Dark State Polarizing a Nuclear Spin in the Vicinity of a Nitrogen-Vacancy Center

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The nuclear spin in the vicinity of a nitrogen-vacancy (NV) center possesses of long coherence time and convenient manipulation assisted by the strong hyperfine interaction with the NV center. It is suggested for the subsequent quantum information storage and processing after appropriate initialization. However, current experimental schemes are either sensitive to the inclination and magnitude of the magnetic field or require thousands of repetitions to achieve successful realization. Here, we propose polarizing a 13C nuclear spin in the vicinity of an NV center via a dark state. We demonstrate theoretically that it is robust to polarize various nuclear spins with different hyperfine couplings and noise strengths.

I. INTRODUCTION

Benefiting from quantum entanglement [1, 2], quantum information processing [3] can effectively speed up computation and ensure security of information [4]. As the basic element, quantum-bit (qubit) lies at the heart of quantum information processing [4, 5]. Solid-state qubits are a promising candidate because they might well explore the well-developed technology of semiconductor industry [6, 7]. Remarkably, the nitrogen-vacancy (NV) center in diamond has been recognized as an intriguing choice since it is of easy accessibility and long coherence time at room temperature [8–12], as can be measured by Landau-Zener-Stückelberg interferometry [13]. To date, various applications including quantum information processing and quantum metrology have been successfully realized in the NV centers [14–16]. For example, different versions of transitionless driving algorithms have been fully utilized to accelerate quantum control in the NV centers [17–20]. Besides, the NV centers have been explored to detect internal dynamics of clusters of nuclear spins by dynamical decoupling [21, 22] with sensitivity further improved by Fluoquet spectroscopy and coupling to collective modes [23, 24]. The NV center has also been proposed to detect the radical-pair chemical reaction in biology [25]. Due to the quantum nature of surrounding nuclear spin bath, the anomalous decoherence effect of the NV center has been theoretically predicted [27] and experimentally verified [28].

Apart from the electron spin of the NV center, the nuclear spins in the vicinity of an NV center is of broad interest to the community. Due to much longer coherence time, nuclear spins are more frequently used in the quantum information storage and processing [29–37]. However, it is difficult to initialize and control the nuclear spins because of their small magnetic moments. Utilizing an ancillary electronic spin to couple with the nuclear spin by the hyperfine interaction may effectively overcome this limitation.

To our best knowledge, there are three kinds of experimental schemes which have been successfully demonstrated to initialize the nuclear spins around the NV centers. A straightforward approach is to repeatedly perform projective measurements until the desired state is observed [32, 33]. An alternative is to bring the excited (ground) state close to the level-anticrossing point by applying a specific static magnetic field [31, 38]. In the last but widely-used approach [29, 30, 35, 39–40], the electron spin is first initialized, and then its polarization is coherently swapped to the nuclear spin. After tens of repetitions of the above process, nearly-complete polarizations of both electronic and nuclear spins are achieved. Here, we remark that in each repetition the swapping of polarization between the electron and nuclear spins is essentially a quantum-state transfer process.

For any state transfer process, the fidelity is inevitably influenced by the noise due to coupling to the bath. On the other hand, we notice that the dark state has been extensively applied to coherently transfer energy in photosynthetic light harvesting [41] and perfectly transfer state in optomechanical systems [12, 42]. The coherent coupling between a surface acoustic wave and an NV center has been experimentally realized via the dark state recently [42]. Inspired by these discoveries, we theoretically propose a novel method to polarize a 13C nuclear spin in the vicinity of an NV center by the dark state. In order to provide an effective guidance for the experimental realization, we performed an analytical analysis on the probability of the nuclear-spin polarized state by the Schrödinger equation with a non-Hermitian Hamiltonian. It is shown that when the Rabi frequencies of the two pulses are equal, the polarization is predict to reach the maximum at the given time. Further numerical simulation demonstrates an anomalous effect that the polarization of a nuclear spin with a smaller hyperfine interaction can be even higher than a nuclear spin.

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at the nearest neighbor site due to the transverse hyperfine interaction. Compared with the above methods, our scheme works effectively over a broad range of magnetic field and only a few repetitions are required.

II. PHYSICAL SETUP

As shown in Fig. 1(a), the NV center in diamond with a C3v symmetry consists of a nitrogen atom associated with a vacancy in an adjacent lattice site. For the negatively-charged NV center with electron spin $S = 1$, the ground state is a spin triplet state $^3A$, with a zero-field splitting $D = 2.87$ GHz [45] between spin sublevels $m_s = 0$ and $m_s = \pm 1$ due to the spin-spin interaction. In this article, we consider a first-shell $^{13}$C nuclear spin coupled with the electronic spin of an NV center. As a result, there is a strong hyperfine coupling $A_1 = 130$ MHz [46] between the nuclear and electronic spins. Figure 1(b) shows the energy-level diagram of the ground-state hyperfine structure associated with a nearby $^{13}$C nuclear spin. We label the states of this bipartite system as $|m_s, \uparrow\rangle$ and $|m_s, \downarrow\rangle$, where $|\uparrow\rangle$ and $|\downarrow\rangle$ are the nuclear spin states.

