Environment spectrum and coherence behaviours in a rare-earth doped crystal for quantum memory: Supplemental information

B. Gong, T. Tu, Z. Q. Zhou, X. Y. Zhu, C. F. Li, and G. C. Guo
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REALISTIC CALCULATIONS OF THE ENVIRONMENT NOISE SPECTRUM

In the present paper we focus on quantum memories for light using the hyperfine states of rare-earth optical centres in crystals. Such a coupled system of the central ion and the bath nuclear spins can be described by [1]

\[ H = H_0 + H_i + H_e, \]

\[ H_0 = \gamma BS_z, \]

\[ H_i = \sum_i b_i I_i^z, \]

\[ H_e = \gamma_i B \sum_i I_i^z + \sum_{i,j} d_{ij} (4f_i^z f_j^z - I_i^z I_j^z - I_i^+ I_j^- - I_i^- I_j^+), \]

with \( d_{ij} = \frac{\mu_0}{4\pi} \frac{\gamma_i \gamma_h}{r_{ij}^3} (1 - 3 \cos^2 \theta_{ij}) \) (S1)

Here \( H_0 \) describes the central ion Hamiltonian, \( H_i \) includes the dipolar coupling of the environment spins to the central ion, and \( H_e \) represents the magnetic dipolar interaction between environment nuclear spins. \( S \) denotes the operator of the central ion, \( I_i \) the operators of the environment nuclear spins, \( r_{ij} \) the distance between spin \( i \) and \( j \), and \( r_i \) the one between environment spin \( i \) and the central ion, \( \theta_{ij} \) and \( \theta_i \) the angles between the vector connecting the two coupled spins involved and the external magnetic field.

From Eq. (S1) the operator for the effective field on the central ion follow as

\[ \delta B = \sum_i b_i I_i^z, \] (S2)

which allows for calculation of the environment noise spectrum \( S(\omega) \) defined as

\[ S(\omega) = \int_{-\infty}^{\infty} dt \langle \delta B(t) \delta B(0) \rangle e^{i\omega t}. \] (S3)

Various approaches, in particular the phenomenological assumption of Ornstein-Uhlenbeck process has been developed for calculating the environment noise spectrum and the corresponding parameters [2, 3]. Here we use the method [1, 4, 5] which systematically deals with the collective quantum evolution of the dipolar coupled environment spins, allows realistic calculations of environment spectrum properties without using any phenomenological assumption.

The environment spin noise is given by the time-dependent correlation function as

\[ \langle \delta B(t) \delta B(0) \rangle = \sum_i b_i I_i^z(t) \sum_j b_j I_j^z(t) \]

\[ = \sum_i b_i^2 \langle I_i^z(t) I_i^z(0) \rangle + \sum_{i,j} b_i b_j \langle I_i^z(t) I_j^z(0) \rangle. \] (S4)

and is in turn determined by the effective Hamiltonian \( H_e \). When any \( I_i \) flips, a corresponding \( I_j \) must flop in the opposite direction. Thus these flip-flop events construct the collective quantum evolution of the dipolar coupled environment spins, and a reasonable description is introduced by

\[ \langle I_i^z(t) I_j^z(0) \rangle \approx \sum_{j \neq i} \langle I_i^z(t) I_j^z(0) \rangle_{(ij)}, \]

\[ \langle I_i^z(t) I_j^z(0) \rangle \approx \langle I_i^z(t) I_j^z(0) \rangle_{(ij)}, \] (S5)
i.e. "pair-correlation" approximation [1, 6]. Here \((ij)\) denotes that two-pair combination of environment spins forms a distinct subspace, allows to significantly reduce the many-body complexity of the original Hamiltonian. Then any operators \(\hat{O}\) acting on different subspaces are completely independent, i.e. \([\hat{O}_{(ij)}, \hat{O}_{(mn)}] = 0\) if \((ij) \neq (mn)\).

Plugging Eq. (S5) in Eq. (S4) and reordering terms we obtain the two-time environment noise correlation function as

\[
\langle \delta B(t) \delta B(0) \rangle = \sum_{i,j} \langle \delta B_{ij}(t) \delta B_{ij}(0) \rangle_{(ij)} , \quad \text{with} \quad \delta B_{ij} = b_i F_i + b_j F_j .
\]

(H6)

Here it offers a unique chance that the expectation value can be evaluated in the subspace \((ij)\).

