Maximizing the purity of a qubit evolving in an anisotropic environment

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We provide a general method to calculate the purity of a qubit interacting with an anisotropic non-Markovian environment. Counter to intuition, we find that the purity is often maximized by preparing and storing the qubit in a superposition of non-interacting eigenstates. For a model relevant to decoherence of a heavy-hole spin qubit in a quantum dot, we show that preparation of the qubit in the Zeeman ground state can actually be the worst choice to maximize purity. We further give analytical results for spin-echo envelope modulations of arbitrary spin components of a hole spin in a quantum dot, going beyond a standard secular approximation. Finally, we account for general dynamics in the presence of a pure-dephasing process and identify a crossover timescale at which it is again advantageous to initialize the qubit in the non-interacting ground state.

I. INTRODUCTION

A source of high-quality pure ancilla qubits is an essential element in a wide variety of applications in quantum information science. Pure ancillas are required to introduce redundancy into quantum error-correcting codes, 10 for the preparation of Greenberger-Horne-Zeilinger (GHZ) states for quantum-enhanced precision measurements, 26 as a low-entropy resource for algorithmic cooling, 43, 44 or direct measurement of the bath state, 19, 21, 24 so studying the purity for these systems is especially important from both a practical and a fundamental point of view.

As we show below, the evolution of qubit purity becomes highly nontrivial for a qubit interacting with a slow anisotropic environment. Anisotropic hyperfine couplings between a central qubit spin and environmental spins are important for nitrogen-vacancy centers in diamond, 35, 27 electrons bound to phosphorus donor impurities in silicon, 28, 29 electrons in graphene or carbon nanotubes, 30, 31 and especially for hole spins in III-V semiconductors or silicon, 32, 33 Heavy-hole spins can indeed approach the extreme-anisotropic limit of a pure Ising-like coupling to nuclear spins. 34

Coherence properties of single hole spins in quantum dots have been probed in detail only relatively recently. 36, 50 Measurements of a coherent-population-trapping effect 41, 45 have suggested long hole-spin coherence times, > 100 ns. These measurements have been supported by time-domain studies for single-hole spin echoes, 39, 10 and mode-locking or spin-echo measurements for ensembles. 41, 12 Alternative measurements of hole-spin dynamics have been performed through spin-noise spectroscopy, revealing a probable anisotropic decay of hole-spin coherence. 38, 39

In addition to optical coherent control of hole spins in self-assembled quantum dots, 37, 38, 39, 40 there are several suggestions for electrical manipulation of hole spins. 37, 40 Such electrical control has recently been demonstrated for hole spins in III-V nanowire quantum dots, 50 and coherence times have now been measured for hole spins in Ge-Si core-shell nanowire quantum dots. 31 The very recent achievement of the few-hole regime in lateral gated double-dot devices, 52 suggests that previous highly successful measurements performed for electron spins, 31, 53 can now be performed for hole spins, which show promise...
for much longer coherence times.\cite{32,34}

In the rest of this paper, we introduce a general method that can be used to calculate and enhance the purity of a qubit interacting with an anisotropic environment. We apply this method to the experimentally relevant problem of heavy-hole spin-echo dynamics. Counter to common intuition, we find that preparation of the hole spin in its Zeeman ground state can be the worst choice if the goal is to maximize purity. This surprising result is not limited to the problem of hole-spin echoes. On quite general grounds, the ideal choice to maximize purity will typically not be initialization in the eigenbasis of the isolated qubit Hamiltonian at sufficiently short time, and whenever pure-dephasing processes are weak or absent.

The rest of this paper is organized as follows. In Sec. II we review properties of the purity and derive general conditions to achieve the maximum purity at time $t$, starting with a factorized initial state of the spin and a generic environment. In Sec. III we calculate spin-echo dynamics for a heavy-hole spin in a quantum dot. In Sec. IV we give general conditions to maximize hole-spin purity in a spin-echo experiment. In Sec. V we consider the more general case of purity/coherence decay accounting for a pure-dephasing process in addition to anisotropic hyperfine coupling. We conclude in Sec. VI with a summary of the main results. Technical details are given in Appendices A-C.

II. QUBIT PURITY

Here we give a brief introduction to the key observable that we will evaluate, the qubit purity, $P(t)$. In addition to its importance for the preparation of high-quality ancillas in quantum error correction schemes,\cite{35} purity characterizes the ability to extract a finite qubit polarization after interacting with a bath for a time $t$. Maximizing the purity is essential for any scheme that aims to maximize the storage-and-retrieval fidelity of a qubit interacting with an uncontrolled environment.

The purity of a two-level system (a qubit) is generally defined as:\cite{22}

$$P(t) = \text{Tr} \left[ \rho_S^2(t) \right] = \frac{1}{2} + 2|\langle S(t) \rangle|^2. \quad (1)$$

Here, $\rho_S(t) = \frac{1}{2} \sigma_0 + \langle S(t) \rangle \cdot \sigma$ is the reduced density matrix of a qubit, where $\sigma_0$ is the identity and $\sigma$ is the vector of Pauli matrices. For a pure state of the qubit, the Bloch vector $|\langle S(t) \rangle\rangle$ lies on the surface of the Bloch sphere, $|\langle S(t) \rangle| = 1/2$, giving $P(t) = 1$, while a mixed state has $|\langle S(t) \rangle| < 1/2$ giving $P(t) < 1$. Understanding the dynamics of the length of the Bloch vector, $|\langle S(t) \rangle|$, therefore allows for a direct evaluation of the purity. $P(t)$.

In particular, we can establish a set of criteria that maximize the purity to avoid information loss.

Provided both a qubit and its environment are initially prepared in a pure state, entanglement between the qubit and environment can be characterized by the von Neumann (entanglement) entropy,

$$E[\rho_S] = -\text{Tr} \rho_S \log_2 \rho_S = -\sum_{s=\pm} p_s \log_2 p_s, \quad (2)$$

$$p_\pm = \frac{1}{2} (1 \pm |\langle S(t) \rangle|). \quad (3)$$

Here, $p_\pm$ give the eigenvalues of $\rho_S$. In this case, a reduction in the length of the Bloch vector $|\langle S(t) \rangle|$ characterizes a finite degree of entanglement, $E[\rho_S] \neq 0$.\cite{22} Provided the environment itself can be prepared in a pure state, the purity $P(t)$ is therefore also an important measure of non-classical evolution.

We assume the dynamics of $\langle S(t) \rangle$ are generated by a Hamiltonian

$$H = H_0 + V(t), \quad H_0 = H_S + H_E, \quad (4)$$

where here, the Hamiltonians $H_S$ and $H_E$ act only on the system and environment Hilbert spaces, respectively, and the (possibly time-dependent) perturbation $V(t)$ typically couples the two spaces. For factorized initial conditions $\rho(0) = \rho_S(0) \otimes \rho_E(0)$ with initial system (environment) density matrix $\rho_{SE}(0)$, we can write generally

$$\langle S(t) \rangle = \left\langle e^{-\frac{1}{2} L(t) \hat{S}(t)} \right\rangle_{S}. \quad (5)$$

Here, the interaction picture is defined (setting $\hbar = 1$) by

$$\hat{O}(t) = U_0(t) O U_0^\dagger(t), \quad U_0(t) = e^{i H_0 t}, \quad (6)$$

corresponding to an SO(3) rotation matrix $[R_0(t)]$ applied to the vector $S = (S_x, S_y, S_z)^T$:

$$\hat{S}(t) = U_0(t) S U_0^\dagger(t) = [R_0(t)] \cdot S. \quad (7)$$

We use the notation

$$\langle \cdot \cdot \cdot \rangle_{S(E)} = \text{Tr}_{S(E)} \left[ \rho_{S(E)}(0) \cdot \cdot \cdot \right] \quad (8)$$

for an average over the initial state of the system (environment). The time evolution is generated by a superoperator that acts exclusively on the qubit space:

$$e^{-\frac{1}{2} L(t)} = \left\langle T e^{\frac{1}{2} \int_0^t dt' L_V(t')} \right\rangle_{E}. \quad (9)$$