We apply a weak static magnetic field $B_z < 673$ G along the NV principle axis by a permanent magnet. The total Hamiltonian of the electron-spin ground state and a nearby $^{13}$C nuclear spin reads [35]

$$H_F = DS_z^2 + \gamma_e B_z S_z + \gamma_e B_x I_z + A_\parallel S_z I_z$$

$$+ A_\perp(S_x I_x + S_y I_y).$$

Here, $S_\alpha$ and $I_\alpha$ ($\alpha = x, y, z$) are respectively the electronic and nuclear spin operators. The first term stands for the zero-field splitting of the electronic ground state. The following two terms $\gamma_e B_z S_z$ and $\gamma_e B_x I_z$ are the electronic and nuclear spin Zeeman energy splittings with the electronic gyromagnetic ratio $\gamma_e = -1.76 \times 10^{11}$ rad $s^{-1}T^{-1}$ [47] and the nuclear gyromagnetic ratio $\gamma_n = 6.73 \times 10^7$ rad $s^{-1}T^{-1}$ [27]. The last two terms describe the hyperfine interaction between the electron spin and the $^{13}$C nuclear spin, where $A_\parallel$ and $A_\perp$ are the longitudinal and transverse hyperfine interactions respectively.

Due to the weak magnetic field strength, the difference between the electronic Zeeman splitting and the zero-field splitting is much larger than the transverse hyperfine interaction, i.e. $|D - \gamma_e B_z| \gg A_\perp$. Therefore the $S_x I_x$ and $S_y I_y$ terms of the hyperfine interaction are sufficiently suppressed. In this case, the longitudinal hyperfine interaction is taken into account and the secular approximation is valid [29, 39, 48, 49]. In the presence of time-varying magnetic fields, the Hamiltonian can be approximated as

$$H_F^2 \simeq DS_z^2 + \gamma_e B_z S_z + \gamma_e B_x I_z + A_\parallel S_z I_z.$$  (2)

III. POLARIZING BY DARK STATE

As shown in Fig. 1(b), the transition $|0, \uparrow\rangle \leftrightarrow |\downarrow, \uparrow\rangle$ is addressed via a microwave pulse with Rabi frequency $\Omega_1$ and driving frequency $\omega_A = D - \gamma_e B_z - \delta - A_\parallel/2$, while the transition $|\downarrow, \uparrow\rangle \leftrightarrow |\downarrow, \downarrow\rangle$ is driven via a radio-frequency pulse with Rabi frequency $\Omega_2$ and driving frequency $\omega_B = A_\parallel - \gamma_e B_z + \delta - \Delta$. Thus, in the presence of the two pulses the whole Hamiltonian of the system reads $H_M = H_F + H_I$, where the interaction Hamiltonian is

$$H_I = \Omega_1 e^{i\omega_A t}|0, \uparrow\rangle\langle -, \uparrow| + \Omega_2 e^{i\omega_B t}|-, \downarrow\rangle\langle -, \downarrow| + \text{h.c.}$$  (3)

Hereafter, we will demonstrate polarizing the nuclear spin by the dark state. The nuclear and electronic spins are initially in a product state [30, 33, 35]. In each cycle there are two steps. First of all, the system evolves under the hyperfine interaction $H_I$ while the transition $|\downarrow, \uparrow\rangle \leftrightarrow |\downarrow, \downarrow\rangle$ is driven via a radio-frequency pulse with Rabi frequency $\Omega_2$ and driving frequency $\omega_B = A_\parallel - \gamma_e B_z + \delta - \Delta$. Thus, in the presence of the two pulses the whole Hamiltonian of the system reads $H_M = H_F + H_I$, where the interaction Hamiltonian is

$$H_I = \Omega_1 e^{i\omega_A t}|0, \uparrow\rangle\langle -, \uparrow| + \Omega_2 e^{i\omega_B t}|-, \downarrow\rangle\langle -, \downarrow| + \text{h.c.}$$  (3)

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$$H_I = \Omega_1 e^{i\omega_A t}|0, \uparrow\rangle\langle -, \uparrow| + \Omega_2 e^{i\omega_B t}|-, \downarrow\rangle\langle -, \downarrow| + \text{h.c.}$$  (3)

The optical excitation with a 532-nm laser pulse leads to a strong spin polarization into the $|0\rangle$ sublevel of the ground state [9], which derives from spin-selective nonradiative intersystem crossing to a metastable state between the ground and excited triplets. In this sense, it is reasonable to choose the electronic initial state $\rho_e(0) = |0\rangle\langle 0|$. Due to the small nuclear Zeeman energy splitting with respect to the thermal energy, the nuclear spin is in the maximum mixed state $\rho_n(0) = (|\uparrow\rangle\langle \uparrow| + |\downarrow\rangle\langle \downarrow|)/2$. When the electronic spin is populated in $m_s = 0$, the hyperfine interaction vanishes.

![Figure 1](image-url)
Thus, the initial state of the total system is given by
\[
\rho(0) = \rho_e(0) \otimes \rho_i(0).
\] (4)

Then, the microwave pulse and the radio-frequency pulse drive the transitions \(|0, \uparrow\rangle \leftrightarrow |-, \uparrow\rangle\) and \(|-, \uparrow\rangle \leftrightarrow |-, \downarrow\rangle\) respectively. The total system evolves under the Hamiltonian \(H_M = H_F + H_I\) for a time interval \(t\). Since the total Hamiltonian \(H_M\) is time-dependent, it is transformed to the rotating frame defined by \(|\Psi(t)^R\rangle = U^\dagger(t)|\Psi(t)\rangle\) with \(U(t) = \exp[-i(H_F^R - \delta \uparrow \downarrow) t].\) Here, \(|\Psi(t)^R\rangle\) satisfies the Schrödinger equation in the rotating frame with the effective Hamiltonian

\[
H_M^R = U^\dagger (H_F^S + H_I) U + iU^\dagger U
\]
\[
= \delta \uparrow \downarrow \langle \uparrow | \uparrow \uparrow | + \Delta \langle \downarrow \downarrow | \uparrow \downarrow |
\]
\[
+ \Omega_1 \langle 0, \uparrow | \downarrow \downarrow | + \Omega_2 \langle -, \uparrow | \downarrow \downarrow | + \text{h.c.} \quad (5)
\]