In the following we employ a technique similar to the method of moments [7–9]. We focus on \(H_e\) as

\[
H_e = \gamma B \sum_i F_i + 4 \sum_{i<j} d_i F_i F_j - \sum_{i<j} d_{ij}(I_i^+ I_j^- + I_i^- I_j^+) \cos(\omega t),
\]

where a frequency variable \(\omega\) is introduced for convenience (eventually we will take the interested limit \(\omega = 0\)). The flip-flop rate for a pair \((ij)\) is given by Fermi’s golden rule

\[
R_{ij}(\omega) = T_{ij}^{-1}(\omega) = 2\pi d^2 \rho_{ij}(\omega),
\]

with the density of states \(\rho_{ij}(\omega) = \sum_{a,b} \langle \{ a | F_{ij} | b \} \rangle \delta(E_a - E_b - \omega)\). We label the two eigenstates of subspace \((ij)\) by \(|a\rangle\) and \(|b\rangle\) and \(F_{ij} = I_i^+ I_j^- + I_i^- I_j^+\).

We define the \(n\)-th moment as [7–9]

\[
\langle \omega^n \rangle = \frac{\int_0^\infty \omega^n \rho_{ij}(\omega) \mathrm{d} \omega}{\int_0^\infty \rho_{ij}(\omega) \mathrm{d} \omega}.
\]

(S9)

Accordingly, the zeroth moment and the second moment are given by the invariance of the trace

\[
\langle \omega \rangle = \overline{\omega} = 2C \text{Tr}[H_e, F_{ij}^+ I_j^-],
\]

\[
\langle \omega^2 \rangle = -C \text{Tr}[H_e, F_{ij}]^2 ,
\]

(S10)

where \(C\) is a normalization constant. It is lengthy but straightforward to derive the following relations:

\[
\langle \omega \rangle = \overline{\omega} = \frac{1}{2} |b_i - b_j| = \Delta_{ij},
\]

(S11)

\[
\langle (\omega - \overline{\omega})^2 \rangle = 4 \sum_{k \neq i,j} (d_{ik} - d_{jk})^2 = \sigma^2_{ij},
\]

(S12)

where we denote \(\Delta_{ij}\) the frequency shift felt by the central ion when one flip-flop event takes place, and \(\sigma^2_{ij}\) the linewidth for the flip-flop. Using these expressions we can discuss the shape of \(\rho_{ij}(\omega)\), which leads to

\[
\rho_{ij}(\omega) = \frac{1}{8 \sqrt{2\pi} \sigma_{ij}} \left\{ \exp\left[ -\frac{(\omega - \overline{\omega})^2}{2\sigma_{ij}^2} \right] + \exp\left[ -\frac{(\omega + \overline{\omega})^2}{2\sigma_{ij}^2} \right] \right\}, \quad \text{with} \quad \sigma_{ij} = \sqrt{\langle (\omega - \overline{\omega})^2 \rangle}. \tag{S13}
\]

The flip-flop rate is then obtained by setting \(\omega = 0\) in Eq. (S13) and (S8),

\[
R_{ij} = T_{ij}^{-1} = \frac{\sqrt{2\pi}}{4} \frac{d_{ij}}{\sigma_{ij}} \exp\left( -\frac{\Delta_{ij}^2}{2\sigma_{ij}^2} \right) . \tag{S14}
\]

i.e., Eq. (5) in the main text. Therefore we can calculate the correlation function

\[
\langle \delta B(t) \delta B(0) \rangle = \frac{1}{4} \sum_i b_i^2 \sum_{i<j} P_{ij} \exp\left( -\frac{2t}{T_{ij}} \right) , \quad \text{with} \quad P_{ij} = \frac{R_{ij} \Delta_{ij}^2}{\sum_{n<m} R_{nm} \Delta_{nm}^2}. \tag{S15}
\]
Eq. (S15) allows realistic calculations of environment noise spectrum without using any phenomenological assumption. All we need are the rare-earth-doped crystal lattice parameters and the magnetic moments $\gamma_s$, $\gamma_I$ available from the experimental literature [10], which in turn determine the fluctuation rate $R_\epsilon$ and the corresponding correlation function. For the studied system europium-ion dopant in yttrium orthosilicate (Eu$^{3+}$:Y$_2$SiO$_5$), Y$_2$SiO$_5$ have C2/c (C$^{2}_2$) space group with cell parameters $a = 1.041$ nm, $b = 0.673$ nm, $c = 1.249$ nm, $\beta = 103.65^\circ$. Each cell contains 64 atoms and Si presents an unique crystallographic site, Y enters two different site in the lattice and Eu ion occupies site 1 of Y position. The Y’s gyromagnetic moment is 2.09 MHz/T, while Eu ion’s state has a large magnetic moment (be of the order of 100 MHz/T). The computation resources of calculations are in general rapidly increasing due to a large number of environment spins. In practice, we wrote a computer program that sums over lattice sites with $10^4$ atoms and the numbers are high enough to guarantee convergence. The correlation function has been numerically evaluated in Fig. 1(b) in the main text and subsequently fitted to a exponential form $b^2 \exp(-t/\tau_c)$ in order to obtain the environment noise amplitude $b$ and correlation time $\tau_c$.

