Engineering nuclear spin dynamics with optically pumped nitrogen-vacancy center

Ping Wang,1,2 Jiangfeng Du,1 and Wen Yang2

1Hefei National Laboratory for Physics Sciences at Microscale and Department of Modern Physics, University of Science and Technology of China, Hefei, Anhui 230026, China
2Beijing Computational Science Research Center, Beijing 100084, China

We present a general theory for using an optically pumped diamond nitrogen-vacancy center as a tunable, non-equilibrium bath to control a variety of nuclear spin dynamics (such as dephasing, relaxation, squeezing, polarization, etc.) and the nuclear spin noise. It opens a new avenue towards engineering the dissipative and collective nuclear spin evolution and solves an open problem brought up by the 13C nuclear spin noise suppression experiment [E. Togan et al., Nature 478, 497 (2011)].

PACS numbers: 03.67.Pp, 71.70.Jp, 76.70.Fz, 03.67.Lx

Introduction.—Diamond nitrogen-vacancy (NV) center is a leading platform for quantum computation and nanoscale sensing [2–5]. The NV spin and a few surrounding nuclear spins form a hybrid quantum register [5, 11]. Its coherence time is ultimately limited by the noise from environmental nuclei. This motivates widespread interest in using the NV spin to control the qubit and environmental nuclei through their hyperfine interaction (HFI). In addition to the remarkable success in manipulating a few qubit nuclei, there is increasing interest in controlling the nuclear spin dissipation, e.g., dephasing and relaxation of individual qubit nuclei [13, 14] and dynamic polarization of many environmental nuclei [15, 16]. Intensive experimental efforts have led to dramatic enhancement of the NMR signal [16] for applications in chemistry and biomedicine and the first demonstration of coherence protection by suppressing the nuclear spin noise [19]. This is an important step towards engineering the nuclear spin evolution for coherence protection, nanoscale sensing [2, 20, 22], and long-time storage of quantum information [23, 24].

This prospect, however, could be hindered by our limited understanding of the dissipative nuclear spin dynamics. At present, theoretical treatments are limited to phenomenological or semiclassical modelling [13, 14, 18, 19, 25] or numerical simulation neglecting the NV coherence [16, 17]. The former provides an intuitive picture, but is qualitative. The latter is more accurate, but is limited to a small number of nuclei and may miss important effects due to the NV coherence. Crucially, it is not clear how to efficiently and quantitatively control the nuclear spin dissipation and especially the nuclear spin noise, e.g., the physical mechanism leading to the most impressive observation of Ref. [19], the unconditional suppression of the 13C nuclear spin noise without appreciable polarization, remains unclear. Subsequent noise suppression experiments [26, 27] are based on the simple but challenging approach of completely polarizing all the nuclei or conditioned on measurement-based postselection [28].

Despite the recent experimental progress in controlling the nuclear spin polarization in certain setups [29, 30], a general guidance for the efficient, unconditional control of the nuclear spin dynamics and noise is still lacking.

In this letter, we present a quantum theory for using the NV center to engineer various nuclear spin dynamics and noise. The essential idea is to introduce tunable dissipation into the NV center by optical pumping, so the NV becomes a tunable, dissipative bath for the nuclei. When the NV dissipation is much faster than the NV-induced nuclei dissipation (i.e., the bath being Markovian), we derive a generalized Lindblad master equation for the many-nuclei density matrix, with analytical expressions for the nuclei transition/dephasing rates. They not only allow easy calculation of various nuclear spin dynamics incorporating the NV coherence, but also allow engineering these dynamics (dephasing, relaxation, squeezing, polarization, etc.) and the nuclear spin noise by controlling the NV. This is illustrated by (i) control of nuclear spin relaxation and dephasing, (ii) nuclear spin squeezing, and (iii) suppression of the noise from many 13C nuclei. Case (i) provides a microscopic basis for the phenomenological spin-fluctuator model [13] and experimental observations [2, 31], and a simple method to suppress the nuclear spin dephasing or relaxation. Case (iii) provides a general and efficient way to suppress or amplify the nuclear spin noise and explains the observed 13C nuclear spin noise suppression [19] as a special case.

General theory.—We consider many nuclei \{I_k\} (described by the Hamiltonian \(\hat{H}_N\)) coupled to an optically pumped NV center. We always work in a suitable NV rotating frame and nuclei interaction picture, so the Hamiltonian consists of the time-independent NV part \(\hat{H}_c\), the longitudinal HFI \(\hat{K}\) that commutes with \(\hat{H}_N\), and the transverse HFI \(\hat{V}(t)\) that flips the nuclei:

\[
\hat{\rho}(t) = -i[\hat{H}_c + \hat{K} + \hat{V}(t), \hat{\rho}(t)] + \mathcal{L}_c \hat{\rho}(t),
\]

where \(\mathcal{L}_c \hat{\rho} \equiv \sum_{f_1} \gamma_{f_1} D[|f\rangle \langle f|] \hat{\rho} \) is the NV dissipation in the Lindblad form \(D[\hat{L}] \hat{\rho} \equiv \hat{L} \hat{\rho} \hat{L}^\dagger - \hat{L}^\dagger \hat{L} \hat{\rho} / 2\). Here we focus on NV-induced nuclei dynamics. The direct nuclei interactions and intrinsic nuclei damping can be easily included at the end of the derivation.

To derive a closed description for the many-nuclei state \(\hat{\rho}(t) \equiv \text{Tr}_c \hat{\rho}(t)\), we use the adiabatic approximation [32]...
to eliminate the fast electron motion. We define the many-nuclei basis \( |\mathbf{m}\rangle \) as the common eigenstates of \( \hat{H}_N \) and \( \hat{K} \) with \( \hat{K}|\mathbf{m}\rangle = \hat{K}_m|\mathbf{m}\rangle \), where \( \hat{K}_m \) is an electron operator. The block \( \hat{\rho}_{m,n} \equiv \langle \mathbf{m}|\hat{\rho}|\mathbf{n}\rangle \) obeys

\[
\dot{\hat{\rho}}_{m,n} = \mathcal{L}_{m,n}\hat{\rho}_{m,n} - i [\hat{\rho}_{m,n}, \delta \hat{K}_{m,n}] / 2 - i \langle \mathbf{m}|\hat{V}|\mathbf{n}\rangle,
\]

where \( \delta \hat{K}_{m,n} \equiv \hat{K}_m - \hat{K}_n \) and \( \mathcal{L}_{m,n}(\bullet) \equiv -i[H_e + (\hat{K}_m + \hat{K}_n)/2, \bullet] + \mathcal{L}_e(\bullet) \). Tracing over the electron yields

\[
\dot{p}_{m,n} = -i \text{Tr}_e [\hat{\rho}_{m,n}, \delta \hat{K}_{m,n}] / 2 - i \text{Tr}_e \langle \mathbf{m}|\hat{V}|\mathbf{n}\rangle
\]

for \( p_{m,n} \equiv \langle \mathbf{m}|\hat{\rho}|\mathbf{n}\rangle \). The above equations contain three dissipation time scales: NV dissipation (time scale \( T_e \)) driven by \( \mathcal{L}_{m,n} \); nuclei dephasing (time scale \( T_2 \)) by \( \delta \hat{K}_{m,n} \) fluctuation, and nuclei relaxation (time scale \( T_1 \)) by \( \hat{V}(t) \) fluctuation. Nuclei dissipation much slower than \( T_e \) can be adiabatically singled out. For specificity, we consider \( T_e \ll T_1, T_2 \) and single out the dynamics of \( \hat{\rho} \) on the coarse grained time scale \( \Delta t \gg T_e \).