$T$ is the usual time-ordering operator. The interaction-picture Liouvillian $L_V(t)$ is defined by its action on an arbitrary operator $O$ through

$$L_V(t) O = \left[ \hat{V}(t), O \right]. \quad (10)$$

The action of Eq. (9) can generally be described by an affine map (see Ref. [31])

$$e^{-\frac{1}{2} L(t)} S = [R'(t)] \cdot [M(t)] \cdot [R(t)] \cdot S + \langle S(t) \rangle. \quad (11)$$
Here, \([R(t)]\) and \([R'(t)]\) are SO(3) rotation matrices. \([M(t)]\) is a magnification matrix that is diagonal with real eigenvalues,

\[
[M(t)] = \begin{pmatrix}
e^{-\lambda_1(t)/2} & 0 & 0 \\
0 & e^{-\lambda_2(t)/2} & 0 \\
0 & 0 & e^{-\lambda_3(t)/2}
\end{pmatrix}.
\] (12)

The inhomogeneous term in Eq. (11), \((\delta S(t))\), typically sets the long-time equilibrium value of the spin, \(\langle \delta S(t) \rightarrow \infty \rangle\), which is independent of the initial state for an ergodic system. For systems interacting with a sufficiently high-temperature thermal environment, the inhomogeneous term may be negligible,

\[
\langle \delta S(t) \rangle \approx 0.
\] (13)

Indeed, this turns out to be the case in the experimentally relevant problem of a hole spin interacting with an unpolarized nuclear-spin bath, which we address below (see also Appendix A). Thus, we first proceed under the (realistic) assumption that Eq. (13) is satisfied.

Inserting Eq. (17) into Eq. (10), and applying Eq. (11) with \((\delta S(t)) = 0\) gives

\[
\langle S(t) \rangle = [R_0(t)] \cdot [R'(t)] \cdot [M(t)] \cdot [R(t)] \cdot \langle S(0) \rangle.
\] (14)

The first two rotations, \([R_0(t)] \cdot [R'(t)]\), preserve the length of the Bloch vector, so they will not enter into the formula for purity. This leaves

\[
\langle S(t) \rangle = \sum_{\mu} [M(t)]_{\mu\mu} \langle S(0) \rangle. \]

The effect of \([R(t)]\) is to align the Bloch vector along principal axes defined by a set of mutually orthogonal unit vectors \(\hat{e}_\mu(t)\) (see, e.g., Fig. 1 below),

\[
\hat{e}_\mu(t) \cdot S = \langle [R(t)] \cdot S \rangle_\mu.
\] (16)

Using Eqs. (12), (13), and (10) in Eq. (1) then gives a compact form for the purity,

\[
P(t) = \frac{1}{2} + \sum_{\mu} e^{-\lambda_\mu(t)} \langle S(0) \rangle \cdot \hat{e}_\mu(t)^2.
\] (17)

The purity of the qubit at time \(t\) therefore depends on the eigenvalues \(\lambda_\mu(t)\) and on the initial conditions through \(\langle S(0) \rangle \cdot \hat{e}_\mu(t)\). In particular, it is always possible to maximize \(P(t)\) by choosing to initialize the qubit along a direction \(\hat{e}_\mu(t)\) associated with the smallest eigenvalue, \(\lambda_\mu(t) < \lambda_\nu(t)\) (\(\mu \neq \nu\)). In this case, the purity is given simply by

\[
P_\mu(t) = \frac{1}{2} \left(1 + e^{-\lambda_\mu(t)}\right), \quad |\langle S(0) \rangle \cdot \hat{e}_\mu(t)\rangle = 1/2.
\] (18)

Note that the general case of finite \((\delta S(t))\) is not significantly more complex—in this case, we simply need to find the initial state \(\langle S(0) \rangle\) that maximizes the magnitude

\[
|\langle S(t) \rangle| = \left| [R'(t)] \cdot [M(t)] \cdot [R(t)] \cdot \langle S(0) \rangle + \langle \delta S(t) \rangle \right|.
\] (19)

However, the result for this general case cannot be expressed in the simple form of Eq. (17).

When \(L(t)\) can be expressed as a real symmetric matrix \([L_{\alpha\beta}] = [L]_{\alpha\beta}\), where \([L_{\alpha\beta}] = 2\text{Tr} \{S_\alpha LS_\beta\}\), this matrix is diagonalized with an orthogonal rotation, i.e., \([R'(t)] = [R^{-1}(t)]\) in Eq. (11). In this case, the parameters \(\lambda_\mu(t)\) are the real eigenvalues of the superoperator \(L(t)\) and the unit vectors \(\hat{e}_\mu(t)\) determine the associated eigenerators through

\[
L(t) \hat{e}_\mu(t) \cdot S = \lambda_\mu(t) \hat{e}_\mu(t) \cdot S.
\] (20)

Decomposing the spin operator \(S\) in terms of its components along the unit vectors \(\hat{e}_\mu\) then gives a simplified expression for the spin expectation values when \([L]\) is symmetric,

\[
\langle S(t) \rangle = \sum_{\mu} e^{-\lambda_\mu(t)/2} \hat{e}_\mu(t) \cdot \langle S(0) \rangle \cdot [R_0(t)] \cdot \hat{e}_\mu(t).
\] (21)

The case of a real symmetric generator \(L(t)\) will be relevant to the example of hole-spin echo dynamics, which we address in the following sections.

To solve the eigenvalue equation, Eq. (20), it is first necessary to derive a suitable approximation for the superoperator \(L(t)\), defined by Eq. (9). As will be shown below, when \(H_0\) generates sufficiently rapid oscillations in \(\tilde{V}(t)\), a leading-order Magnus expansion can be performed on the time-ordered exponential in Eq. (2). For sufficiently large environment with initial state described by many uncorrelated degrees of freedom, the moments associated with the average \(\langle \cdot \cdot \cdot \rangle_E\) will be approximately Gaussian. When \(\langle L_\nu(t) \rangle_E = 0\), the combination of these two approximations leads to

\[
L(t) \simeq L_0(t) = \int_0^t dt_1 \int_0^t dt_2 \langle L_\nu(t_1) L_\nu(t_2) \rangle_E.
\] (22)

Of course, the applicability of Eq. (22) depends sensitively on the details of the physical system under study. In the remaining sections we will evaluate and justify this formula for a model with a pure Ising-like anisotropic hyperfine coupling. This model is directly relevant to hole spins in quantum dots.

III. HEAVY-HOLE SPIN-ECHO DYNAMICS

We consider a model Hamiltonian, appropriate for a heavy-hole spin qubit in a flat semiconductor quantum dot\(^{14}\) with an in-plane magnetic field applied along the \(x\)-axis (see Fig. 1):
In the sums above, $j$ labels the distinct nuclear isotopes and $k_j$ labels the set of sites occupied by nuclear spins of isotope $j$. Here, $H_Z$ gives the hole-spin and nuclear-spin Zeeman terms, and $H_{hf}$ describes the anisotropic hyperfine interaction between hole and nuclear spins. $S = \sigma/2$ is a pseudospin-1/2 operator in the heavy-hole subspace, and $I_k$ is the nuclear spin at site $k$. The hole gyromagnetic ratio is $\gamma_H = \mu_B g_{\perp}$, with $\mu_B$ the Bohr magneton and $g_{\perp}$ the in-plane hole $g$-factor. The gyromagnetic ratio of the nucleus at site $k_j$ of isotopic species $j$ having total spin $I_j$ is denoted by $\gamma_{j}$. The hyperfine couplings, $A_{k_j}$, are given by $A_{k_j} = A^{I}v_{0}\vert\psi(\mathbf{r}_{k_j})\vert^{2}$, where $A^{I}$ is the hyperfine coupling for nuclear species $j$, $v_{0}$ is the volume per nuclear spin, and $\psi(\mathbf{r}_{k_j})$ is the heavy-hole envelope wavefunction evaluated at site $\mathbf{r}_{k_j}$. For a Gaussian envelope function in two dimensions, $A_{k_j} \simeq \frac{A}{N} e^{-k/N}$, $k = 0, 1, 2, \ldots$, where $N$ is the number of nuclear spins within a quantum-dot Bohr radius.