Generally speaking, the quantum dynamics of the electron and nuclear spins is subject to the noise, which can be described by the master equation

\[
\partial_t \rho = -i[H_M, \rho] + \mathcal{L}\rho, \tag{6}
\]

where \(\mathcal{L}\rho = \kappa \langle \uparrow | \langle \uparrow | - \langle \uparrow | \downarrow \rangle \langle \downarrow | \uparrow \rangle - \frac{1}{2} \frac{i}{\hbar} \langle \downarrow | \downarrow \rangle \langle \uparrow | \uparrow \rangle \) describes the decoherence induced by the bath with \(\kappa\) being the decoherence rate. \(|\cdot, \rho\rangle\) is the anti-commutator. Because \(T_1\) is at least larger than \(T_2 = \kappa^{-1}\) by one order \([50-52]\), without loss of generality, we only take the pure-dephasing process into consideration in our simulation.

When the decoherence is sufficiently slow as compared to the coherent processes described by \(H_M\), the total quantum dynamics including the decoherence can be simulated by the Schrödinger equation with a non-Hermitian Hamiltonian \(H = H_M^R - \frac{i}{2} \kappa \langle \uparrow | \downarrow \rangle \langle \downarrow | \uparrow \rangle \). Because we apply two selective-resonance drivings to the NV center, the state \(|0, \downarrow\rangle\) is effectively decoupled from the other three states. Therefore, hereafter we can separately consider the quantum dynamics of an initial state \(|\psi(0)\rangle = |0, \uparrow\rangle\) driven by two microwave pulses. As presented in Appendix A at any time the state of the system reads

\[
|\psi(t)\rangle = \sum_{j=1}^3 \frac{N_j e^{-ix_j t}}{1 - F_j(x_j - x_k)} |E_j\rangle, \tag{7}
\]

where the three eigen states of \(H\) are

\[
|E_j\rangle = \frac{1}{N_j} \{ \Omega_1 (x_j - \omega_2), \uparrow \uparrow + \Omega_1 \Omega_2 (-, \downarrow) \}
\]
\[
+ \{ (x_j - \omega_1)(x_j - \omega_2) - \frac{\Omega_2^2}{\Omega_1^2} |0, \uparrow\rangle \} \tag{8}
\]

with \(N_j\)‘s being the normalization constants, \(x_j\)‘s being the eigen energies, \(\omega_1 = \delta - ik/2, \omega_2 = \Delta - ik/2\). When we employ two strong drivings to polarize the nuclear spin, i.e. \(\omega_1, \omega_2 \ll \Omega_1, \Omega_2\),

\[
|E_1\rangle \approx \frac{\Omega_2}{N_1} (-\Omega_2 |0, \uparrow\rangle + \Omega_1 |-, \downarrow\rangle) \tag{9}
\]

is the dark state because it is lack of the component of the lossy intermediate state \(|-, \uparrow\rangle\), while

\[
|E_2\rangle \approx \frac{\Omega_1}{N_2} (\Omega_1 |0, \uparrow\rangle + \Omega \langle -, \uparrow | + \Omega_2 |-, \downarrow\rangle) \tag{10}
\]

\[
|E_3\rangle \approx \frac{\Omega_1}{N_3} (\Omega_1 |0, \uparrow\rangle - \Omega \langle -, \uparrow | + \Omega_2 |-, \downarrow\rangle) \tag{11}
\]

are the bright states suffering from relaxation. In this case, the state of system is simplified as

\[
|\psi(t)\rangle = \frac{\Omega_1}{2\Omega} e^{-i(\omega_1 + \Omega^2/2 |E_2\rangle + e^{i\Omega t} |E_2\rangle + e^{i\Omega t} |E_3\rangle) \tag{12}
\]

with

\[
\Omega = \sqrt{\Omega_1^2 + \Omega_2^2}. \tag{13}
\]

By solving the Schrödinger equation with a non-Hermitian Hamiltonian, we numerically simulate the population dynamics of all three states for the resonance case, i.e. \(\delta = \Delta = 0\), as shown by the solid lines in Fig. 2. In one cycle, almost 100% population in \(|0, \uparrow\rangle\) can be coherently transferred to \(|-, \downarrow\rangle\) even in the presence of noise. In order to derive the non-Hermitian Hamiltonian, several approximations have been utilized, i.e. dropping the quantum jump terms in the master equation, and ignoring the transverse hyperfine interactions, and disregarding transitions due to large-detuning condition. In order to validate these approximations, we also present the numerical simulation with the dashed lines in Fig. 2.
Figure 3. Comparison between the probabilities of the nuclear spin in the $|\downarrow\rangle$ vs time by applying two simultaneous pulses (black dash-dotted line) and two separate pulses (red solid line). In the simultaneous case, $\Omega_1 = \Omega_2 = 13$ MHz, while $\Omega_1 = 4.3$ MHz in the separate case. Other parameters are the same in both cases, i.e. $A_\parallel = 130$ MHz, $\delta = \Delta = 0$, $\kappa = 1/58$ MHz.

by the exact master equation without the above approximations. Obviously, the differences between the two approaches are relative small and thus it is valid to simulate the quantum dynamics of the nuclear-spin polarization in the presence of noise.