By treating \( \mathcal{L}_{m,n} \) exactly and \( \delta \hat{K}_{m,n} \), \( \hat{V}(t) \) perturbatively, an application of the adiabatic approximation \([35]\) gives the nuclear spin dynamics order by order \( \hat{p} \equiv \langle \mathbf{m} | \hat{\rho} | \mathbf{n} \rangle \), \( \hat{p}_2 \equiv \langle \mathbf{m} | \hat{\rho} | \mathbf{n} \rangle \), \( \hat{p}_3 \equiv \langle \mathbf{m} | \hat{\rho} | \mathbf{n} \rangle \), \( \ldots \). The first-order dynamics describes nuclear spin precession in the electron Knight fields,

\[
\langle \mathbf{p}(t) \rangle = -i \text{Tr}_e [\hat{K} + \hat{V}(t), \hat{\rho}_0(t)],
\]

equivalent to a renormalization of the nuclei Hamiltonian \( \hat{H}_N \), where \( \hat{\rho}_0(t) = \sum_{m,n} \langle m | \hat{\rho} | n \rangle \hat{P}_{m,n} \) and \( \hat{P}_{m,n} \)

is the normalized electron steady state: \( \mathcal{L}_{m,n} \hat{P}_{m,n} = 0 \). For \( \langle \mathbf{p}|\hat{V}(t)|\mathbf{m}\rangle = \hat{F}_e^{-i\omega t} \),

\[
(\hat{p}_{m,m})_2 = 2 \text{Re} \int_0^\infty e^{\omega \tau} \text{Tr}_e \hat{F}^\dagger e^{\hat{p}_{m,m} \tau} \hat{F} \hat{P}_{m,m} d\tau,
\]

The off-diagonal coherences \( p_{m,n} (m \neq n) \) obeys

\[
(\hat{p}_{m,n})_2 = - \left( \Gamma^{\varphi}_{m,n} + \frac{\sum_p (W_{p,m+n} + W_{p,m-n})}{2} \right) p_{m,n},
\]

\[
\Gamma^{\varphi}_{m,n} \equiv \text{Re} \int_0^\infty \text{Tr}_e \delta \hat{K}_{m,n} e^{\lambda \tau} \delta \hat{K}_{m,n} \hat{P}_{m,n} d\tau,
\]

where \( \delta \hat{K}_{m,n} \equiv \delta \hat{K}_{m,n} - \text{Tr}_e \delta \hat{K}_{m,n} \hat{P}_{m,n} \). The expression for \( W_{p,m-n} \) is involved \([35]\), but it reduces to \( W_{p,m} \) upon neglecting the difference between \( \hat{K}_m \) and \( \hat{K}_n \). The key quantities of our theory, the transition rate \( W_{p,m} \) \([\text{Eq. (4)}]\) and pure dephasing rate \( \Gamma^{\varphi}_{m,n} \) \([\text{Eq. (6)}]\) are obtained by calculating the inverse \( (\mathcal{L}_{m,n} + i\omega)^{-1} \) and \( \mathcal{L}_{m,n}(\bullet) \equiv \lim_{\lambda \to 0}(\mathcal{L}_{m,n} + iv)^{-1} \). Here we notice that if we focus on the dynamics of \( p_{m,n} \) on the time scale \( \Delta t \gg T_1, T_2 \), then we can treat \( \delta \hat{K}_{m,n} \) exactly and still derive Eqs. \( \text{[4]} \) and \( \text{[6]} \), with \( \mathcal{L}_{p,m} \) in Eq. \( \text{[4]} \) replaced with \( \mathcal{L}_{p,m}^\text{id} \equiv \mathcal{L}_{p,m}(\bullet) - i (\bullet, \delta \hat{K}_{m,n}) / 2 \).
we can always tune the nuclear spin quantization axis \( \mathbf{e}_z \propto \mathbf{F} = (g | \hat{P} | g)a_0 + (e | \hat{P} | e)a_0 - \gamma_N < \mathbf{B} > \) to \( \mathbf{e}_z = \mathbf{a}_y - \mathbf{a}_x \) such that \( \Gamma_\varphi = 0 \) (\( \Gamma_1 = 0 \)). Interestingly, the sum rule \( \Gamma_\varphi + 2 \Gamma_1 \propto (\mathbf{a}_y - \mathbf{a}_x)^2 \) suggests that reducing \( \Gamma_\varphi \) (\( \Gamma_1 \)) inevitably increases \( \Gamma_1 \) (\( \Gamma_\varphi \)) and it is impossible to suppress \( \Gamma_\varphi \) and \( \Gamma_1 \) simultaneously, unless the NV states are tuned such that \( \mathbf{a}_y = \mathbf{a}_x \). For more NV levels, analytical results are no longer available, but our general theory is still applicable.