Further assuming a uniform distribution of different nuclear species across the dot and $N \gg 1$ (typically $N \simeq 10^{4} - 10^{6}$), we define the average hyperfine constant $A$ as

$$A = \sum_{k} A_{k} \simeq \sum_{j} \nu_{j} A^{j},$$

where $\nu_{j}$ is the isotopic abundance of species $j$. In numerical estimates we will assume, for simplicity, a single average value $A$ of the hyperfine constant corresponding to $\nu_{j}$ for an In$_{0.5}$Ga$_{0.5}$As quantum dot ($A \simeq 13\mu$eV), and $\gamma_{j}$, $I_{j}$ appropriate for natural abundances of isotopes of Ga, As, and In taken from Table 1 of Ref. 63. For heavy holes, the ratio $|A/A^{(e)}|$ of hole to electron hyperfine coupling strengths has been estimated theoretically in GaAs and confirmed experimentally in InGaAs and InP/GaInP to be of order $|A/A^{(e)}| \sim 0.1$. This is consistent with $A \simeq 13\mu$eV since $A^{(e)} \simeq 90\mu$eV in GaAs.

Random fluctuations in the nuclear field cause rapid hole-spin decoherence via the hyperfine coupling described above. A spin-echo sequence can remove fluctuations that are approximately static over the time scale of hole-spin preparation and measurement. A Hahn echo sequence corresponds to a free evolution for time $t < \tau$, application of a $\pi$-rotation about the $x$-axis, $U_{x}(\pi)$, at $t = \tau$, followed by another free evolution for time $t \in (\tau, 2\tau)$. We consider a second $\pi$-rotation, $U_{x}^{\dagger}(\pi)$, at $t = 2\tau$ to return the spin to its original orientation (see Fig. 2). Noting that $U_{x}(\pi)S_{z}U_{x}^{\dagger}(\pi) = -S_{z}$, but $U_{x}(\pi)S_{x}U_{x}^{\dagger}(\pi) = S_{x}$, we account for the Hahn echo sequence illustrated in Fig. 2 with the identifications:

$$H_{0} = H_{Z}, \quad V(t) = s(t)H_{hf},$$

where

$$s(t) = \begin{cases} +1 & 0 \leq t < \tau, \\ -1 & \tau \leq t \leq 2\tau. \end{cases}$$

With the associations given in Eq. (28), we can now apply the analysis of Sec. IB to the problem of hole spin echo, using the leading-order Magnus expansion and Gaussian approximation to obtain the approximate generator $\mathcal{L}(2\tau) \simeq \mathcal{L}_{0}(2\tau)$ given in Eq. (22). See Refs. 34, 36 and 67 for further details on implementing the Magnus expansion and Gaussian approximation specific to this problem.

To make analytical progress, we rewrite the superoperator $\mathcal{L}_{0}$ in matrix form. In the basis of hole-spin operators, $\{S_{x}, S_{y}, S_{z}\}$, the matrix elements $[\mathcal{L}_{0}(2\tau)]_{\alpha\beta}$ are given by

$$\mathcal{L}_{0}(2\tau)S_{\alpha} = \sum_{\beta} [\mathcal{L}_{0}(2\tau)]_{\beta\alpha} S_{\beta}; \quad \alpha, \beta = x, y, z. \quad (30)$$

The matrix $[\mathcal{L}_{0}(2\tau)]$ can be found explicitly in terms of
bath correlation functions $\langle B_\alpha(2\tau)B_\beta(2\tau) \rangle$, with bath operators $B_\alpha$ defined by (see Appendix A):
\[
\int_0^{2\tau} dt \dot{V}(t) = \sum_\alpha B_\alpha(2\tau)S_\alpha.
\] (31)

We assume the initial state of the nuclear-spin bath describes uncorrelated spins without second-order coherences and with vanishing polarization, so that
\[
\langle h_j^+ h_j^- \rangle = \langle h_j^- h_j^+ \rangle = 0,
\] (32)
\[
\langle h_j^+ h_j^+ \rangle = \langle h_j^- h_j^- \rangle = 2\sigma_j^z \delta_{jj'}.
\] (33)

Here we have introduced the nuclear field operators over an isotope $j$,
\[
h_j = \sum_k A_{kj} I_{kj}, \quad h_j^z = h_j^y \pm i h_j^x.
\] (34)

Equation (33) above defines the nuclear-field fluctuation $\sigma_j$ due to isotope $j$. For the purposes of studying system-bath entanglement, it may be interesting to prepare a pure state of the bath and observe the resulting purity dynamics [see the discussion leading to Eq. (22) above]. We note that the conditions given in Eqs. (32) and (33) will be approximately satisfied for a pure state with suitably random initialization (e.g., by choosing a random orientation for each nuclear spin independently). For practical measurements, the initial conditions of the nuclear-spin bath are often well-described by an infinite-temperature thermal state, for which
\[
\sigma_j^2 = \frac{I_j(I_j + 1)}{3} \sum_k (A_{kj})^2.
\] (35)

For explicit estimates, we will make use of the total nuclear-field variance,
\[
\sigma_N^2 = \sum_j \sigma_j^2.
\] (36)

As suggested by the form of Eq. (31), the bath operators can be conveniently rewritten in terms of the complex-valued filter functions
\[
Z_j^\pm(2\tau) = \sigma_j \int_0^{2\tau} dt s(t) e^{i\omega_j t},
\] (37)
\[
\omega_j^\pm = (\gamma_H \pm \gamma_j) B.
\] (39)

In the conventional theory of spin-echo decay, functions such as Eq. (37) determine a filter function $F$, that restricts the frequency-content of the noise that can act to dephase a qubit through the absolute magnitude of $Z_j^\pm(2\tau)$,
\[
F(\omega_j, 2\tau) \propto |Z_j^\pm(2\tau)|^2.
\] (40)

Here, we will find both the magnitude and the phase of the functions $Z_j^\pm(2\tau)$ will be essential in determining spin dynamics. While the magnitude of the functions $Z_j^\pm(2\tau)$ will modify the spectral content of the noise, the phase of these functions will be crucial in determining a set of principal axes that determine the anisotropy of the decay process. Quite significantly, we will be able to exploit information about this decay anisotropy to identify optimal initialization/storage protocols to maximize the purity of a hole spin qubit.

Direct evaluation of the bath correlation functions and application of the relationships derived in Appendix A gives the matrix
\[
[L_0(2\tau)] = \begin{pmatrix} \lambda_x(2\tau) & 0 \\ 0 & [L^{\gamma^2}(2\tau)] \end{pmatrix}.
\] (41)

Here, one eigenvalue of the superoperator is $\lambda_x(2\tau)$. The $2 \times 2$ submatrix $[L^{\gamma^2}(2\tau)]$ can be written as
\[
[L^{\gamma^2}(2\tau)] = \frac{1}{2} \left[ \lambda_x(2\tau) \tau_0 + \text{Re} Z^2(2\tau) \tau_3 + \text{Im} Z^2(2\tau) \tau_1 \right],
\] (42)
where we have introduced the $2 \times 2$ identity matrix $\tau_0$ and usual Pauli matrices $\tau_\mu$. In Eq. (42), we have also introduced the complex function $Z(2\tau)$:
\[
Z^2(2\tau) = \sum_j Z_{j+}(2\tau) Z_{j-}(2\tau).
\] (43)

By diagonalizing the matrix in Eq. (41), we solve Eq. (29) for the eigenvalues, $\lambda_\mu(2\tau)$, and vectors, $\hat{e}_\mu(2\tau)$, with $\mu = x, \pm$:
\[
L_0(2\tau) [S \cdot \hat{e}_\mu(2\tau)] = \lambda_\mu(2\tau) [S \cdot \hat{e}_\mu(2\tau)].
\] (44)

We find the eigenvalues:
\[
\lambda_x(2\tau) = \frac{1}{2} \sum_j (|Z_{j+}(2\tau)|^2 + |Z_{j-}(2\tau)|^2),
\] (45)
\[
\lambda_\pm(2\tau) = \frac{1}{2} \left( \lambda_x(2\tau) \pm |Z(2\tau)|^2 \right).
\] (46)

When either $\gamma_H > \gamma_j$ or $\gamma_j > \gamma_H$ for all nuclear-spin species $j$, Eq. (46) becomes
\[
\lambda_\pm(2\tau) = \frac{1}{4} \sum_j (|Z_{j+}(2\tau)|^2 + |Z_{j-}(2\tau)|^2).
\] (47)

The associated unit vectors (illustrated in Fig. 3) are
\[
\hat{e}_x = \hat{x},
\] (48)
\[
\hat{e}_+(2\tau) = \cos \theta(2\tau) \hat{y} + \sin \theta(2\tau) \hat{z},
\] (49)
\[
\hat{e}_-(2\tau) = -\sin \theta(2\tau) \hat{y} + \cos \theta(2\tau) \hat{z}.
\] (50)

The angle $\theta(2\tau)$ is determined by the SU(2) rotation that diagonalizes Eq. (42). This angle is given by
\[
\theta(2\tau) = \arg Z(2\tau).
\] (51)
The vectors $\hat{e}_\mu(2\tau)$ are parametrized by the time $\tau$ between $\pi$-pulses. However, we stress that these are not dynamical quantities, evolving during the echo sequence. Instead, $\hat{e}_\mu(2\tau)$ determines the initial condition for a spin that should be chosen to recover a given purity $P_\mu(2\tau)$ after a Hahn echo sequence.