NUMERICAL SIMULATIONS OF COHERENCE BEHAVIORS

The calculation of environment spectrum outlined above provides good description for the bath around the central ion. The environment noise effects can be modelled [11],

$$H_{\text{eff}} = \epsilon_0 S_z + \epsilon_1 \delta B S_z + \epsilon_2 (\delta B)^2 S_z + ...$$

where we expand the Hamiltonian in the perturbation of the fluctuating magnetic field $\delta B$: $\epsilon_1 = \frac{\partial \omega_0}{\partial B}$ and $\epsilon_2 = \frac{1}{2} \frac{\partial^2 \omega_0}{\partial B^2}$ with the correlation function $C(t) = \langle \delta \epsilon(t) \delta \epsilon(0) \rangle = b^2 \exp(-t/\tau_c)$.

In practice, we can simulate the evolution of the central ion Eq. (S16) by numerically solving the time-dependent Schrodinger equation. The numerical calculations utilize the Runger-Kutta scheme and the result is averaged over 500 realizations of the $\delta \epsilon$ noise of the environment. Each run has a stochastic term $\delta \epsilon$, and the initial value $\delta \epsilon(0)$ is sampled from the Gaussian distribution of the noise with width $b$. At each time step, the value of the random field $\delta \epsilon(t)$ can be randomly sampled using the previous value and the transition probability for the environment process with the correlation function $C(t)$. The simulation results for a wide range of dynamical decoupling protocols are presented in Figs 2, 3, 4 of the main text. Similar numerical analysis has been used in a variety of superconducting, quantum dot and NV center systems [11–13].

ANALYTICAL EXPRESSIONS OF COHERENCE BEHAVIORS

We quantify the coherence of the central ion by using the function $\exp[-\chi(t)]$, which is defined as the off-diagonal elements of the density matrix $|\rho_{+-}(t)|$. Formally, the dynamics of the central ion can be analysed using the evolution operator [11]. Dynamical decoupling sequences can be considered as applying a certain number of $\pi$ pulses at time $t_i$. Since the $\pi$ pulse is given by $\exp(-i \sigma_z) = -i \sigma_x$, we can write the evolution operator by slicing the time interval into small pieces at $t_1, t_2, ...t_n$,

$$U(t) = ...(-i \sigma_x) \exp\left(-i \sigma_z \frac{t}{2} \int_{t_{i-1}}^{t_{i}} [\epsilon_0 + \delta \epsilon] dt \right)...(-i \sigma_x) \exp\left(-i \sigma_z \frac{t}{2} \int_{0}^{t_1} [\epsilon_0 + \delta \epsilon] dt \right).$$

When the fluctuations are Gaussian, the average over noise realizations can be expressed as a Gaussian functional integral over all possible values of $\delta \epsilon$. Therefore $\chi(t)$ can be calculated through the spectral density of the environment noise $S(\omega)$ as [11]

$$\chi(t) = \int_0^\infty \frac{d\omega}{\pi} S(\omega) F(\omega t) \frac{F(\omega t)}{\omega^2}.$$

Here $F(\omega t)$ is the filter function derived from the Fourier transform of the evolution of the pulse sequence on the ion,

$$F(\omega t) = \left( \frac{1}{2} \sum_{n=0}^{n} (-1)^n (e^{i \omega t_{n+1}} - e^{i \omega t_n}) \right)^2.$$
Analytical expressions of $F(\omega t)$ for the CPMG sequences under consideration can be obtained:

$$F(\omega t) = 8 \sin^2\left(\frac{\omega t}{2\tau_c}\right) \frac{\sin^2\left(\frac{\omega t}{4\tau_c}\right)}{\cos^2\left(\frac{\omega t}{2\tau_c}\right)}$$

for even $n$,

$$F(\omega t) = 8 \sin^2\left(\frac{\omega t}{2\tau_c}\right) \cos^2\left(\frac{\omega t}{4\tau_c}\right)$$

for odd $n$.