**Nuclear spin squeezing.**—Here we explore \(^{13}\text{C}\) nuclear spin squeezing \(^{33}\) by engineering the first-order evolution Eq. \(^{2}\). With a magnetic field \( \mathbf{B} \mathbf{e}_z \) to quantize all \(^{13}\text{C}\) nuclei along \( \mathbf{e}_z \parallel \mathbf{N} \)-axis and a microwave to couple the NV ground states \( |0\rangle \) and \( |+1\rangle \) with detuning \( \Delta \), the rotating frame Hamiltonian consists of \( \hat{H}_z = \left( \Omega_R/2 \right) (|+1\rangle \langle 0| + h.c.) + \Delta |+1\rangle \langle +1| + \hat{K} = \hat{h}_1 + |+1\rangle \langle +1| \) and \( \hat{K} = \sum_n (|+1\rangle \langle 0| + 1) \cdot \mathbf{A}_n \cdot \mathbf{I}_n \approx \sum_n a_n |I_{n,z}\rangle \) comes from the dipolar HFI with \(^{13}\text{C}\) nuclei and \( a_n \equiv (|+1\rangle \langle 0| + 1) \cdot \mathbf{A}_n \cdot \mathbf{e}_z \). To introduce tunable dissipation, in the steady state \( |\gamma, +1\rangle \) (with rate \( \gamma \)) to orbital excited state \( |+, +1\rangle \) which decays back to \( |+, +1\rangle \) (with rate \( \gamma \)), or to a singlet \( |S\rangle \) (with rate \( \gamma_{1c} \)) and then to \( |0\rangle \) (with rate \( \gamma \)). This creates a unidirectional transition from \( |+, +1\rangle \) to \( |0\rangle \). The transition rate \( \gamma_1 \) is tunable from \( R_{\gamma_1}/(\gamma + R) \) (small \( R \)) to \( \gamma_3 \) (large \( R \)). We define the nuclear spin basis \( \{|m\}\rangle \) as eigenstates of \( \hat{h} \) with eigenvalues \( \{\gamma_m\} \). For \( \gamma_3 \gg |\gamma_m - \gamma_0| \), Eq. \(^{2}\) gives \( \langle \hat{P} | \mathbf{m} \rangle \approx -i \mathbf{m} \cdot \left( \hat{h}_1 (\hat{h}_1 + \hat{h}_0) / \hbar \right) \) |m\rangle \), where \( \hat{P} \) is the polarization of a r.f. pulse), the evolution under the nonlinear Hamiltonian \( \hat{h}_1 \approx \hat{h}_1 (\hat{h}_0) / h \) could lead to nuclear spin squeezing, even for non-uniform coupling \( \{a_n\} \). Taking \( \alpha_n = \alpha \) for an estimate, the characteristic squeezing time \(^{33}\) for \( N \) nuclei is \( \gamma_0^{(N)} = |\hat{P} (0) | N a^2 |^{-1} \). The maximal \( N \)-nuclear collective dephasing rate \( \Gamma_\varphi \) obtained analogous to Eq. \(^{2}\). For \( \Delta = 4 \Omega_R = 2 \gamma_1 \), we have \( \gamma_0^{(N)} = N \gamma_0^{(1)} \approx 100 / 100 \), suggesting significant squeezing for \( N \approx 100 \) nuclei without appreciable dephasing.

**Controlling nuclear spin noise.**—Here we consider continuous pumping the NV to control the noise from many \(^{13}\text{C}\) nuclei coupled to the NV via dipolar HFI \( \sum_{n=1}^{13} \mathbf{S} \cdot \mathbf{A}_n \cdot \mathbf{I}_n \). To focus on noise control, we neglect the squeezing effect, so Eq. \(^{2}\) gives \( \langle \hat{P} | \mathbf{m} \rangle \approx -i \mathbf{m} \cdot \left( \hat{h}_1 (\hat{h}_1 + \hat{h}_0) / \hbar \right) \) |m\rangle \). Here \( \mathbf{b}_n \equiv \left( \hat{S} \mathbf{P} \right) \cdot \mathbf{A}_n - \gamma_N \mathbf{B} \) defines a local coordinate \( \{\mathbf{e}_n, \mathbf{e}_n, \mathbf{e}_n, \mathbf{e}_n, \mathbf{e}_n\} \) for \( \mathbf{I}_n \), where \( \mathbf{e}_n, \mathbf{e}_n \propto \mathbf{b}_n \) and \( \mathbf{P} \) is the steady NV state in the absence of \(^{13}\text{C}\) nuclei. We decompose the dipolar HFI into \( \mathbf{K} \equiv \mathbf{S} \cdot \mathbf{h} \approx \mathbf{S}_z \hat{h}_z \) (\( \mathbf{e}_z \) along N-V axis) and the remaining part \( \mathbf{V} \), where \( \mathbf{h} \equiv \sum_n (\mathbf{A}_n \cdot \mathbf{e}_n) \mathbf{I}_n \). The \( \mathbf{K} \) term not only allows the thermal fluctuation of \( \hat{h}_z \) to rapidly decohere the NV spin, but also allows the nuclear spin magnetometer to monitor and control the slow \( \hat{h}_z \) fluctuation by engineering the following feedback loop \(^{12}\). Here for simplicity we take \( \mathbf{A}_n = \mathbf{A} \), so that \( \hat{h}_z = a \sum_n \mathbf{I}_n \mathbf{z} \leq N \hat{h}_{\text{MAX}} \equiv a N / 2 \) and \( a \equiv \mathbf{e}_z \cdot \mathbf{A}_n \cdot \mathbf{e}_n \mathbf{z} \), although the physical mechanism is general: (i) \( \mathbf{K} = \hat{S} \mathbf{h}_z \), the NV spin \( \hat{S} \) monitors the fluctuation of \( \hat{h}_z \) and records its instantaneous value \( h \) into the NV steady state \( \mathbf{P}(h) \), as determined by \( \mathbf{P}(h) = \hat{S} \mathbf{h}_z \); (ii) Where \( \mathbf{V} \), the NV state \( \mathbf{P}(h) \) flips each \(^{13}\text{C}\) with rates \( \hat{W}_h \mathbf{P}(h) \) and \( \hat{W}_h \mathbf{P}(h) \) [given by Eq. \(^{3}\) with \( \mathbf{P}_m \mathbf{m} \) replaced by \( \mathbf{P}(h) \)] and drives \( \hat{h}_z \) from \( h \) to the steady-state value \( \mathbf{H}(h) \equiv \mathbf{N} \sum_n \mathbf{I}_n \mathbf{z} \), where \( \mathbf{I}_n \mathbf{z} = (1/2) [\mathbf{W}_{h} \mathbf{P}(h) - \mathbf{W}_{h} \mathbf{P}(h)] \). For example, to lock \( \hat{h}_z \) to a pre-defined value \( h_{\text{PRE}} \), we can design the feedback loop such that \( \mathbf{H}(h_{\text{PRE}}) = \mathbf{P}(h_{\text{PRE}}) \) and the derivative \( \mathbf{H}',(h_{\text{PRE}}) < 0 \), i.e., the NV flips the nuclei to decrease (increase) \( \hat{h}_z \) upon detecting \( \hat{h}_z > h_{\text{PRE}} \) (\( \hat{h}_z < h_{\text{PRE}} \)). To describe this feedback loop, we quantify the \( \hat{h}_z \) noise by the width of the \( \hat{h}_z \) distribution \( \mathbf{P}(\hat{h}) \equiv \mathbf{P}(\hat{h} - \hat{h}_z) \mathbf{P} \) in the \( N \)-nuclear steady state \( \mathbf{P} \) and apply the nuclear spin feedback theory \(^{12}\) to Eq. \(^{3}\) and obtain

\[
\mathbf{P}(\hat{h}) \propto \frac{e^{-(\hat{h}_z + \hat{h}_z)^2 / 2(\sigma^2)^2}}{|\mathbf{W}_{h} \mathbf{P}(h) - \mathbf{W}_{h} \mathbf{P}(h)| - h \mathbf{H}(h) / h_{\text{MAX}}^2}, \quad \text{(9)}
\]

