau_0 \approx (2/15)N/\mathcal{A}$, it is clear that the coherence decays almost completely before time $t = 1/A_{\max} \approx N/\mathcal{A}$ is reached. The right panel of Fig. 1 depicts the extent to which the decoherence time increases with increasing bath polarization, a well known effect which can be understood in terms of a reduction of phase space for flip-flops. It is also evident that positive net polarization leads to longer decoherence times compared with negative polarization, since for the latter the decrease of $|\Omega_p|$ facilitates virtual flip-flops. Our results provide quantitative predictions for the enhancement of decoherence time resulting from bath polarization.

A salient feature of the uniform-polarization result, Eq. (9), is that it depends on the couplings only through the quantities \mathcal{A} and N . This implies that any set of couplings which yield the same values of \mathcal{A} and N will give rise to the same central spin evolution for $t \ll N/\mathcal{A}$. For example, this evolution should be reproduced by a model in which all the couplings are equal, $A_k = \mathcal{A}/N$, the so-called “box” model, which is exactly solvable (see, e.g., Ref. [13] for the solution in the case of a polarized bath). A comparison of Eq. (9) with the exact box model solution is shown in the left panel of Fig. 2, and it is evident that the two solutions agree very well. This insensitivity to the particular values of the A_k on time scales $t \ll N/\mathcal{A}$ was anticipated in Ref. [13] based on energy-time uncertainty; here, we have given a direct proof of this result, and we show below that its validity requires uniform polarization. The exact box model solution is not known to have a closed form, so the fact that Eq. (9) constitutes a very good approximate closed-form solution is an added bonus of the present Letter [Eq. (8)].

The right panel of Fig. 2 reveals that Eq. (9) does not capture a high-frequency, small-amplitude modulation exhibited by the exact box model solution [13]. However, it turns out that it is easy to correct for this by relaxing the RPA-like approximation at the second order of the TCL

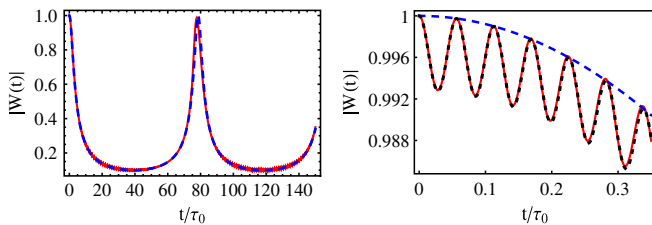


FIG. 2 (color online). Left panel: Coherence function from Eq. (9) (blue, dashed) vs exact box model solution from Ref. [13] (red, solid) with $I = \frac{1}{2}$, $\mathcal{A}/\Omega = 30$, $N = 10^4$, $p = \frac{1}{10}$. Right panel: Zoom in of left panel with modified coherence function $\tilde{W}(t)$ (black, dotted) included as well.

flip-flop expansion, in which case one finds that $\mathcal{G}_2^{(\chi)} = \frac{1}{4\Omega_x^2} [i\Omega_x t + e^{-i\Omega_x t} - 1](d_\chi^+ + d_\chi^-)$, leading to the modified coherence function $\tilde{W} = W \exp[(e^{-i\Omega_p t} - 1)/(\Omega_p \tau_p)]$. This modified function reproduces very well the small modulations as is evident in the right panel of Fig. 2. The condition $\delta_\chi = \frac{\mathcal{A}}{\sqrt{N}\Omega_x} \ll 1$ ensures that higher-order corrections to these modulations are negligible.

We stress that the coherence function given in Eq. (8) can describe more general bath polarization states. To illustrate this, we consider a simple example of a nonuniformly polarized initial bath state where the bath spins possess an average polarization which depends linearly on their coupling: $\langle m_k \rangle = \sum_\chi \rho_{\chi\chi} m_k^\chi = NIpA_k/\mathcal{A}$. This describes a bath configuration where the spins closest to the central spin are more polarized, while those further away are less polarized. In the case of spin qubits in quantum dots, this is qualitatively a physically plausible configuration since the nuclear spins are typically polarized through manipulation of the central electron spin [20,21,33–36]. This time, the coherence function computed from Eq. (8) depends not only on $\sum_k A_k$ and $\sum_k A_k^2$, but also on $\sum_k A_k^3$, meaning that the result now depends on details of the distribution of the A_k . Assuming the A_k are distributed in accordance with a two-dimensional Gaussian wave function of the central spin electron, and taking the continuum limit (e.g., $\sum_k f(A_k) \rightarrow \int_0^{A_{\max}} \rho(A)f(A)dA$ with $\rho(A) = \frac{N}{2A}$ and $A_{\max} = \frac{2\mathcal{A}}{N}$), we obtain

$$W(t) = \frac{pe^{i(\Omega + \mathcal{A}Ip)t}}{p \cos(\frac{8Ip t}{3\tau_p}) - \frac{3i}{4} p_\perp^2 \sin(\frac{8Ip t}{3\tau_p})}. \quad (10)$$

Figure 3 shows that while for lower bath polarizations this result is similar to what we found in the uniform-polarization case, for larger polarizations, the differences between the two solutions become quite pronounced. This illustrates how our results can be used to distinguish between different narrowed polarization distributions produced using empirical nuclear state preparation schemes [20,21,34,35] which are not yet understood microscopically.

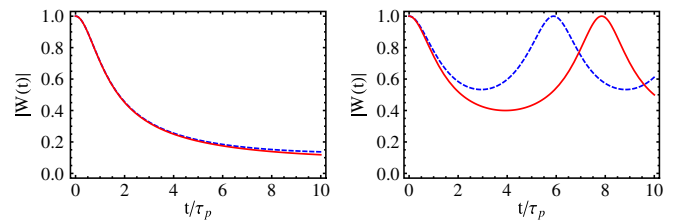


FIG. 3 (color online). Coherence function with uniform polarization Eq. (9) (red, solid) vs nonuniform polarization Eq. (10) (blue, dashed) with $I = \frac{1}{2}$, $\mathcal{A}/\Omega = 30$, and (left panel) $p = \frac{1}{10}$, $\tau_p = \frac{1}{3} \frac{N}{\mathcal{A}}$, (right panel) $p = \frac{2}{5}$, $\tau_p = \frac{14}{15} \frac{N}{\mathcal{A}}$.

In conclusion, we have presented a nonperturbative solution to the central spin problem. Our solution is an elegant, closed-form expression which is valid for an arbitrary set of couplings to the spin bath and for a large class of initial bath states. In the context of GaAs spin qubits, our result solves completely the pure-hyperfine decoherence problem and can be used to quantitatively predict decoherence times and to better understand nuclear state preparation protocols.

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