By means of the Schrödinger equation with a non-Hermitian Hamiltonian, we can effectively analyze the effects of parameters on the nuclear-spin polarization and obtain a set of optimal parameters to guide the experiment for different conditions of nuclear spins. In the previous experimental realizations, cf. Refs. [39, 40], two $\pi$-pulses are sequentially applied to swap the electron-spin polarization into the nuclear-spin polarization. In our proposal, because we simultaneously apply two balanced pulses to make use of the dark state to avoid the noise suffered by the intermediate state, the nuclear-spin polarization in our proposal is much higher than theirs. As shown in Fig. 2 the polarization for our proposal is further optimized by choosing optimal parameters to reduce the swapping time, i.e. 0.90 vs 0.48.

The swapping of electron-spin polarization into nuclear-spin polarization is intrinsically a quantum state transfer process from $|0, \uparrow\rangle$ to $|-, \downarrow\rangle$ via a lossy state $|-, \uparrow\rangle$. Intuitively, the fidelity of state transfer is subject to the noise strength. In Fig. 2(a), we explore the noise’s effect on the fidelity of nuclear spin in the state $|\downarrow\rangle$ with $\kappa = 1$ MHz for different nuclear spins, i.e. different hyperfine interactions. For a nuclear spin in the first shell, the probability in $|\downarrow\rangle$ vs time manifests a damped vibration due to couplings to the environment. The maximum fidelity is more than 0.9 around the first peak at $t = \pi/\sqrt{2}\Omega_1 \simeq 0.17\mu$s as a longer pulse duration yields more loss. When a nuclear spin in the second shell is to be polarized, the maximum fidelity is about 0.85, less than that for the nuclear spin in the first shell as the Rabi frequency is smaller due to a weaker hyperfine interaction. If we choose a nuclear spin even further apart from the NV center, e.g. $A_\parallel = -7.5$ MHz, the maximum fidelity observably declines to 0.65. Because Rabi frequencies are limited by the large-detuning condition, the descending of maximum fidelity along with reducing of hyperfine interaction results from the increasing pulse duration. When the pure-dephasing rate $\kappa$ is reduced from 1 MHz to 1/5.8 MHz, e.g. Fig. 2(a) vs Fig. 2(b), an anomalous phenomena occurs, the maximum fidelity achieved for the nuclear spin in the second shell is a little bit larger than that for the nuclear spin in the first shell. That is because the transverse hyperfine interaction provides an additional pathway for the intermediate state $|-, \uparrow\rangle$ to the nuclear-spin polarized state $|0, \downarrow\rangle$. If the noise strength is further suppressed to $\kappa = 1/58$ MHz, the first peak for a nuclear spin with $A_\parallel = -7.5$ MHz rises although more time is required for the evolution.

IV. DISCUSSION AND CONCLUSION

The initialization of the nuclear spin is critical to the subsequent quantum information storage and processing. Facilitated by the strong hyperfine interaction with the electron spin, the nuclear spin in the vicinity of an NV center can be polarized by the swapping of the electron-spin polarization. And the swapping process is intrinsically a quantum-state process via a lossy intermediate state. In this paper, we propose to polarize the $^{13}$C nuclear spin coupled to the electron spin of an NV center through the dark state. Our simulation demonstrates that the nuclear-spin polarization can reach more than 96.7% for the next-next-nearest neighbor site. In the following, we will discuss the feasibility in the experiment and advantages of this proposal.

In theory, in the case of stronger Rabi frequencies $\Omega_1$ and $\Omega_2$, it takes a shorter time $t$ for the nuclear spin to reach the maximum polarization. In our scheme, the magnitudes of $\Omega_1$ and $\Omega_2$ are limited by the selective-excitation condition. The driving frequency $\omega_A$ is set to be in close resonance with the transition $|0, \uparrow\rangle = |-, \downarrow\rangle$, i.e. $\Omega_1 \gtrsim \delta$. On the other hand, the level spacing between $|0, \downarrow\rangle$ and $|-, \downarrow\rangle$ is $D - \gamma_e B_z + A_\parallel/2$. To selectively address the transition between $|0, \uparrow\rangle$ and $|-, \downarrow\rangle$, the Rabi frequency $\Omega_1$ must satisfy the large-detuning condition, i.e. $\Omega_1 \ll \delta + A_\parallel$. In the same way, we can deduce that $\delta - \Delta \ll \Omega_2 \ll A_\parallel - \delta + \Delta$. To be specific, because the hyperfine interaction between the electronic spin and the $^{13}$C nuclear spin in the first shell $A_\parallel$ is 130MHz, the Rabi frequencies can be no more than 13MHz. Furthermore, in order to validate the secular approximation, the magnetic field strength and the hyperfine interaction should fulfill the requirement $D - \gamma_e B_z + \gamma_e B_z - \frac{1}{2}A_\parallel \gg A_\parallel/\sqrt{2}$. In other words, $B_z \leq 673$G for the case with a $^{13}$C nuclear
spin in the first shell.

Compared with Refs. 31, 32, our scheme does not require a specific magnetic field to result in a level anti-crossing in the ground or excited states. Besides, our proposal is not sensitive to the inclination of applied magnetic field. In Refs. 33, 34, the initialization of nuclear spin with fidelity 85% is realized by mapping the electronic spin polarization into the nuclear spin state with a weak hyperfine interaction. Furthermore, our proposal can reach 96% in a weaker hyperfine interaction. Since about 10 repetitions are required in our proposal, it is more convenient than the single-shot readout approach in Refs. 32, 33.