with \( h \equiv \mathbf{H}(h) \) the most probable value of \( h \), \( \sigma^2 = \gamma_N > \langle (\hat{h}_z^2)^2 - h^2 / h_{\text{MAX}}^2 \rangle [1 - \mathbf{H}'(h)] \rangle^{-1} \), and \( \gamma_N = \gamma / \sqrt{N} / 2 \) the thermal fluctuation of \( \hat{h}_z \). Equation \(^{9}\) summarizes three ways to suppress the \( \hat{h}_z \) noise. (i) Narrow \( e^{-h_z^2 / (2\sigma^2)^2} \)
by high nuclear polarization, e.g., $h_z/h_{\text{max}} \approx \pm 90\%$ reduces $\sigma$ by a factor of 2. (ii) Narrow $e^{-(h-h_z)^2/(2\sigma^2)}$ by strong negative feedback $H^2(h_z) \ll -1$. This provides a general, measurement-free scenario to control the $h_z$ noise by operating the NV as a magnetometer: any scheme in which the NV steady state $P(h)$ and hence the NV-induced steady-state nuclear polarization $H(h)$ is sensitive to the value $h$ of $h_z$ could significantly suppress or amplify the $h_z$ noise. A possible scheme is to use very weak optical pumping at the NV ground state anticrossing to polarize $^{13}$C nuclei without significantly degrading the NV sensitivity $\Delta h \sim$ NV linewidth, ultimately limited to $\gtrsim 1/T_{2,\text{NV}}$ by the true NV dephasing time $T_{2,\text{NV}}$ (not the inhomogeneous dephasing time $T_{2,\text{NV}}$). For $\Delta h < h_{\text{max}}$, we can tune $h_z$ to the region with the most negative $H^2(h_z) \sim -h_{\text{max}}/\Delta h$ to reduce $\sigma$ from $\sigma_{\text{th}} = a/\sqrt{N}/2$ to an $N$-independent value $\sigma \sim \sqrt{a\Delta h}$. For an estimate, we take $h_{\text{max}} = 1$ MHz to obtain the noise reduction factor $\sigma_{\text{th}}/\sigma = \sqrt{h_{\text{max}}}/\Delta h \approx 10$ (for $\Delta h = 10$ kHz) or 30 (for $\Delta h = 1$ kHz).

The third approach to suppressing $h_z$ noise is (iii) to generate a sharp dip in the denominator of Eq. (9), e.g., by $h_z$-dependent coherent population trapping (CPT) [43]. Now we show that this mechanism leads to the first observation of $^{13}$C nuclear spin noise suppression [19]. The setup of Ref. [19] consists of a Λ system ($|\pm\rangle$ and $|\Omega\rangle$) and a two-level system ($|0\rangle$ and $|E_g\rangle$), both under resonant pumping. Relevant processes are shown in Fig. (1a) and the NV Hamiltonian can be found in Ref. [19] or [45]. The NV-nuclei coupling includes the contact HFI $(A_g S_y + A_s S_y) \cdot I_0$ with the $^{14}$N nucleus $I_0$ and the dipolar HFI $\sum_{n=1}^N S \cdot A_n \cdot I_n$ with the $^{13}$C nuclei $\{I_n\}$, where $S_g (S_s)$ is the NV ground (excited) state spin and $S \equiv S_g + S_s + \cdots$ (the electron Knight fields on $I_0$ and $I_n$ are along $e_z \parallel$ N-V axis and $e_z \cdot A_n = a_n e_{n,z}$, respectively. So we define the local coordinate $(e_{n,x}, e_{n,y}, e_{n,z})$ and decompose the HFI into $\hat{K} = \hat{S}_g \cdot \hat{h}_z$ and the remaining part $\hat{V}$, where $\hat{h}_z \equiv A_g \hat{I}_{0,z} + \sum_n a_n \hat{I}_{n,z}$. We define the nuclei basis $|\tilde{m}\rangle \equiv |m_0\rangle \otimes_{n=1}^N |m_n\rangle$ as the product of eigenstates of each nucleus: $\hat{I}_{0,z}|m_0\rangle = m_0|m_0\rangle$ and $\hat{I}_{n,z}|m_n\rangle = m_n|m_n\rangle$. Then we calculate the transition rates from Eq. (4) (the $^{13}$C nuclear spin flip by the NV ground state has no Fermi golden rule part, only the coherent part $W_{\text{coh}}^{\text{pump}}$ contributes) and solve Eq. (9) numerically to obtain the steady-state nuclear spin populations $\{n_{m,m}\}$. The intrinsic $^{13}$C-$^{13}$C interaction and $^{13}$C relaxation is included as a phenomenological depolarization rate $\gamma_C$ for each $^{13}$C nucleus. The calculated $^{14}$N population on $|m=0\rangle$ and the relevant time scale $\sim 200 \mu$s (vs. experimental value $\sim 353 \pm 34 \mu$s) agree reasonably with the experiment [Fig. (1b)]. We also confirm that the observed decrease of the population at large $\Omega_A$ arises from the off-resonant excitation to $|A_2\rangle$, as expected in Ref. [19]. An impressive observation [19] is the suppressed $\hat{h}_z$ noise from $^{13}$C nuclei in the absence of appreciable $^{13}$C polarization, manifested as the narrowed CPT dip of the NV fluorescence. Using realistic and experimental parameters, we obtain the steady nuclear populations $\{n_{m,m}\}$ at the preparation magnetic field and use them to calculate the unconditional and post-selected population on $|E_g\rangle$ at the readout magnetic field [35]. When normalized to unity at large readout magnetic field $B_{re} = \mu_B e / (g_\mu_B B)$, the results agree reasonably with the experimental fluorescence [Fig. (1c)].

Finally we use Eq. (9) to analyze qualitatively how the NV detects and suppresses $h_z$ noise in the CPT experiment [19]. Given an instantaneous value $h_z$, the NV rapidly records $h$ as a two-photon detuning in the NV steady-state $P(h)$. Our calculation shows that the NV-induced nuclei flip always yields vanishing steady-state polarization $H(h) = 0$, so $h_z = 0$, $\sigma = \sigma_{\text{th}}$, and Eq. (9) gives $p(h) \propto e^{-h_z^2/(2\sigma_{\text{th}}^2)}(W_{\uparrow\uparrow}(h)+W_{\downarrow\downarrow}(h))^{-1}$. The key is that the nuclei flip rates $W_{\uparrow\downarrow}(h), W_{\downarrow\uparrow}(h) \propto |E_g\rangle$ population [35], which has a sharp dip at the two-photon resonance $h = 0$. This generates a sharp peak in $p(h)$ and hence suppresses the fluctuation of $h_z$. Further analysis shows that at $h = 0$, off-resonant excitation to $|A_2\rangle$ gives rise to non-vanishing $|E_g\rangle$ population and hence $^{13}$C spin flip that fundamentally limit the noise suppression efficiency. To avoid this limitation, a possible scheme is to exploit the strain-induced non-vanishing $|E_g\rangle/S_z|E_g\rangle$ and hence the term $|E_g\rangle (I_z + \hat{L}_z)$ of the dipolar HFI. In a magnetic field that quantizes $^{13}$C nuclei along the N-V axis, a negative feedback is expected to significantly suppress the $h_z$ noise without being limited by off-resonant excitation to $|A_2\rangle$.