Inserting Eq. (38) for the complex-valued filter functions $Z_{j\pm}(2\tau)$ into Eq. (13) shows that $\theta(2\tau)$ can generally alternate between two simple forms:

$$\theta(2\tau) = \left\{ \begin{array}{ll} \gamma_H B\tau - \pi, & G(2\tau) > 0 \\ \gamma_H B\tau, & G(2\tau) < 0 \end{array} \right., \quad (52)$$

with

$$G(2\tau) = \sum_j \sigma_j^2 \sin^2 \left( \frac{\omega_{j\pm} \tau}{2} \right) \sin^2 \left( \frac{\omega_{j\pm} \tau}{2} \right) \frac{\gamma_H^2 - \gamma_j^2}{B^2} \cdot (53)$$

Alignment of the spin along $\hat{e}_-(2\tau)$ at time $t = 0$ will maximize the purity at time $t = 2\tau$ since $\lambda_-(2\tau)$ gives the smallest eigenvalue [see Eqs. (45), (46)]. Referring to Fig. 3 and Eq. (52) for the angle $\theta(2\tau)$, we see that when the nuclear-spin system can be taken as approximately static compared to the hole-spin precession, $\gamma_H > \gamma_j$ (giving $G > 0$), a spin initialized along $\hat{e}_-(2\tau)$ will advance in time at an angle $\phi = \theta(2\tau) + \pi/2 - \gamma_H B\tau = \gamma_H (\tau - t)$ from the $y$-axis. The spin will then be aligned with the $y$-axis at the time of the first $\pi$-pulse ($t = \tau$).

That this choice is optimal can be simply understood from a semiclassical model of a fluctuating magnetic field along $z$ in the limit of a purely static nuclear field and arises from the perfect symmetry of this problem for reflections through the $x$-axis within the $x$-$y$ plane (see Appendix). In the opposite limit of a slow hole-spin precession compared to the nuclear-spin precession, $\gamma_H < \gamma_j$ ($G < 0$), the optimal choice is to prepare the hole spin so that it aligns with the $z$-axis at the first $\pi$-pulse. This result is easy to understand in the limit $\gamma_H = 0$, since in this case the $z$-component of hole spin is a constant of the motion, and is therefore preserved for all time. More generally, when $\gamma_H \sim \gamma_j$, the optimal initialization axis will alternate nontrivially as a function of $\tau$ to favor alignment with either $\hat{y}$ or $\hat{z}$ at the time of the first $\pi$-pulse. When we consider additional pure-dephasing processes in Section below, we will find such a nontrivial behavior even when $\gamma_H \gg \gamma_j$, the limit typically realized in current experiments.

For this problem, spin dynamics in the $S_z$-subspace have been discussed previously. In this subspace, we find

$$\left\langle S_z(2\tau) \right\rangle / \left\langle S_z(0) \right\rangle \simeq e^{-\frac{1}{2} \lambda_0(2\tau)} \cdot (54)$$

A motional-averaging regime is reached for $\lambda_0 \lesssim 1$, corresponding to $\omega \gtrsim \sigma_N$, where $\sigma_N \sim A/\sqrt{N}$ is the typical amplitude of nuclear-field fluctuations and $\omega = B \cdot \max[\gamma_i, \gamma_H]$ gives the frequency of rapid oscillations. In this regime, the hole spin experiences envelope modulations with amplitude $\sim \lambda_0 \sim |\sigma_N/\omega|^2 < 1$.

FIG. 3. (Color online) Unit vectors satisfying the eigenvalue equation, Eq. (14), forming an orthonormal basis. $\hat{e}_x = \hat{x}$ while $\hat{e}_+(2\tau)$ and $\hat{e}_-(2\tau)$ correspond to $\hat{y}$ and $\hat{z}$ rotated by an angle $\theta(2\tau)$ [given by Eq. (51)] about the $x$-axis.

From Eq. (54), it is already clear that a hole spin initially aligned along $\hat{x}$ will have a purity that is modulated in time according to the envelope modulations.

Further setting $g_{1B} = \gamma_H = 0$ in the expressions above, Eq. (54) recovers the result previously given in Ref. 34

$$\left\langle S_z(2\tau) \right\rangle / \left\langle S_z(0) \right\rangle \simeq \exp \left[ -\sum_j \frac{8\sigma_j^2}{(\gamma_j B\tau/2)^2} \sin^4 \left( \gamma_j B\tau \right) \right]. \quad (55)$$

We now evaluate $\langle S_y(2\tau) \rangle$ and $\langle S_z(2\tau) \rangle$ by inverting Eqs. (49) and (50) for $\hat{e}_+(2\tau)$ and $\hat{e}_-(2\tau)$,

$$S_y = \cos \theta(2\tau) S \cdot \hat{e}_+(2\tau) - \sin \theta(2\tau) S \cdot \hat{e}_-(2\tau), \quad (56)$$

$$S_z = \sin \theta(2\tau) S \cdot \hat{e}_+(2\tau) + \cos \theta(2\tau) S \cdot \hat{e}_-(2\tau). \quad (57)$$

The evolution takes a simple form in terms of the operators $S_{\pm}^z$:

$$S_{\pm}^z = S_y \pm iS_z. \quad (58)$$

The spin evolution in the $y$-$z$ plane is then described by

$$\langle S_{\pm}^z(2\tau) \rangle = e^{i\phi(2\tau)} \sum_{\mu = \pm} \sqrt{\mu} e^{-\frac{1}{2} \lambda_\mu(2\tau)} \langle S(0) \rangle \cdot \hat{e}_\mu(2\tau), \quad (59)$$

where $\phi(2\tau) = -\gamma_H B2\tau + \phi(2\tau)$, $\sqrt{-} = \sqrt{1} = i$, and $\sqrt{+} = \sqrt{-1} = i$. The phase $\phi(2\tau)$ tracks the mismatch in evolution of the interaction-picture rotating frame and the rotation to principal axes for the generator $L_0(2\tau)$ (see Fig. 3). The eigenvalues $\lambda_\mu(2\tau)$ control the degree of damping/modulation in the amplitude of the spin.