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Appendix A: Quantum Dynamics and Dark State

In the basis of \{0, ↑, −, ↑, 0, ↓\}, the Hamiltonian is written in the matrix form as

\[
H = \begin{pmatrix} 0 & \Omega_1 & 0 \\ \Omega_1 & \omega_1 & \Omega_2 \\ 0 & \Omega_2 & \omega_2 \end{pmatrix},
\]

where

\[
\begin{align*}
\omega_1 &= \delta - i\kappa/2, \\
\omega_2 &= \Delta - i\kappa/2.
\end{align*}
\]

We consider the time evolution of an initial state \( |\psi(0)\rangle = |0, \uparrow\rangle \). At any time, the state reads

\[
|\psi(t)\rangle = u(t)|0, \uparrow\rangle + v(t)|-, \uparrow\rangle + w(t)|-, \downarrow\rangle.
\]

According to Schrödinger equation, we obtain a set of differential equations for the probability amplitudes as

\[
\begin{align*}
iu(t) &= \Omega_1 v(t), \\
i\dot{v}(t) &= \Omega_1 u(t) + \omega_1 v(t) + \Omega_2 w(t), \\
i\dot{w}(t) &= \Omega_2 v(t) + \omega_2 w(t),
\end{align*}
\]

with the initial condition \( u(0) = 1, v(0) = w(0) = 0 \). By Laplace transformation, \( \tilde{\alpha}(p) = \int_0^\infty \alpha(t) e^{-pt} dt \) (\( \alpha = u, v, w \)),

\[
\begin{align*}
i[p\tilde{u}(p) - 1] &= \Omega_1 \tilde{v}(p), \\
p\tilde{v}(p) &= \Omega_1 \tilde{u}(p) + \omega_1 \tilde{v}(p) + \Omega_2 \tilde{w}(p), \\
p\tilde{w}(p) &= \Omega_2 \tilde{v}(p) + \omega_2 \tilde{w}(p).
\end{align*}
\]

or equivalently in the matrix form

\[
\begin{bmatrix} -i p & \Omega_1 & 0 \\ \Omega_1 & \omega_1 - ip & \Omega_2 \\ 0 & \Omega_2 & \omega_2 - ip \end{bmatrix} \begin{bmatrix} \tilde{u}(p) \\ \tilde{v}(p) \\ \tilde{w}(p) \end{bmatrix} = \begin{bmatrix} -i \\ 0 \\ 0 \end{bmatrix}.
\]

We define

\[
\det D \equiv \begin{vmatrix} -i p & \Omega_1 & 0 \\ \Omega_1 & \omega_1 - ip & \Omega_2 \\ 0 & \Omega_2 & \omega_2 - ip \end{vmatrix} = (x_1 - ip)(x_2 - ip)(x_3 - ip),
\]

\[
\det D_1 \equiv \begin{vmatrix} -i & \Omega_1 & 0 \\ 0 & \omega_1 - ip & \Omega_2 \\ 0 & \Omega_2 & \omega_2 - ip \end{vmatrix} = -i(\omega_1 - ip)(\omega_2 - ip) - (i)\Omega_2^2.
\]
where \( x_j \)'s are the eigen energies of Hamiltonian (A1), which will be determined later. And thus we have

\[
\tilde{u}(p) = \frac{\det D_1}{\det D} = -i\frac{(\omega_1 - ip)(\omega_2 - ip) - \Omega^2}{(x_1 - ip)(x_2 - ip)(x_3 - ip)},
\]

(A9a)

\[
\tilde{v}(p) = \frac{\det D_2}{\det D} = -i\frac{\Omega_1(\omega_1 - \omega_2)}{(x_1 - ip)(x_2 - ip)(x_3 - ip)},
\]

(A9b)

\[
\tilde{w}(p) = \frac{\det D_3}{\det D} = -i\frac{\Omega_1\Omega_2}{(x_1 - ip)(x_2 - ip)(x_3 - ip)}.
\]

(A9c)

Furthermore, by inverse Laplace transformation, \( \alpha(t) = (2\pi i)^{-1}\int_\sigma^{\sigma+i\infty} \tilde{\alpha}(p)e^{pt}dp \) \( (\alpha = u, v, w) \), we obtain the probability amplitudes as

\[
u(t) = \sum_{j=1}^3 \frac{(x_j - \omega_1)(x_j - \omega_2) - \Omega^2}{\Omega_1(x_j - x_k)}e^{-ix_jt},
\]

(A10a)

\[
v(t) = \sum_{j=1}^3 \frac{\Omega_1(x_j - \omega_2)}{\Omega_1(x_j - x_k)}e^{-ix_jt},
\]

(A10b)

\[
w(t) = \sum_{j=1}^3 \frac{\Omega_1\Omega_2}{\Omega_1(x_j - x_k)}e^{-ix_jt}.
\]

(A10c)

According to Schrödinger equation, the eigen energies \( x_j \)'s are the solutions to the following equation

\[
x_j^2 - (\omega_1 + \omega_2)x_j + \omega_1\omega_2 - \Omega^2 = \Omega_1^2\omega_2 = 0,
\]

(A11)

where

\[
\Omega^2 = \Omega_1^2 + \Omega_2^2.
\]

(A12)

When \( \omega_2 \) is small, assuming

\[
x_j \simeq x_{0j} + A_j\omega_2,
\]

(A13)

we can obtain the approximate solutions by the perturbation theory. The zeroth-order terms are determined by

\[
x_{0j}[x_{0j}^2 - (\omega_1 + \omega_2)x_{0j} + \omega_1\omega_2 - \Omega^2] = 0,
\]

(A14)

where

\[
x_{01} = 0,
\]

(A15a)

\[
x_{02} = \omega_+,
\]

(A15b)

\[
x_{03} = \omega_-,
\]

(A15c)

\[
\omega_\pm = \frac{1}{2}[(\omega_1 + \omega_2) \pm \sqrt{(\omega_1 - \omega_2)^2 + 4\Omega^2}] = \frac{\Omega^2}{\Omega_1^2} + \omega_\pm.
\]