To summarize, we have presented a quantum theory for using an optically pumped NV center as a tunable bath to engineer various dissipative and collective nuclear spin dynamics, as illustrated by the control of the nuclear spin dephasing, relaxation, and squeezing. It also reveals a general and efficient way to control the nuclear spin noise and clarifies the physical mechanism leading to the first observation of nuclear spin noise suppression [19]. Apart from NV centers, our theory can be readily applied to other quantum information platforms such as quantum dots and defect centers to engineer $^{13}$C-$^{13}$C interaction and $^{13}$C relaxation is included as a phenomenological depolarization rate $\gamma_C$ for each $^{13}$C nucleus. The calculated $^{14}$N population on $|m=0\rangle$ and the relevant time scale $\sim 200 \mu$s (vs. experimental value $\sim 353 \pm 34 \mu$s) agree reasonably with the experiment [Fig. (1b)]. We also confirm that the observed decrease of the population at large $\Omega_A$ arises from the off-resonant excitation to $|A_2\rangle$, as expected in Ref. [19]. An impressive observation [19] is the suppressed $\hat{h}_z$ noise from $^{13}$C nuclei in the absence of appreciable $^{13}$C polarization, manifested as the narrowed CPT dip of the NV fluorescence. Using realistic and experimental parameters, we obtain the steady nuclear populations $\{n_{m,m}\}$ at the preparation magnetic field and use them to calculate the unconditional and post-selected population on $|E_g\rangle$ at the readout magnetic field [35]. When normalized to unity at large readout magnetic field $B_{re} = \mu_B e / (g_\mu_B B)$, the results agree reasonably with the experimental fluorescence [Fig. (1c)].

The authors thank Nan Zhao and L. J. Sham for helpful discussions. This work was supported by NSFC (Grant No. 11274036 and No. 11322542) and the MOST (Grant No. 2014CB848700).
Supplementary materials for “Engineering nuclear spin dynamics with optically pumped nitrogen-vacancy center”

Ping Wang,1,2 Jiangfeng Du,1 and Wen Yang2
1Hefei National Laboratory for Physics Sciences at Microscale and Department of Modern Physics, University of Science and Technology of China, Hefei, Anhui 230026, China
2Beijing Computational Science Research Center, Beijing 100084, China

Section I provides a detailed derivation of Eqs. (2)-(6) of the main text. Sec. II provides a perturbative, explicit expression for the nuclear spin transition rate \( W_{p-m} \), which can be easily applied to a given experimental setup. Sec. III details the NV Hamiltonian, NV-induced nuclear spin transition rates, and calculation of NV fluorescence under the CPT condition [7].

I. DERIVATION OF NUCLEAR SPIN EQUATIONS OF MOTION

The starting point is

\[
\dot{\rho}_{m,n}(t) = -i \frac{[\delta \hat{K}_{m,n}, \hat{\rho}]}{2} - i \langle \hat{\mathcal{V}}(t), \hat{\rho}(t) \rangle \mathbf{n},
\]

\[ \tag{1} \]

\[
\dot{\rho}_{m,n}(t) = -i \text{Tr}_e \left[ \frac{[\delta \hat{K}_{m,n}, \hat{\rho}]}{2} - i \langle \hat{\mathcal{V}}(t), \hat{\rho}(t) \rangle \mathbf{n} \right].
\]

\[ \tag{2} \]

On the coarse grained time scale \( T_c \ll \Delta t \ll T_1, T_2 \), we treat \( \langle \mathbf{p}_{m,n}(t) \rangle \) as slow variables, others as fast variables, and \( \delta \hat{K}_{m,n} \) and \( \hat{\mathcal{V}}(t) \) as first-order small quantities. Correspondingly, we decompose \( \hat{\rho}(t) = \hat{\rho}^{(0)}(t) + \hat{\rho}^{(1)}(t) + \cdots \), where \( \hat{\rho}^{(k)}(t) \) is a \( k \)-th order small quantity. Since we always work in the nuclear spin interaction picture, the zeroth-order dynamics vanishes: \( \langle \hat{\rho} \rangle_0 = 0 \).

The first-order dynamics \( \langle \hat{\rho}^{(0)}_{m,n} \rangle \) [Eq. (2) of the main text] is obtained from Eq. (2) by replacing \( \hat{\rho}(t) \) with \( \hat{\rho}^{(0)}(t) = \sum_{m,n} \langle \mathbf{m} | \hat{\rho}^{(0)}_{m,n} | \mathbf{n} \rangle \), obtained by coarse-graining

\[ \hat{\rho}^{(0)}_{m,n}(t) = \mathcal{L}_{m,n} \hat{\rho}^{(0)}_{m,n}(t) \]

for an interval \( T_c \ll \Delta t \ll T_1, T_2 \). Here \( \hat{P}_{m,n} \) is the normalized electron steady state determined by \( \mathcal{L}_{m,n} \hat{P}_{m,n} = 0 \). The second-order dynamics \( \langle \hat{\rho}^{(1)}_{m,n} \rangle \) is obtained from Eq. (2) by replacing \( \hat{\rho}(t) \) with \( \hat{\rho}^{(1)}(t) \), the solution to

\[ \langle \hat{\rho}^{(0)}_{m,n} \rangle + \langle \hat{\rho}^{(1)}_{m,n} \rangle = \mathcal{L}_{m,n} \langle \hat{\rho}^{(1)}_{m,n} \rangle = -i \frac{[\delta \hat{K}_{m,n}, \hat{\rho}^{(0)}_{m,n}]}{2} - i \langle \hat{\mathcal{V}}(t), \hat{\rho}^{(0)}_{m,n} \rangle \mathbf{n}, \]

where \( \langle \hat{\rho}^{(0)}_{m,n} \rangle = \hat{P}_{m,n} \langle \hat{\rho}^{(0)}_{m,n} \rangle \) comes from the first-order evolution \( \langle \hat{\rho}^{(0)}_{m,n} \rangle \). Substituting into the above equation and coarse-graining for an interval \( 1/\gamma_e \ll \Delta t \ll T_1, T_2 \) gives