The spin dynamics under the action of the anisotropic interactions presented here are strongly dependent on the initial direction of the spin and on the measurement axis. We will find it convenient to parametrize the initial state
for a spin in the $y$-$z$ plane by an angle $\varphi$ between the $y$-axis and the initial spin vector:

$$\langle S'_y(0) \rangle = \langle S_y(0) \rangle + i \langle S_z(0) \rangle = \frac{1}{2} e^{i\varphi}. \quad (60)$$

For a spin prepared at an angle $\varphi$ to the $y$-axis, we define the coherence factor in the rotating frame,

$$C_\varphi(2\tau) = 2e^{i\gamma_H B^2 \tau} \langle S'_+(2\tau) \rangle, \quad \langle S'_+(0) \rangle = \frac{1}{2} e^{i\varphi}. \quad (61)$$

We then find the general expression for this coherence factor,

$$C_\varphi(2\tau) = e^{-\lambda_+(2\tau)/4 + i\varphi} \left[ \cosh \left( \frac{|Z(2\tau)|^2}{4} \right) - e^{-i2(\varphi - \theta(2\tau))} \sinh \left( \frac{|Z(2\tau)|^2}{4} \right) \right]. \quad (62)$$

The first term in Eq. (62) [$\propto \cosh (|Z|^2/4)$] varies slowly in the rotating frame, while the second term [$\propto \sinh (|Z|^2/4)$] experiences violent modulations at a frequency determined by the hole-spin Zeeman energy, $2\theta(2\tau) \sim \gamma_H B^2 \tau$, due to non- secular “counter-rotating” corrections. This second contribution, $\sim |Z|^2 \ll 1$, evolves slowly in the lab frame, in spite of the hole-spin Zeeman term. Thus, while the first term would likely decay rapidly due to electric-field-induced fluctuations in the hole Zeeman energy, as reported in recent experiments, we expect some contribution from the second term to survive this dephasing mechanism. Such a pure-dephasing process is investigated in detail in Sec. V below.

Equation (62) recovers the expected results for initialization along one of the principal axes: $\varphi = \varphi_{\pm}(2\tau)$, corresponding to alignment of the initial spin with $\hat{e}_\pm(2\tau)$ (see Fig. 3):

$$C_{\varphi \pm}(2\tau) = \exp [i\varphi_{\pm}(2\tau) - \lambda_{\pm}(2\tau)/2], \quad (63)$$

$$\varphi_+(2\tau) = \theta(2\tau), \quad (64)$$

$$\varphi_-(2\tau) = \theta(2\tau) + \pi/2. \quad (65)$$

To explore the general spin dynamics of this problem, in which no special care has been taken to initialize the spin along one of the principal axes $\hat{e}_\pm$, we define the general correlators $C_{\alpha\beta}$ corresponding to the coherence for initialization along axis $\beta$ and measurement along axis $\alpha$ in the rotating frame:

$$C_{yy}(2\tau) = \Re \left[ C_{\varphi=0}(2\tau) \right], \quad (66)$$

$$C_{yx}(2\tau) = \Im \left[ C_{\varphi=0}(2\tau) \right], \quad (67)$$

$$C_{xz}(2\tau) = \Im \left[ C_{\varphi=\pi/2}(2\tau) \right], \quad (68)$$

$$C_{yz}(2\tau) = \Re \left[ C_{\varphi=\pi/2}(2\tau) \right]. \quad (69)$$

Correlators such as those given above have been measured, for example, in recent experiments on hole spins in single quantum dots. Those experiments showed similar modulations as seen here, although the authors of Ref. [40] have interpreted the modulations in their data in terms of a dynamic nuclear polarization effect. Two of the correlators above are shown for typical experimental parameters in Fig. 4. We note that the general correlators will contain contributions from each of the eigenvalues $\lambda_{\pm}(2\tau)$. While each of these experiences modulations at the nuclear Larmor frequency, the modulations for $\lambda_{\pm} = \sum_j (|Z_{j+}| + |Z_{j-}|)^2/4$ are $\pi$ out of phase with respect to each other [see, e.g., the modulations of $P_+$ (determined by $\lambda_+$) relative to those for $P_-$ (determined by $\lambda_-$) in Fig. 4]. These out-of-phase modulations generally lead to a sequence of maxima at twice the nuclear Larmor frequency, similar to the result seen for modulations in the experiment of Ref. [40]. The amplitude of modulations ($\propto 1/B^2$) is strongly suppressed in a large magnetic field $B$, so for high-field experiments, it may be difficult to see this effect. However, for $B \lesssim 1$ T and for typical quantum-dot parameters, the modulations can be a substantial fraction of the decay, as we show here.

IV. MAXIMIZING PURITY

The spin-echo purity, $P(2\tau)$, characterizes our ability to recover a pure ancilla qubit at a time $2\tau$ after prepara-
As discussed in Sec. II following Eq. (17), and as is clear from Eq. (70), the purity of a qubit recovered at time $2\tau$ can be maximized by initializing along the direction $\hat{e}_x(2\tau)$ associated with the smallest eigenvalue $\lambda_x(2\tau)$.

Naively, one might expect that the best strategy would be to prepare an ancilla qubit in an eigenstate (e.g., the ground state) of the unperturbed Hamiltonian, $H_0$. In the case of hole-spin qubits, this would correspond to preparing the spin along the applied magnetic field (along the $z$-axis for the geometry shown in Fig. 1), $|0\rangle = |\uparrow_x\rangle$ [where $S_z |\uparrow_x\rangle = +(1/2) |\uparrow_x\rangle$]. However, in this case, with the generator given in Eq. (41), we find the following general relationship, valid for all $\tau$ within the range of validity of the Gaussian approximation and leading-order Magnus expansion:

$$\lambda_x(2\tau) \geq \lambda_+(2\tau) \geq \lambda_-(2\tau). \quad (71)$$

The inequalities in Eq. (71) follow directly from Eqs. (45) and (46).

Quite generally, the purity is maximized by preparing the hole spin in the $y$-$z$ plane, in an equal superposition of Zeeman eigenstates:

$$P_{-}(2\tau) \geq P_{+}(2\tau) \geq P_{x}(2\tau). \quad (72)$$

The three quantities in Eq. (72) are shown in Fig. 5 for typical experimental parameters, illustrating the inequality. In the limit $\gamma_H = \mu_B g_\perp = 0$, this result can be intuitively understood. When $g_\perp = 0$, $|S_z, H\rangle = 0$, so a spin initialized along the $z$-axis will be preserved for all time, while a spin initialized along $\hat{x}$ or $\hat{y}$ will decay due to fluctuations along $\hat{z}$ (see the lower panel of Fig. 5 in this case, $\hat{e}_- = \hat{z}, \hat{e}_+ = \hat{y}$). That this relationship [Eq. (72)] continues to hold for $g_\perp \neq 0$ in any magnetic field and for all $\tau$ (within the range of validity of the approximations used here) is less trivially obvious.

It is straightforward to extend the above analysis to the more general case of an arbitrary anisotropic hyperfine tensor (see Appendix A). In this case, when leading non-secular corrections are included using the leading-order Magnus expansion and Gaussian approximations, the Zeeman ground state will not generally be optimal for initialization. The procedure described here can be used to predict an optimal state in which to store an ancilla. This may be useful in other systems with anisotropic interactions, including nitrogen-vacancy (NV) centers in diamond or phosphorus donors in silicon where the high-fidelity preparation of electron-spin ancillas is important for nuclear-spin readout.

The non-intuitive result given in Eq. (72) presupposes the absence of additional decoherence mechanisms. A rapid pure-dephasing process would typically reduce the purity for states initialized perpendicular to the magnetic field, relative to those initialized along the magnetic field. One source of pure dephasing for hole spins arises due to electric-field-induced fluctuations in the Zeeman energy. Such a mechanism has been identified as the predominant dephasing source for hole spins in Refs. 39 and 38. In the presence of a Markovian pure dephasing process that takes place on a time scale $T_\phi$, our conclusions remain valid in the limit $2\tau < T_\phi$ whenever the decay due to pure dephasing is small compared to the amplitude of envelope modulations, i.e. when $\gamma_H \gg \gamma_i$.

$$2\tau \lesssim 2\tau_c = T_\phi \left( \frac{A}{\sqrt{\mathcal{N}\gamma_H B}} \right)^2. \quad (73)$$

For storage of ancilla qubits beyond the time scale indicated in Eq. (73), it will be advantageous to prepare the qubit in the Zeeman eigenbasis.