(A15d)

Therefore, Eq. (A11) can be rewritten as

\[
x(x - \omega_+)(x - \omega_-) + \Omega_1^2\omega_2 = 0.
\]

(A16)

By inserting \( x_1 = A_1\omega_2 \) into Eq. (A16), to the zeroth order of \( \omega_2 \), we obtain

\[
A_1 = -\frac{\Omega_1^2}{\omega_+\omega_-} \simeq \frac{\Omega_1^2}{\Omega^2}.
\]

(A17)

By inserting \( x_2 = \omega_+ + A_2\omega_2 \) into Eq. (A16), to the zeroth order of \( \omega_2 \), we obtain

\[
A_2 = -\frac{\Omega_2^2}{\omega_\pm(\omega_+ - \omega_-)} \simeq -\frac{\Omega_2^2}{2\Omega^2}.
\]

(A18)

By inserting \( x_3 = \omega_\pm + A_3\omega_2 \) into Eq. (A16), to the zeroth order of \( \omega_2 \), we obtain

\[
A_3 = -\frac{\Omega_1^2}{\omega_\pm(\omega_+ - \omega_-)} \simeq -\frac{\Omega_1^2}{2\Omega^2}.
\]

(A19)

The eigen states are

\[
|E_i\rangle = \frac{1}{N_i}\{(|x_i - \omega_1\rangle)(x_i - \omega_2) - \Omega_2^2|0, \uparrow\}
\]

\[+\Omega_1|x_i - \omega_2\rangle|\downarrow, \uparrow\} + \Omega_1\Omega_2|\downarrow, \downarrow\},
\]

(A20)

where the normalization constants are

\[
N_i^2 = \|(x_i - \omega_1)(x_i - \omega_2) - \Omega_2^2\|^2 + |\Omega_1(x_i - \omega_2)|^2
\]

\[+|\Omega_1\Omega_2|^2.
\]

(A21)

In the eigen basis, the time evolution of the initial state \( |\psi(0)\rangle = |0, \uparrow\> \) is

\[
|\psi(t)\rangle = \sum_{j=1}^3 \frac{N_j e^{-i\Omega_jt}}{\prod_{k\neq j}(x_j - x_k)}|E_j\rangle.
\]

(A22)

When \( \omega_1, \omega_2 \ll \Omega_1, \Omega_2 \), to the first order of \( \omega_j \)'s, we have

\[
\omega_\pm = \frac{1}{2}\{(\omega_1 + \omega_2) \pm \sqrt{\Omega^2(1 + \frac{(\omega_1 - \omega_2)^2}{4\Omega^2})}\}
\]

\[\simeq \frac{1}{2}\{(\omega_1 + \omega_2) \pm 2\Omega[1 + \frac{(\omega_1 - \omega_2)^2}{4\Omega^2}]\}
\]

\[\simeq \frac{1}{2}\{(\omega_1 + \omega_2) \pm 2\Omega\}.
\]

(A23)

The eigen energies are approximated to the first order of \( \omega_j \)'s as

\[
x_1 \simeq \Omega_1^2 \frac{\Omega^2}{4[\Omega_1^2 + (\omega_1 + \omega_2)^2]^{3/2}} \omega^2
\]

\[\simeq \frac{\Omega_1^2}{\Omega^2} \omega_2,
\]

(A24a)
In order to obtain nearly-complete polarization, the following conditions should be fulfilled, i.e.

\[ x_2 \simeq \frac{1}{2}(\omega_1 + \omega_2) + 2\Omega - \frac{\Omega_1^2}{(\omega_1 + \omega_2) + 2\Omega} \omega_2 \]

\[ x_3 \simeq \frac{1}{2}(\omega_1 + \omega_2) - 2\Omega + \frac{\Omega_1^2}{(\omega_1 + \omega_2) - 2\Omega} \omega_2 \]

\[ \simeq \Omega + \frac{1}{2}(\omega_1 + \frac{\Omega_1^2}{\Omega^2} \omega_2) \]  \hspace{1cm} (A24b)

\[ \simeq -\Omega + \frac{1}{2}(\omega_1 + \frac{\Omega_1^2}{\Omega^2} \omega_2). \]  \hspace{1cm} (A24c)

Furthermore, the probability amplitudes can be obtained with the coefficients to the zeroth order of \( \omega_j \)'s and the phases to the first order of \( \omega_j \)'s as

\[ u(t) = \frac{(x_1 - \omega_1)(x_1 - \omega_2) - \Omega_1^2}{(x_1 - x_2)(x_1 - x_3)} e^{-ix_1 t} + \frac{(x_2 - \omega_1)(x_2 - \omega_2) - \Omega_1^2}{(x_2 - x_1)(x_2 - x_3)} e^{-ix_2 t} + \frac{(x_3 - \omega_1)(x_3 - \omega_2) - \Omega_1^2}{(x_3 - x_1)(x_3 - x_2)} e^{-ix_3 t} \]

\[ = \frac{(0 - \omega_1)(0 - \omega_2) - \Omega_1^2}{(0 - \omega_1)(0 - \omega_2)} e^{-i\frac{\Omega_1^2}{\Omega^2} \omega_1 t} + \frac{(\omega_1 - \omega_2)(\omega_2 - \omega_2) - \Omega_1^2}{(\omega_1 + 0)(\omega_2 - \omega_2)} e^{-i[\Omega + \frac{\Omega_1^2}{\Omega^2} \omega_2] t} \]