\[ \hat{\rho}^{(1)}_{m,n} = i \mathcal{L}_{m,n} \langle \hat{\rho}^{(1)}_{m,n} \rangle \]

\[ + i \sum_{p} \mathcal{L}_{m,n} \langle \mathbf{m} | \hat{\rho}^{(0)}_{m,n} \rangle \mathbf{n} = \frac{1}{2} \left[ \mathcal{L}_{m,n} \langle \hat{\rho}^{(0)}_{m,n} \rangle \delta \hat{K}_{m,n} \right] - i \langle \mathcal{V}(t), \hat{\rho}^{(0)}_{m,n} \rangle \mathbf{n}, \]

where \( \langle \hat{\rho}^{(0)}_{m,n} \rangle = \hat{P}_{m,n} \langle \hat{\rho}^{(0)}_{m,n} \rangle \) is defined by Eq. (2) of the main text and \( \langle \mathcal{V}(t), \hat{\rho}^{(0)}_{m,n} \rangle \mathbf{n} \). Then replacing \( \hat{\rho}(t) \) with \( \hat{\rho}^{(1)}(t) \) in Eq. (2) gives the desired second-order nuclear spin dynamics. For \( \mathbf{m} = \mathbf{n} \), neglecting the coupling of \( \rho_{m,m} \) to other \( \rho_{p,q} \) (\( p \neq q \)), which amounts to neglecting the small second-order corrections to \( \langle \mathcal{V}(t) \rangle \) and electron-mediated nuclear spin interactions (they induce nuclear spin diffusion and depolarization, which can be included phenomenologically at the end of the derivation), we obtain Eq. (3) of the main text. The transition rate

\[ \dot{W}_{p-m} \equiv -2 \text{Re} \text{Tr}_e \langle \mathbf{m} | \mathcal{V}_{p,m} | \mathbf{m} \rangle \langle \mathbf{m} | \hat{\rho}^{(0)}_{p,m} \rangle - i \langle \hat{\mathcal{V}}_{p,m} \rangle \mathbf{n} \mathbf{n} \mathbf{n} \mathbf{n}, \]

Using \( \mathcal{L}_{p,m} + i \omega_{p,m} \) \( \hat{P}_{p,m} = \hat{P}_{p,m} / i \omega_{p,m} \) and \( P_{p,m} \approx P_{m,m} \) (\( \mathbf{p} \) and \( \mathbf{m} \) differs only by a single nuclear spin flip), we see that the term \( \propto \mathcal{L}_{p,m} + i \omega_{p,m} \) \( \hat{P}_{p,m} \) is \( (2 / i \omega_{p,m}) \text{Im} \langle \mathbf{m} | \hat{\rho}^{(0)}_{p,m} \rangle \mathbf{n} \mathbf{n} \mathbf{n} \mathbf{n} \approx 0 \) and hence recover Eq. (4) of the main text. For \( \mathbf{m} \neq \mathbf{n} \), neglecting the coupling of \( \rho_{m,n} \) to other variables and second-order self-energy corrections, which amounts to neglecting the small second-order corrections to \( \langle \mathcal{V}(t) \rangle \) and electron-mediated nuclear spin interactions, we obtain Eqs. (5) and (6) of the main text, where

\[ \dot{W}_{p-m} \equiv -2 \text{Re} \text{Tr}_e \langle \mathbf{m} | \mathcal{V}_{p,m} | \mathbf{m} \rangle \langle \mathbf{m} | \hat{\rho}^{(0)}_{p,m} \rangle - i \langle \hat{\mathcal{V}}_{p,m} \rangle \mathbf{n} \mathbf{n} \mathbf{n} \mathbf{n}, \]
II. PERTURBATIVE, EXPLICIT EXPRESSION FOR $W_{p-m}$

If $\langle p| \hat{V}(t)|m \rangle$ has multiple frequencies that are widely separated compared with the nuclei transition/dephasing rates, then $W_{p-m}$ is the sum of contributions from different frequency components. So we consider the case $\langle p| \hat{V}(t)|m \rangle = \hat{P} e^{-i \omega t}$ with a single frequency. Equation (4) of the main text gives $W_{p-m} = -2Re Tr_p F^2 \hat{G} \hat{P}$, where $\hat{G} \equiv (i \omega + \hat{L})^{-1}$, $\hat{P} \equiv \hat{P}_{m,m}$, and $\mathcal{L} \equiv \mathcal{L}_{p,m}$. The key is to calculate $\mathcal{G}$ perturbatively by dividing $\mathcal{L}(\bullet) \equiv -i[\hat{H}, \bullet] + \mathcal{L}_c(\bullet)$ into the unperturbed part

$$\mathcal{L}^u(\bullet) \equiv -i[\hat{H}^u, \bullet] - \frac{1}{2} \sum_i \Gamma_i [\langle i|, \bullet] + \sum_i y_{i0} \langle i| \langle i| i \rangle| i \rangle$$

and the perturbation

$$\mathcal{L}^{np}(\bullet) \equiv -i[\hat{H}^{np}, \bullet] + \sum_{f \neq i} \gamma_{fi} \langle f| \langle f| i \rangle| i \rangle,$$

where $\hat{H}^u = \sum_i \epsilon_i | i \rangle \langle i |$ ($\hat{H}^{np}$) is the diagonal (off-diagonal) part of $\hat{H}$ and $\Gamma_i \equiv \sum_f \gamma_{fi}$. When $||\mathcal{L}^u + i \omega|| \gg ||\mathcal{L}^{np}||$, an explicit expression for $W_{p-m}$ is obtained by replacing $\hat{G}$ by $\mathcal{G}^u - \mathcal{G}^u \mathcal{L}_c^{np} \hat{L}^u$ and using $\mathcal{G}^u [k] \langle j| \equiv (\mathcal{L}^u + i \omega)^{-1}[k] \langle j| = [k] \langle j| (i \epsilon_{k,l})$ with $z_{k,l} = \epsilon_k - \epsilon_j - i(\Gamma_k + \Gamma_j - 2y_{k,l} \delta_{k,l})/2$. For $\hat{P} = \sum_{f \neq i} \hat{V}_{fi} / \langle f| i \rangle$, we obtain $W_{p-m}$ as the sum of the golden rule part

$$W_{p-m}^{golden} = 2 \sum_{i,j} \text{Im} \left( V_{i,j} V_{f,i} P_{i,f} \right) \left( \Delta \right), \tag{3}$$

and the coherent part

$$W_{p-m}^{coh} = 2 \text{Im} \left( \sum_{i,j} \left( H_{i,j}^u V_{i,f} V_{f,i} - V_{f,i} V_{i,j} H_{i,j}^u \right) \right) \left( \Delta \right), \tag{4}$$

where $O_{i,j} \equiv \langle i| \hat{O} |j \rangle$.