It is interesting to note that, even in the limit of Markovian decay, it is possible to gain an advantage by storing an ancilla qubit in a superposition of non-interacting
eigenstates. In the absence of pure dephasing, $1/T_\phi = 0$, Bloch-Redfield theory gives $T_2 = 2T_1$, with longitudinal (transverse) spin relaxation time $T_1$ ($T_2$). For an infinite-temperature environment, decay of the component of spin transverse to an applied magnetic field occurs at a rate that is half that for the longitudinal component. The advantage in preparation perpendicular to an applied magnetic field will persist for moderately low-temperature environments.\footnote{We consider the detailed role of a pure-dephasing process on the general dynamics of a hole-spin qubit and on purity decay in the next section.}

$$\gamma_H B 2\tau \rightarrow \gamma_H B 2\tau + \phi(2\tau); \quad \phi(2\tau) = \int_0^{2\tau} dt \delta \omega(t),$$

(74)

with $\delta \omega(t)$ a Gaussian random variable describing a stationary white-noise process

$$\langle \langle \delta \omega(t) \delta \omega(t') \rangle \rangle = \frac{2}{T_\phi} \delta(t - t').$$

(75)

Here, double angle brackets $\langle \langle \cdots \rangle \rangle$ indicate an average over realizations of the noise $\delta \omega(t)$. The white-noise form given in Eq. (75) is a reasonable approximation for, e.g., Johnson-Nyquist noise due to nearby metallic gates.\footnote{This assumption will break down for, e.g., colored noise due to slowly-varying charged impurities.\footnote{It would be straightforward to extend the analysis presented here to the case of Gaussian colored noise. Indeed, in the case of colored Zeeman noise, it would be very interesting to explore two-axis echo sequences that could simultaneously refocus both nuclear-spin and electric-field noise. These studies are, however, beyond the scope of the present work. To emphasize the limitations of our earlier conclusions in the presence of pure dephasing, here we focus on the simplest (and often realistic) white-noise form given in Eq. (75).}}

Accounting for the modification to the Zeeman term, Eq. (74), the coherence factor in the rotating frame becomes

$$C_\phi(2\tau) = 2 \left\langle \langle e^{-i\phi} e^{-\frac{1}{2} \phi |\mathcal{L}|^2(2\tau)} \rangle \right\rangle_{S_+}, \langle S_+^0(0) \rangle = \frac{1}{2} e^{i\phi}.$$  

(76)

In the absence of hyperfine coupling, we have $\mathcal{L} = 0$, leaving a simple exponentially-decaying coherence factor,

$$C_\phi(2\tau) = e^{i\varphi - \frac{1}{2} \langle \phi^2(2\tau) \rangle} = e^{i\varphi - 2\tau/T_\phi},$$  

(77)

where we have used the fact that the noise is Gaussian in the first step and the fact that it is white [Eq. (75)] in the second. While $\phi$ is Gaussian-distributed, $\mathcal{L} |\phi\rangle$ is a highly nonlinear function of $\phi$, making a direct Gaussian average difficult. In general, we would like an expansion valid up to time scales $t \gtrsim T_\phi$ (giving $\langle \langle \phi \rangle \rangle \gtrsim 1$), so an expansion for small $\phi$, which was justified in evaluating the longitudinal spin $\langle S_x(2\tau) \rangle$,\footnote{is not generally possible for the coherence factor. Instead, here we perform a moment expansion, valid for $|\mathcal{L}| \lesssim 1$,

$$\langle \langle e^{-i\phi} e^{-\frac{1}{2} \phi |\mathcal{L}|^2} \rangle \rangle = \langle \langle e^{-i\phi} \rangle \rangle (e^{-\frac{1}{2} \phi |\mathcal{L}|^2}).$$  

(78)

Here, for an arbitrary operator $\mathcal{O}$, the average $\langle \langle \cdots \rangle \rangle$ is defined by

$$\langle \langle \mathcal{O} |\phi\rangle \rangle = \frac{\langle \langle e^{-i\phi} \mathcal{O} |\phi\rangle \rangle}{\langle \langle e^{-i\phi} \rangle \rangle} = e^{\frac{i}{2} \langle \phi^2 \rangle} \langle \langle e^{-i\phi} \mathcal{O} |\phi\rangle \rangle.$$  

(79)

At leading order in the moment expansion,

$$\langle \langle e^{-\frac{1}{2} \phi |\mathcal{L}|^2} \rangle \rangle \approx e^{-\frac{1}{2} \phi |\mathcal{L}|^2}.$$  

(80)

From a leading-order Magnus expansion, we have $\mathcal{L} \approx \mathcal{L}_0$ and in the regime of applicability of the Magnus expansion, $|\mathcal{L}_0(2\tau)| \ll 1$ for all time, allowing us to neglect all higher moments with small corrections.

Averaging over random instances of the fluctuating Zeeman term, $\delta \omega(2\tau)$, then gives an analogous expression to Eq. (82), but accounting for pure dephasing:

$$\langle \langle C_{\phi}^{2\tau} \rangle \rangle |\phi\rangle = \frac{1}{2} \left\langle \langle \lambda_\varphi \rangle \tau_0 + \langle \text{Re} Z^2 \rangle \phi \tau_3 + \langle \text{Im} Z^2 \rangle \phi \tau_1 \right\rangle.$$  

(81)

The averages above are evaluated explicitly in Appendix C. The coefficients in Eq. (81) are no longer real, but the matrix can nevertheless be exponentiated directly to determine the coherence factor

$$C_\varphi(2\tau) = e^{-2\tau/T_\phi - \langle \lambda_\varphi \rangle \phi/4} \left[ e^{i\varphi \cosh Q^2/4} - e^{-i\varphi \langle \text{Re} Z^2 \rangle \phi \sinh Q^2/4} \right].$$  

(82)

where

$$Q^2 = \left[ \langle \text{Re} Z^2 \rangle_{\phi}^2 + \langle \text{Im} Z^2 \rangle_{\phi}^2 \right]^{1/2}.$$  

(83)

The angle $\theta$ is then the value $\theta = \varphi$ for which the length of the Bloch vector (set by $|C_{\varphi}|$) is minimal (corresponding to alignment along the vector $\hat{e}_+\phi$). This value can be read off directly from Eq. (82), giving

$$\theta = \frac{1}{2} \arg \left[ \langle Z^2 \rangle_{\phi} \tanh Q^2/4 \right].$$  

(84)
at $t = \tau [\theta(2\tau) - \gamma_H B \tau \simeq -\pi/2]$ or the $z$-axis at $t = \tau [\theta(2\tau) - \gamma_H B \tau \simeq 0]$. The additional dynamics induced through the average over random Zeeman fields gives rise to a nontrivial evolution of the angle $\theta(2\tau)$ beyond this simple picture (see Fig. 4). While these corrections may be small here, they can be accurately determined using the procedure outlined above provided the dephasing model itself is known accurately.

The correlators $C_{\alpha\beta}$ corresponding to initialization along direction $\beta \in \{y, z\}$ and measurement along direction $\alpha \in \{y, z\}$ are shown in Fig. 5 for typical experimental parameters. Here we account for both pure dephasing from electric-field fluctuations and modulations of the decay envelope due to hyperfine coupling. Notably, $C_{yy}$ and $C_{zz}$ show a strong full-amplitude decay with small modulations [Figs. 8(a) and 8(b)]. In contrast, $C_{zy}$ and $C_{yz}$ [Figs. 8(c) and 8(d)] grow on a very short time scale on the order of the inverse hole-spin precession frequency, and subsequently slowly decay. Within the approximations made above, there will generally be a small non-decaying portion of the coherence arising from counter-rotating contributions to $C_C$ that are independent of the fluctuating Zeeman energy to leading order.

Detail of the purities for initialization along each of the three principal directions ($\hat{e}_x, \hat{e}_y$) is shown in Fig. 4 accounting for pure dephasing. For time $\tau < \tau_c$, with $\tau_c$ given by Eq. (73), the optimal initialization axis alternates between $\hat{e}_x$ (giving $P_+ + i, \text{i.e., initialization perpendicular to the magnetic-field quantization axis}$) and $\hat{e}_y$ (giving $P_{-y}$, initialization along the magnetic field). In contrast, for $\tau > \tau_c$, pure-dephasing processes dominate over the effect of envelope modulations and it will be advantageous once again to initialize a hole spin along the magnetic field.

\section{Conclusions}

We have given a general procedure for the calculation of the non-Markovian dynamics of qubit purity for qubits interacting with an anisotropic environment. Applying this procedure to the case of a hole-spin qubit interacting with a nuclear-spin bath, we find that (at sufficiently short times), the qubit purity is maximized by storing the qubit in a superposition of Zeeman eigenstates. Storage of the qubit in its Zeeman ground state can actually be the worst choice for this system. That storage in the computational basis (non-interacting eigenbasis) is suboptimal is not unique to hole spins. We expect this to be true for a wide variety of qubit systems when ancillas are required a short time after preparation and if pure-dephasing processes are weak. This effect is especially pronounced for systems interacting with anisotropic non-Markovian environments, including hole spins, nitrogen-vacancy center spins, and spins bound to phosphorus donor impurities.