\[ + \frac{\Omega_1^2}{\Omega^2} e^{-i\frac{\Omega_1^2}{\Omega^2} \omega_2 t} + \frac{\Omega_1^2 - \Omega_1^2}{\Omega \Omega} e^{-i[\Omega + \frac{\Omega_1^2}{\Omega^2} \omega_2] t} + \frac{\Omega_1^2}{-2\Omega^2} e^{-i[-\Omega + \frac{\Omega_1^2}{\Omega^2} \omega_2] t} \]

\[ = \frac{\Omega^2}{\Omega} e^{-i\frac{\Omega_1^2}{\Omega^2} \omega_2 t} - i \frac{\Omega^2}{\Omega} e^{-i\frac{1}{2}(\omega_1 + \frac{\Omega_1^2}{\Omega^2} \omega_2) t} \sin \Omega t, \] \hspace{1cm} (A25)

\[ v(t) = \frac{\Omega_1(x_1 - \omega_2)}{(x_1 - x_2)(x_1 - x_3)} e^{-ix_1 t} + \frac{\Omega_1(x_2 - \omega_2)}{(x_2 - x_1)(x_2 - x_3)} e^{-ix_2 t} + \frac{\Omega_1(x_3 - \omega_2)}{(x_3 - x_1)(x_3 - x_2)} e^{-ix_3 t} \]

\[ = \frac{\Omega_1(0 - \omega_2)}{(0 - \omega_1)(0 - \omega_2)} e^{-i\frac{\Omega_1^2}{\Omega^2} \omega_2 t} + \frac{\Omega_1(\omega_1 - \omega_2)}{(0 - 0)(0 + \Omega)} e^{-i[\Omega + \frac{\Omega_1^2}{\Omega^2} \omega_2] t} + \frac{\Omega_1(\omega_1 - \omega_2)}{(-\Omega - 0)(-\Omega - \Omega)} e^{-i[-\Omega + \frac{\Omega_1^2}{\Omega^2} \omega_2] t} \]

\[ = \frac{\Omega_1}{\Omega} e^{-i\frac{1}{2}(\omega_1 + \frac{\Omega_1^2}{\Omega^2} \omega_2) t} \cos \Omega t, \] \hspace{1cm} (A26)

\[ w(t) \simeq \Omega_1 \Omega_2 \left\{ \frac{e^{-i\frac{\Omega_1^2}{\Omega^2} \omega_2 t}}{(0 - \omega_1)(0 - \omega_2)} + \frac{e^{-i[\Omega + \frac{\Omega_1^2}{\Omega^2} \omega_2] t}}{(\omega_1 - 0)(\omega_2 - \omega_2)} + \frac{e^{-i[-\Omega + \frac{\Omega_1^2}{\Omega^2} \omega_2] t}}{(\omega_1 - 0)(\omega_1 - \omega_2)} \right\} \]

\[ \simeq -\frac{\Omega_1 \Omega_2}{\Omega^2} e^{-i\frac{\Omega_1^2}{\Omega^2} \omega_2 t} + \frac{\Omega_1 \Omega_2}{2\Omega^2} e^{-i[\Omega + \frac{\Omega_1^2}{\Omega^2} \omega_2] t} + \frac{\Omega_1 \Omega_2}{2\Omega^2} e^{-i[-\Omega + \frac{\Omega_1^2}{\Omega^2} \omega_2] t} \]

\[ \simeq -\frac{\Omega_1 \Omega_2}{\Omega^2} e^{-i\frac{\Omega_1^2}{\Omega^2} \omega_2 t} - e^{-i\frac{1}{2}(\omega_1 + \frac{\Omega_1^2}{\Omega^2} \omega_2) t} \cos \Omega t. \] \hspace{1cm} (A27)

When \( \Omega t = \pi \),

\[ w(t) = -\frac{\Omega_1 \Omega_2}{2\Omega^2} \left[ e^{-i\frac{\Omega_1^2}{\Omega^2} \omega_2 t} + e^{-i\frac{\Omega_1^2}{\Omega^2} \omega_2} \right] \]

\[ = -\frac{\Omega_1 \Omega_2}{\Omega^2} \left[ e^{-i\pi \frac{\Omega_1^2}{\Omega^2} \Delta} + e^{-i\frac{\Omega_1^2}{\Omega^2} \Delta} \right] \hspace{1cm} (A28) \]

In order to obtain nearly-complete polarization, the two following conditions should be fulfilled, i.e.

\[ e^{-i\frac{\Omega_1^2}{\Omega^2} \Delta} = e^{-i\frac{2\Omega^2}{\Omega^2} \Delta}, \]

\[ 1 \gg e^{-\frac{\Omega_1^2}{\Omega^2} \kappa}, e^{-\frac{\Omega_1^2 + \Omega_2^2}{\Omega^2} \kappa}, \]

or equivalently

\[ \delta = \frac{2\Omega_1^2 - \Omega_2^2}{\Omega^2}, \]