III. NV HAMILTONIAN, NUCLEAR SPIN TRANSITION RATES, AND NV FLUORESCENCE UNDER CPT

A. NV Hamiltonian under CPT

The NV Hamiltonian for the coherent population trapping (CPT) experiment has been discussed in [? ]. Here we reproduce it with greater detail using our own notations. The strain effect is neglected as we have confirmed that it produces a small influence on the nuclear spin polarization and noise suppression. The NV states of relevance include 3 ground triplet states $|0\rangle$ (energy $\epsilon_0$, $| \pm \rangle$ (energy $D_{ps}$), and 6 excited triplet states $| E_1 \rangle, | E_2 \rangle$ (energy $\epsilon_{E_1} = \epsilon_{E_2}$), $| E_1 \rangle, | E_2 \rangle$ (energy $\epsilon_{E_1} = \epsilon_{E_2}$), $| A_1 \rangle$ (energy $\epsilon_{A_1}$), $| A_2 \rangle$ (energy $\epsilon_{A_2}$), and one metastable singlet $| S \rangle$ (energy $\epsilon_S$). A linearly polarized laser with electric field $E_1 e^{-i \omega t} / 2 + c.c.$ and frequency $\omega_1 = \epsilon_{A_1} - D_{ps}$ resonantly excites the ground states $| \pm \rangle$ to the excited state $| A_1 \rangle$ and, at the same time, off-resonantly excited $| \pm \rangle$ to the excited state $| A_2 \rangle$ with detuning $\Delta = \epsilon_{A_2} - \epsilon_{A_1}$. Another linearly polarized laser with electric field $E_2 e^{-i \omega t} / 2 + c.c.$ and frequency $\omega_2 = \epsilon_{E_2}$ resonantly excites $| 0 \rangle$ to $| E_1 \rangle$. The relevant optical transition matrix element of the electric dipole operator $\hat{d} \equiv -e \hat{r}$ are $\langle A_1 | \hat{d} | \pm \rangle = \pm d_{a,E} e_{\pm} / (2 \sqrt{2})$, $\langle A_2 | \hat{d} | \pm \rangle = \pm d_{b,E} e_{\pm} / (2 \sqrt{2})$, and $\langle E_1 | \hat{d} | 0 \rangle = d_{a,E} e_{\pm} / \sqrt{2}$, where $e_{\pm} \equiv e_{\pm} \pm ie_{\pm}$ and $d_{a,E}$ is the reduced matrix element of the electric dipole moment [? ]. Thus the laser coupling Hamiltonian

$$\hat{H}_L(t) = \frac{\Omega E}{2} (e^{-i \phi} \hat{\sigma}_{A_1,1} + e^{-i \phi} \hat{\sigma}_{A_1,-1} + i e^{i \phi} \hat{\sigma}_{A_1,1} + i e^{i \phi} \hat{\sigma}_{A_1,-1}) e^{-i \omega t} + \frac{\Omega E}{2} e^{-i \omega t} \hat{\sigma}_{E_1,0} + h.c.,$$

where we have defined $\hat{\sigma}_{ij} \equiv | i \rangle \langle j |$, $\Omega E = E_2 \cdot \langle E_2 | \hat{d} | 0 \rangle = E_2 d_{a,E} / \sqrt{2}$, and $\Omega A \hat{\sigma}_{ij} \equiv E_1 \cdot \langle A_1 | \hat{d} | 0 \rangle = E_2 d_{a,E} (E_{1,x} + i E_{1,y}) / (2 \sqrt{2})$. Defining the bright state $| b \rangle = (e^{-i \phi} + 1 - e^{i \phi} - 1) / \sqrt{2}$ and dark state $| d \rangle = (e^{-i \phi} + 1 + e^{i \phi} - 1) / \sqrt{2}$, the laser coupling Hamiltonian simplifies to

$$\hat{H}_L(t) = \frac{\Omega A}{2} (\hat{\sigma}_{A_1,b} + i \hat{\sigma}_{A_1,d}) e^{-i \omega t} + \frac{\Omega E}{2} e^{-i \omega t} \hat{\sigma}_{E_1,0} + h.c..$$
In the rotating frame $|\psi(t)\rangle$ connected to the laboratory frame $|\psi_{\text{lab}}(t)\rangle$ via $|\psi(t)\rangle = e^{i\hat{H}_0 t}/|\psi_{\text{lab}}(t)\rangle$ with $\hat{H}_0 = \epsilon E_e (\hat{\sigma}_{E_e,E_s} + \hat{\sigma}_{E_s,E_e}) + \epsilon E_s (\hat{\sigma}_{E_e,E_s} + \hat{\sigma}_{E_s,E_e}) + \epsilon A_1 (\hat{\sigma}_{A_1 A_1} + \hat{\sigma}_{\bar{A}_1 A_1} + \hat{\sigma}_{S_S} S_S + D_{gs}(\hat{\sigma}_{1,1} + \hat{\sigma}_{1,-1})), the NV Hamiltonian is

$$\hat{H}_e = \omega_e \hat{S}_z^e + \Delta \hat{\sigma}_{A_1 A_2} + \frac{\Omega_a}{\sqrt{2}} (\hat{\sigma}_{A_1 b} + i \hat{\sigma}_{A_2 d} + h.c.) + \frac{\Omega_e}{2} (\hat{\sigma}_{E_e 0} + \hat{\sigma}_{E_s 0}),$$

where we have included the ground state Zeeman term $g_e \mu_B B \hat{S}_z^e \equiv \omega_e \hat{S}_z^e$ of an external magnetic field $B$ along the $z$ (N-V) axis. We also include the dissipation of the NV center in the Lindblad form, including the spontaneous emission from $|A_1\rangle$ and $|A_2\rangle$ to $|\pm 1\rangle$ (rate $\gamma/2$), from $|E_e\rangle$ to $|0\rangle$ (rate $\gamma$), and non-radiative intersystem crossing from $|A_1\rangle$ to $|S\rangle$ (rate $\gamma_1$), from $|A_2\rangle$ to $|S\rangle$ (rate $\gamma_2$), from $|S\rangle$ to $|0\rangle$ (rate $\gamma_s$), from $|E_e\rangle$ to $|\pm 1\rangle$ (rate $\gamma_{se}$), and pure dephasing $\gamma_\phi$ for each excited state.