We will consider the case for large $n$ limit or pulse interval $\tau \ll \tau_c$ (i.e., the case of long correlation time $\tau_c$ which our system belongs to). For CPMG sequence $F(\omega t) < (\omega t/2n)^4$ for $\omega t < 2n$, i.e., $F(\omega t)$ strongly suppresses the low-frequency noise. Since the spectral density is a Lorentzian type $S(\omega) = \frac{2\nu}{\omega^2 + \nu^2}$, the filter function $F(\omega t)$ will suppress the low-frequency part ($\omega < 1/\tau_c$). While for large $\omega t$, the filter function can be approximated by a periodic train (with period $1/2n\tau_c$) of square peaks of height $2n^2$ and width $1/2n^2$. Thus the tail part as $\frac{2\nu}{\omega^2} (\omega > 1/\tau_c)$ contributes to the integral Eq. (S16). Then, to the lowest order of $1/n$, we can sum all of the terms of narrow peaks for $F(\omega t)$ and get

$$\chi(t) \approx \frac{(b\tau_c)^2}{12n^2} \left(\frac{t}{\tau_c}\right)^3,$$

which is the result Eq. (11) in the main text.

**ANALYSIS FOR A PRASEODYMIUM DOPED CRYSTAL**

Several ion-doped crystals offer interesting perspectives as quantum memories for optical states. Among them, the best known systems with the required properties are Eu$^{3+}$:Y$_2$SiO$_5$ and Pr$^{3+}$:Y$_2$SiO$_5$ [14]. In the main text we have described the general theory of rare-earth-doped crystals and given the numerical results for a particular case (Eu$^{3+}$:Y$_2$SiO$_5$). This general theory can be directly applied to other ion-doped solids.

We start by calculating the correlation function of Y-bath spins in a Pr$^{3+}$:Y$_2$SiO$_5$ system. Fig. S1(a) shows that this correlation function decays smoothly over 1 second, and its behavior is fitted by an exponential form $b^2 \exp(-t/\tau_c)$ with $b = 90$ Hz and $\tau_c = 0.5$ s. Then using this bath spin dynamics, Fig. S1(b) presents the signals of central Pr ion under dynamical decoupling pulses as a function of time. The coherence envelope can be compared with experimental result [15]. For example, the calculated value of decoherence rate $1/T_2$ for pulse interval $\tau = 10$ ms is $6.8 \times 10^{-2}$ s$^{-1}$, which is agreement with the measured gradient of the dynamical decoupling decay curve ($4.7 \times 10^{-2}$ s$^{-1}$).

Next we discuss its implications for the different systems. Clearly the results of Pr$^{3+}$:Y$_2$SiO$_5$ are quite similar to those of Eu$^{3+}$:Y$_2$SiO$_5$, but the time scale of coherence envelope is twenty times smaller than the value of Eu-doped system. Actually our theory suggests this small time scale is coming from the incomplete suppression of Y bath spin dynamics. The key mechanism is that the strong couplings between the central ion and the neighbour Y spins is responsible for that Larmor frequencies of neighbour Y spins are detuned from each other. Including the relatively smaller magnetic moment of Pr ion, the calculation shows that the couplings between the central ion and the neighbour Y spins is one order of magnitude smaller in Pr$^{3+}$:Y$_2$SiO$_5$ than those in Eu$^{3+}$:Y$_2$SiO$_5$. The smaller $b_i$ reduces the degree of detuning of neighbour Y spins, hence increase the flip-flop rate $R_{ij}$. Therefore the Y bath spin dynamics is not entirely suppressed in Pr$^{3+}$:Y$_2$SiO$_5$ and the correlation time $\tau_c$ is only 0.5 s (in comparison, the correlation time is about 12 s in Eu-doped system). Furthermore, the scaling behaviors of coherence envelope extend over a range on several tens of seconds time scale (in comparison, the scaling spans an hour-long time scale in Eu-doped system).

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FIG. 1. (a) Numerically calculated environment correlation function $C(t)$ for Pr-doped crystals (red circles) and the fitted shape $b^2 \exp(-t/\tau_c)$ (green solid line) with $b = 90$ Hz and correlation time $\tau_c = 0.5 \, s$. (b) Circles are simulation data of coherence envelope for Pr-doped crystals under CPMG sequences with pulse interval $\tau = 7.5 \, ms$, $10 \, ms$, $15 \, ms$, $20 \, ms$. Solid lines are obtained by the scaling expression $(t/T_2)^\beta$ with $\beta = 1$. Insert: Extracted $1/T_2$ for CPMG pulse interval. Solid line is also the scaling function $1/T_2 \sim \tau^\delta$ with $\delta = 2$. 