In the process of calculating purity for a hole-spin qubit, we have given closed-form analytical expressions
FIG. 8. (Color online) Correlators $C_{\alpha\beta}(2\tau)$ showing dynamics for preparation along axis $\beta$ and measurement along axis $\alpha$ in the rotating frame. The material parameters used are the same as in Figs. 4 and 5 for an In$_{0.5}$Ga$_{0.5}$As quantum dot, but with magnetic field $B = 400$ mT and with a pure dephasing process giving rise to $T_\phi = 1$ $\mu$s.

for all spin components describing the spin-echo dynamics of hole spins in the presence of a nuclear-spin bath. In particular, we have shown how echo envelope modulations can be described by a combination of terms arising from (i) a rotation to a set of principal axes $\hat{e}_\mu(2\tau)$ for the generator of evolution $\mathcal{L}(2\tau)$, and (ii) modulations in a set of eigenvalues $\lambda_\mu(2\tau)$. While both contributions enter into the spin dynamics in general, the eigenvalues are most important for determining the purity, provided the spin is initialized along an appropriate principal axis. We have fully accounted for a pure-dephasing process arising from white-noise fluctuations in the hole-spin Zeeman energy and have illustrated the resulting rich dynamics.

We expect the calculations for qubit purity given here to be useful in quantum-information protocols that require high-purity ancillas, including quantum error correction, algorithmic cooling, and methods for high-fidelity readout. The general approach taken here emphasizes the fact that, for anisotropic systems, optimizing coherence is not simply a matter of manipulating the spectral content of the noise [associated with eigenvalues $\lambda_\mu(2\tau)$], but also the geometry of the noise, determined by initializing with respect to principal axes $\hat{e}_\mu(2\tau)$.

FIG. 9. (Color online) Purities for preparation along the unit vectors $\hat{e}_\mu$ in the presence of a Markovian pure dephasing process giving $T_\phi = 1$ $\mu$s. The purities are $P_+$ (red solid line), $P_\parallel$ (blue dashed line), and $P_\perp$ (black dotted line). The material parameters are as in Figs. 4 and 5 for an In$_{0.5}$Ga$_{0.5}$As quantum dot, and for this plot we have taken a magnetic field of $B = 400$ mT. In the presence of the pure-dephasing process, at certain times it becomes advantageous to prepare the qubit along the $\hat{e}$ direction [when $P_\parallel(2\tau) > P_+(2\tau)$].
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Appendix A: Average Hamiltonian and generator

We take the leading-order Magnus Hamiltonian to have the general form

$$H^{(0)}(t) = \int_0^t dt' \tilde{V}(t') = \sum_{\alpha} \mathcal{B}_\alpha(t) S_\alpha,$$

where $\mathcal{B}_\alpha(t)$ are Hermitian bath operators that act exclusively on the environment.

The matrix elements of $\mathcal{L}_0$ [defined by Eq. (22) of the main text] can then be written in terms of $\mathcal{B} = (\mathcal{B}_x, \mathcal{B}_y, \mathcal{B}_z)^T$ as

$$[\mathcal{L}_0]_{\alpha} = \frac{1}{2} \text{Tr} \{ \sigma_0 \mathcal{L}_0 S_\alpha \},$$

where

$$\mathcal{L}_0\alpha = -\frac{1}{2} \text{Im} \langle \mathcal{B} \times \mathcal{B}\rangle_E \cdot \hat{e}_{\alpha},$$

$$[\mathcal{L}_0]_{\alpha\beta} = 2 \text{Tr} \{ S_\alpha \mathcal{L}_0 S_\beta \},$$

$$= \text{Re} \{ \delta_{\alpha\beta} \mathcal{B} \cdot \mathcal{B}_E - \langle \mathcal{B}_\beta \mathcal{B}_\alpha \rangle_E \}. $$

Here, $\sigma_0$ is the identity in the qubit Hilbert space and $\hat{e}_{\alpha}$ is a unit vector along an axis in Cartesian coordinates, $(\hat{e}_x = \hat{x}, \hat{e}_y = \hat{y}, \hat{e}_z = \hat{z})$. A sufficient condition for the inhomogeneous term, Eq. (A3), to vanish within a leading-order Magnus expansion, Eq. (A1), is then:

$$\text{Im} \langle \mathcal{B} \times \mathcal{B}\rangle_E = 0 \Rightarrow \langle \delta S(t) \rangle = 0. $$

We now consider the most general anisotropic hyperfine Hamiltonian,

$$V = H_{\text{shf}} = \sum_{\gamma, \delta, k} A_{k}^{\gamma, \delta} I_{k}^{\gamma} S_{\delta}. $$

If the environment Hamiltonian is described by a general inhomogeneous Zeeman term,

$$H_I = -\sum_{k} \gamma_k \mathcal{B}_k \cdot I_k,$$

the interaction picture results in a rotation

$$\mathcal{B}_\beta(t) = \sum_{k, \gamma \delta} A_{k}^{\gamma, \delta} g_{k}^{\gamma \delta, \alpha \beta}(t) I_{k}^{\alpha}.$$

For the case of nuclear spin $I = 1/2$, for example, the coefficients in the expansion of Eq. (A9) are given explicitly by

$$g_{k}^{\gamma \delta, \alpha \beta}(t) = \int_0^t dt' 2 \text{Tr} \left\{ \tilde{I}_{k}(t') I_k^0 \right\} \cdot 2 \text{Tr} \left\{ \tilde{S}_{\delta}(t') S_{\beta} \right\}.$$ (A10)

With Eq. (A9), it is straightforward to estimate the matrix elements given in Eq. (A3). All terms are proportional to the initial polarization of the nuclear-spin system and therefore vanish:

$$\text{Im} \langle \mathcal{B} \times \mathcal{B}\rangle_I = 0. $$ (A11)

Here, we have used the subscript $I = E$ for the nuclear-spin environment. Thus, for an initially unpolarized nuclear spin bath, we are justified in neglecting the inhomogeneous term to leading order in a Magnus expansion,

$$\langle I_k^2 \rangle_I = 0 \Rightarrow \langle \delta S(t) \rangle \simeq 0. $$ (A12)

Specializing to the case of an Ising-like hyperfine interaction,

$$A_k^{\alpha \beta} = \delta_{\alpha \sigma} \delta_{\beta \sigma} A_k,$$ (A13)

and the spin-echo problem discussed in Sec. III, we find explicit forms for the bath operators $\mathcal{B}_n$, in terms of the complex-valued filter functions $Z_j(2\tau)$ given in Eq. (37) of the main text:

$$\mathcal{B}_x = 0,$$ (A14)

$$\mathcal{B}_y = \sum_j \frac{1}{4\sigma_j} \left[ (Z_{j_{-}} - Z_{j_{+}})^{h_j^+} - (Z_{j_{+}} - Z_{j_{-}})^{h_j^-} \right],$$

$$\mathcal{B}_z = \sum_j \frac{-i}{4\sigma_j} \left[ (Z_{j_{-}} + Z_{j_{+}})^{h_j^+} - (Z_{j_{+}} - Z_{j_{-}})^{h_j^-} \right].$$

Applying the rules in Eq. (33) for an uncorrelated and unpolarized nuclear-spin state immediately gives the nonvanishing correlators,

$$\langle \mathcal{B}_y \mathcal{B}_y \rangle = \frac{1}{2} \langle \lambda_x - ReZ^2 \rangle,$$ (A15)

$$\langle \mathcal{B}_z \mathcal{B}_z \rangle = \frac{1}{2} \langle \lambda_x + ReZ^2 \rangle,$$ (A16)

$$\langle \mathcal{B}_y \mathcal{B}_z \rangle = \langle \mathcal{B}_z \mathcal{B}_y \rangle = -\frac{1}{2} \text{Im} Z^2.$$ (A17)

Here, $Z^2(2\tau)$ and $\lambda_x(2\tau)$ are given in Eqs. (43) and (45) of the main text, respectively. Inserting these correlators into Eq. (A5) directly gives the matrix form found in the main text [Eqs. (41) and (42)].