**B. NV-induced nuclear spin transition rate**

To calculate the NV-induced nuclear spin transition rates, we need to determine the NV steady state $\hat{P}(h)$ from $-i[\hat{H}_e + \hat{S}_z^e h, \hat{P}] + \mathcal{L}_e \hat{P}(h) = 0$, where $\hat{H}_e + \hat{S}_z^e h = \hat{H}_{\text{lab}} - \Delta \hat{\sigma}_{A_1 A_2} + h$ and $\delta \equiv \omega_e + h$ is the two-photon detuning. The calculation is straightforward and it turns out that relevant matrix elements of $\hat{P}(h)$ can be expressed in terms of the $|E_i\rangle$ population $\langle E_i|\hat{P}(h)|E_j\rangle$, e.g., $\langle S|\hat{P}(h)|S\rangle = (2\gamma_{se}/\gamma_s)\langle E_s|\hat{P}(h)|E_s\rangle$, $\langle 0|\hat{P}(h)|0\rangle = [1 + (\gamma + 2\gamma_{se})/W_A]\langle E_1|\hat{P}(h)|E_s\rangle$, and $\langle 0|\hat{P}(h)|E_s\rangle = [i(\gamma + 2\gamma_{se})/\Omega_e]\langle E_1|\hat{P}(h)|E_s\rangle$, where $W_A = \Omega_e^2/2(\gamma + \gamma_s + \gamma_{se})$ is the transition rate between $|0\rangle$ and $|E_s\rangle$, $W_A = \Omega_e^2/2(\gamma + \gamma_s + \gamma_{se})$ is the transition rate between $|\pm 1\rangle$ and $|A_1\rangle$, and $\eta_1 \equiv \gamma_{se}/\gamma_{s1}$. We treat the off-resonant $|\pm 1\rangle \rightarrow |A_2\rangle$ excitation perturbatively. Up to zeroth order, near the two-photon resonance $\delta \approx 0$, we neglect $O(\delta^4)$ and high order terms and obtain

$$\langle E_i|\hat{P}(h)|E_j\rangle \approx D_i \delta^2,$$

where

$$D_i = \frac{1}{\eta_1 + 2\eta_1 \frac{\gamma + \gamma_{se}}{W_A} + \frac{\gamma + 2\gamma_{se}}{W_e}},$$

$$\delta^2 \approx \frac{\eta_1 \eta_2}{\eta_1 + \eta_2 + \frac{W_A}{\gamma + \gamma_{se}} (1 + \frac{\gamma + 2\gamma_{se}}{W_e})},$$

with $\eta_2 \equiv \gamma_{se}/(\gamma + \gamma_s + \gamma_{se})$ and we have used $\eta_1 \ll 1$. We find that only $|E_s\rangle, |0\rangle$, and $|d\rangle$ are significantly populated since $\gamma_{ce} \ll \gamma_{s}, \gamma_{s1}, \gamma_s$. Under saturated pumping $W_A \gg \gamma_{se} + \gamma_{s1}$, the leading order correction from the off-resonant excitation to $|A_2\rangle$ is

$$\langle E_i|\hat{P}^2(h)|E_j\rangle \approx \frac{1}{2\eta_1 \gamma + \gamma_s} \left(1 - 2\langle E_i|\hat{P}(h)|E_s\rangle\right),$$

where $W_A = (\Omega_e^2/2)(\gamma + \gamma_{se} + \gamma_s)/\Delta^2$ is the off-resonant transition rate between $|\pm 1\rangle$ and $|A_2\rangle$ and we have used $\gamma_{se} \ll \gamma_{s}, \gamma_{s1}$. Now the $|E_s\rangle$ population $\langle E_s|\hat{P}(h)|E_s\rangle + \langle E_s|\hat{P}^2(h)|E_s\rangle$ no longer vanish at $\delta = 0$. This limits the efficiency of $^{14}\text{N}$ nuclear spin cooling (in agreement with the expectation of Ref. [2] for $^{13}\text{C}$ nuclear spin suppression).

Next we use Eqs. (3) and (4) [with $p_{\text{nm}}$ replaced by $\hat{P}(h)$ and neglecting the dependence of $\mathcal{L}_p_{\text{nm}}$ on $p$ and $m$] to calculate the $^{14}\text{N}$ and $^{13}\text{C}$ nuclear spin transition rate. The transition rate $W_{m+n-m,n}(h) = W_{m-n-m,n}(h)$ for $^{14}\text{N}$ from $|m_0\rangle$ to $|m_\pm 1\rangle \propto \tilde{E}_n^m |m_0\rangle$ is dominated by the following contributions from different NV transitions: $A_{p_{\text{nm}}}(\langle E_i|\hat{P}(h)|E_j\rangle) \propto \delta^2$ from $|E_i\rangle \rightarrow |f\rangle$, where $f$ runs over $A_1, A_2, E_1, E_2$ states, $\chi_{p_{\text{nm}}} \equiv (\gamma + 2\gamma_{se})/D^2$, and $\chi_f \equiv (1/4)(\Gamma_f + \gamma_p)/(\gamma_p - E_{f})^2$, with $E_f$ the energy of $|f\rangle$ in the laboratory spin. These transition rates differ from the phenomenological expression $A_{p_{\text{nm}}}(\langle E_i|\hat{P}(h)|E_j\rangle)$ in Ref. [?]. Similarly, the transition rate $W_{m+n-m,n}(h) = W_{m-n-m,n}(h)$ for $^{13}\text{C}$ from $|m_0\rangle$ to $|m_\pm 1\rangle \propto \tilde{E}_n^m |m_0\rangle$ is dominated by the following contributions: $\chi_{p_{\text{nm}}}(A_{p_{\text{nm}}} - \frac{\gamma_{se}}{2} + A_{p_{\text{nm}}} - \frac{\gamma_s}{2})(\langle E_i|\hat{P}(h)|E_j\rangle) \propto \delta^2$ from $|E_i\rangle \rightarrow |f\rangle$, where $f$ runs over $A_1, A_2, E_1, E_2$ states, $\chi_{p_{\text{nm}}} \equiv (\gamma + 2\gamma_{se})/D^2$, and $\chi_f \equiv (1/4)(\Gamma_f + \gamma_p)/(\gamma_p - E_{f})^2$, with $E_f$ the energy of $|f\rangle$ in the laboratory spin. These transition rates differ from the phenomenological expression $A_{p_{\text{nm}}}(\langle E_i|\hat{P}(h)|E_j\rangle)$ in Ref. [?].

**C. Calculation of NV fluorescence**

To compare with the experimentally observed suppressed $^{13}\text{C}$ nuclear spin noise, we first set $\omega_e = 0.18\text{MHz}$ to obtain the steady nuclear state populations $|p_{\text{nm}}\rangle$ and then calculate the nuclear-state-dependent NV population $\langle E_i|\hat{P}(h)|E_j\rangle$ at the readout magnetic field $\omega_{\text{ref}} = g_e \mu_B B_{\text{ref}}$. The fluorescence before post-selection is proportional to $\sum_m p_{\text{nm}}(\langle E_i|\hat{P}(h)|E_j\rangle e^{-\eta_\text{cond}T_{\text{cond}}(E_i|\hat{P}(h)|E_j)}$, where $\epsilon$ is the photon collection efficiency and $T_{\text{cond}}$ is the photon collection time.