\[ \kappa \ll \frac{\Omega^3}{\pi \Omega_1}, \frac{4\Omega^3}{\pi (\Omega^2 + \Omega_2^2)}. \] \hspace{1cm} (A30a, A30b)
For $\Omega_1 = \Omega_2$, we have
\[
\delta = \frac{1}{2} \Delta, \quad (A31a)
\]
\[
\Omega \gg \frac{3}{8} \pi \kappa. \quad (A31b)
\]
When the above condition is fulfilled, to the first order of $\kappa$, the polarization deviates from the unity as
\[
1 - |w(t)|^2 \simeq 1 - \frac{\Omega_1^2 \Omega_2^2}{\Omega^4} \left\{ e^{-\frac{\pi \Omega_1^2}{2 \Omega^2} \kappa} + e^{-\frac{\pi \Omega_2^2}{2 \Omega^2} \kappa} + 2 \text{Re}\left[ e^{-\frac{\pi \Omega_1^2}{2 \Omega^2} \kappa} \left( e^{i \frac{\pi \Omega_1^2}{2 \Omega^2} \kappa} \right) e^{i \frac{\pi \Omega_2^2}{2 \Omega^2} \kappa} \right] \right\}
\]
\[
= 1 - \frac{\Omega_1^2 \Omega_2^2}{\Omega^4} \left\{ e^{-\frac{\pi \Omega_1^2}{2 \Omega^2} \kappa} + e^{-\frac{\pi \Omega_2^2}{2 \Omega^2} \kappa} + 2 \cos\left(\frac{\pi \Omega_1^2}{\Omega^2} \kappa + \frac{\pi \Omega_2^2}{\Omega^2} \kappa\right) \right\}
\]
\[
\simeq 1 - \frac{\Omega_1^2 \Omega_2^2}{\Omega^4} \left\{ 1 - \frac{\Omega_1^2}{2 \Omega^2} \kappa + 1 - \frac{\Omega_2^2}{2 \Omega^2} \kappa + 2 \left( \frac{\pi \Omega_1^2}{\Omega^2} \kappa - \frac{\pi \Omega_2^2}{\Omega^2} \kappa \right) \right\}
\]
\[
= 1 - \frac{\Omega_1^2 \Omega_2^2}{\Omega^4} \left\{ 1 - \frac{3 \pi \Omega_1^2}{2 \Omega^2} \kappa + 2 \left( \frac{3 \pi \Omega_1^2}{2 \Omega^2} \kappa \right) \right\}
\]
\[
= \frac{3 \pi}{8 \Omega^2} \kappa. \quad (A32)
\]
To the zeroth order of $\omega_j$'s, the eigen states are
\[
|E_1\rangle \simeq \frac{1}{N_1} \left\{ \left[ (0 - \Omega_1)(0 - \Omega_2) - \Omega_2^2 \right] |0, \uparrow\rangle + \Omega_1 (0 - \Omega_2) |-, \uparrow\rangle + \Omega_1 \Omega_2 |-, \downarrow\rangle \right\}
\]
\[
\simeq \frac{\Omega_2}{N_1} (-\Omega_2 |0, \uparrow\rangle + \Omega_1 |-, \downarrow\rangle), \quad (A33a)
\]
\[
|E_2\rangle \simeq \frac{1}{N_2} \left\{ \left[ (\Omega - \Omega_1)(\Omega - \Omega_2) - \Omega_2^2 \right] |0, \uparrow\rangle + \Omega_1 (\Omega - \Omega_2) |-, \uparrow\rangle + \Omega_1 \Omega_2 |-, \downarrow\rangle \right\}
\]
\[
\simeq \frac{\Omega_1}{N_2} (\Omega_1 |0, \uparrow\rangle + \Omega |-, \uparrow\rangle + \Omega_2 |-, \downarrow\rangle), \quad (A33b)
\]
\[
|E_3\rangle \simeq \frac{1}{N_3} \left\{ \left[ (\Omega - \Omega_1)(\Omega - \Omega_2) - \Omega_2^2 \right] |0, \uparrow\rangle + \Omega_1 (\Omega - \Omega_2) |-, \uparrow\rangle + \Omega_1 \Omega_2 |-, \downarrow\rangle \right\}
\]
\[
\simeq \frac{\Omega_1}{N_3} (\Omega_1 |0, \uparrow\rangle + \Omega |-, \uparrow\rangle + \Omega_2 |-, \downarrow\rangle), \quad (A33c)
\]
where the normalization constants are
\[
N_1^2 = \Omega_2^2 \Omega^2, \quad (A34a)
\]
\[
N_2^2 = N_3^2 = 2 \Omega_2^2 \Omega^2. \quad (A34b)
\]
Here $|E_1\rangle$ is the dark state, while the other two are the bright states. Notice that all expanding coefficients in the bright states are the same except there is a sign difference in the expanding coefficients of $|-, \uparrow\rangle$.

In the eigen basis, the time evolution of the initial state $|\psi(0)\rangle = |0, \uparrow\rangle$ is
\[
|\psi(t)\rangle = \frac{\Omega_2 \Omega e^{-\frac{\Omega_2^2}{2 \Omega^2} \kappa t}}{(0 - \Omega)(\Omega + \Omega)} |E_1\rangle + \frac{\sqrt{2} \Omega_1 \Omega e^{-\frac{\Omega_1^2}{2 \Omega^2} \kappa t}}{(\Omega - \Omega)(\Omega + \Omega)} |E_2\rangle + \frac{\sqrt{2} \Omega_1 \Omega e^{-\frac{\Omega_1^2}{2 \Omega^2} \kappa t}}{(-\Omega - \Omega)(\Omega - \Omega)} |E_3\rangle
\]
\[
= - \frac{\Omega_2 e^{-\frac{\Omega_2^2}{2 \Omega^2} \kappa t}}{\Omega} |E_1\rangle + \frac{\sqrt{2} \Omega_1 e^{-\frac{\Omega_1^2}{2 \Omega^2} \kappa t}}{2 \Omega} (e^{-i \Omega t} |E_2\rangle + e^{i \Omega t} |E_3\rangle).
\]
Because there is a sign difference in the expanding coefficients of $|-, \uparrow\rangle$ in $|E_2\rangle$ and $|E_3\rangle$, the probability in $|-, \uparrow\rangle$ vanishes as long as $\Omega t = \pi n$ with $n$ being integer. To summarize, we utilize the dark state and quantum interference to achieve nearly-complete polarization of the nuclear spin.

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