Parenthetically, we note that arbitrary initial conditions for the bath can be taken above, in principle, including pure-state environment initial conditions (required to measure entanglement through purity). However, for each bath initial state, it will be important to justify the Gaussian approximation used to derive Eq. (22). This approximation is very good for an uncorrelated thermal bath or a sufficiently random ‘narrowed’ state, but may break down for pure initial conditions with strong (classical or quantum) correlations.
Appendix B: Simple example: $\gamma_j = 0$

It is useful to consider a simple and direct application of the analytical expressions derived in Appendix A. Here we consider dynamics at a time scale short compared to the nuclear-spin precession period, and neglect the nuclear gyromagnetic ratio, so there is effectively one nuclear-spin species $j$ with $\gamma_j \approx 0$ and we assume a single nuclear-field variance $\sigma_j = \sigma_N$. Under these conditions, there is only one complex filter function, $Z_{j+}(2\tau) = Z(2\tau)$, for one fixed $j$ [see Eq. (37)]. Setting $\omega = \gamma_H B$,

$$Z(2\tau) = \frac{4\sigma_N}{\omega} \sin^2 \frac{\omega\tau}{2} e^{i(\omega\tau - \pi/2)}. \quad (B1)$$

From Eq. (A2), we see that the submatrix can be rewritten as an outer product (a projector onto a vector constructed from the real and imaginary parts of $Z = X + iY$):

$$[\mathcal{L}_0^\gamma(2\tau)] = \begin{pmatrix} X^2 & XY \\ XY & Y^2 \end{pmatrix} = \begin{pmatrix} X \\ Y \end{pmatrix} \begin{pmatrix} X \\ Y \end{pmatrix}. \quad (B2)$$

The eigenvectors giving $\hat{e}_\pm$ are then simply this vector and the vector orthogonal to it:

$$[\mathcal{L}_0^\gamma(2\tau)] \begin{pmatrix} X \\ Y \end{pmatrix} = |Z|^2 \begin{pmatrix} X \\ Y \end{pmatrix}, \quad (B3)$$

$$[\mathcal{L}_0^\gamma(2\tau)] \begin{pmatrix} Y \\ -X \end{pmatrix} = 0. \quad (B4)$$

In terms of unit vectors,

$$\hat{e}_+(2\tau) = \frac{1}{|Z(2\tau)|} X(2\tau)\hat{y} + Y(2\tau)\hat{z}, \quad (B5)$$

$$\hat{e}_-(2\tau) = \frac{1}{|Z(2\tau)|} (-Y(2\tau)\hat{y} + X(2\tau)\hat{z}). \quad (B6)$$

Equation (B5) gives a quick shortcut to find the angle $\theta(2\tau)$ in Fig. 3:

$$\theta(2\tau) = \arg Z(2\tau) = \gamma_H B\tau - \frac{\pi}{2}. \quad (B7)$$

From Eqs. (B3) and (B4), we can read off the eigenvalues,

$$\lambda_+(2\tau) = |Z(2\tau)|^2 = \left(\frac{4\sigma_N}{\gamma_H B}\right)^2 \sin^4 \frac{\gamma_H B\tau}{2}, \quad (B8)$$

$$\lambda_-(2\tau) = 0. \quad (B9)$$

Equation (B9) indicates that a spin initially aligned along $\hat{e}_-$ will show no decay or modulations under a spin-echo sequence within the range of applicability of approximations made here. On the surface, this may not seem surprising since we have assumed $\gamma_j = 0$, making the bath static and the spin-echo dynamics reversible. However, a spin prepared along any other direction will show violent modulations, as described by Eq. (B8). The distinction between these two cases can be understood by considering the specific geometry and the phase in Eq. (B7). Before the first $\pi$-pulse, a spin initialized along $\hat{e}_+$ will evolve with a phase $\phi(t)$ [see also Eq. (59) for an analogous expression after the echo sequence has been carried out]:

$$\langle S_+^x(t) \rangle \propto e^{i\phi(t)}, \quad \phi(t) = -\gamma_H B\tau + \theta(2\tau). \quad (B10)$$

Note that $\theta(2\tau)$ does not evolve with $t$ since it determines the initial condition. Inserting Eq. (B7), we see that the initial condition is such that the spin lies along $\hat{z}$ at the time of the first $\pi$-pulse ($t = \tau$):

$$\phi(\tau) = -\frac{\pi}{2}. \quad (B11)$$

This situation leads to rapid envelope modulations. In contrast, a spin initialized along $\hat{e}_-$ will be oriented along $\hat{y}$ at the first $\pi$-pulse and will show no modulations. We can understand this difference by considering a model of a spin evolving in the presence of a classical magnetic field, $B = B_z\hat{x} + \delta B\hat{z}$, having a fixed $x$-component $B_z$ and slowly-varying random $z$-component $\delta B_z$. For initialization along $\hat{e}_+$, a finite $\delta B_z$ will result in a finite component along $\hat{x}$ at the time of the first $\pi$-pulse, but the spin will lie approximately in the $y-z$ plane due to the choice of initial condition (with small corrections in $\delta B_z/B_z \ll 1$). In this plane, the system shows perfect mirror symmetry for a reflection through the $x$-axis, so a $\pi$-pulse about $\hat{y}$ induces symmetric time-reversed dynamics, returning the spin precisely to its starting point in the rotating frame after a second $\pi$-pulse is performed at $t = 2\tau$. In contrast, if the spin is initialized along $\hat{e}_-$, it will lie approximately in the $x-z$ plane at the time of the first $\pi$-pulse. In this plane, for any finite value of $\delta B_z$, there is no reflection symmetry for a $\pi$-rotation about the $x$-axis. The spin’s cone of precession after the $\pi$-pulse can be quite different from that before the $\pi$-pulse, resulting in a mismatch in evolutions causing the modulations indicated by Eq. (B8) for any finite $\delta B_z$.

Appendix C: Averages for pure dephasing

Here we give expressions for the averages required to evaluate the associated generator $\langle \mathcal{L}_0 \rangle_\phi$, accounting for averages over realizations of the Gaussian random variable $\delta\omega(t)$ described by Eq. (75).

Explicitly, the eigenvalue $\langle \lambda_x \rangle_\phi$ can be written as

$$\langle \lambda_x \rangle_\phi = \frac{1}{2} \sum_j (\langle |Z_{j+}|^2 \rangle_\phi + \langle |Z_{j-}|^2 \rangle_\phi), \quad (C1)$$

where

$$\langle |Z_{j\pm}|^2 \rangle_\phi = 2\sigma_j^2 \int_0^{2\tau} dt_1 \int_0^{t_1} dt_2 s(t_1)s(t_2)F_{j\pm}(t_1 - t_2), \quad (C2)$$

and the functions $F_{j\pm}(t)$ are:

$$F_{j\pm}(t) = e^{-t/T_0} \cosh [(i\omega_{j\pm} + 2/T_0) t]. \quad (C3)$$
The remaining coefficients in the matrix representation of \((\mathcal{L}_0)_\phi\), \((\text{Re}Z^2)_\phi\), and \((\text{Im}Z^2)_\phi\), are given by

\[
(\text{Re}Z^2)_\phi = \sum_j \sigma_j^2 \int_0^{2\tau} dt_1 \int_0^{t_1} dt_2 K^+_j(t_1, t_2),
\]

\[
(\text{Im}Z^2)_\phi = -i \sum_j \sigma_j^2 \int_0^{2\tau} dt_1 \int_0^{t_1} dt_2 K^-_j(t_1, t_2),
\]

with integral kernels

\[
K^+_j(t_1, t_2) = s(t_1)s(t_2) \cos[\omega_j(t_1 - t_2)] e^{(t_1 - t_2)/T_\phi} \times \\
\times \left[ e^{i\gamma B(t_1 + t_2)} \pm e^{-i\gamma B(t_1 + t_2) - 4(t_1 + t_2)/T_\phi} \right].
\]

The integrals can all be evaluated analytically, but we leave them unevaluated here for notational convenience.

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