Dressed state control of effective dipolar interaction between strongly-coupled solid-state spins : Supplementary Information

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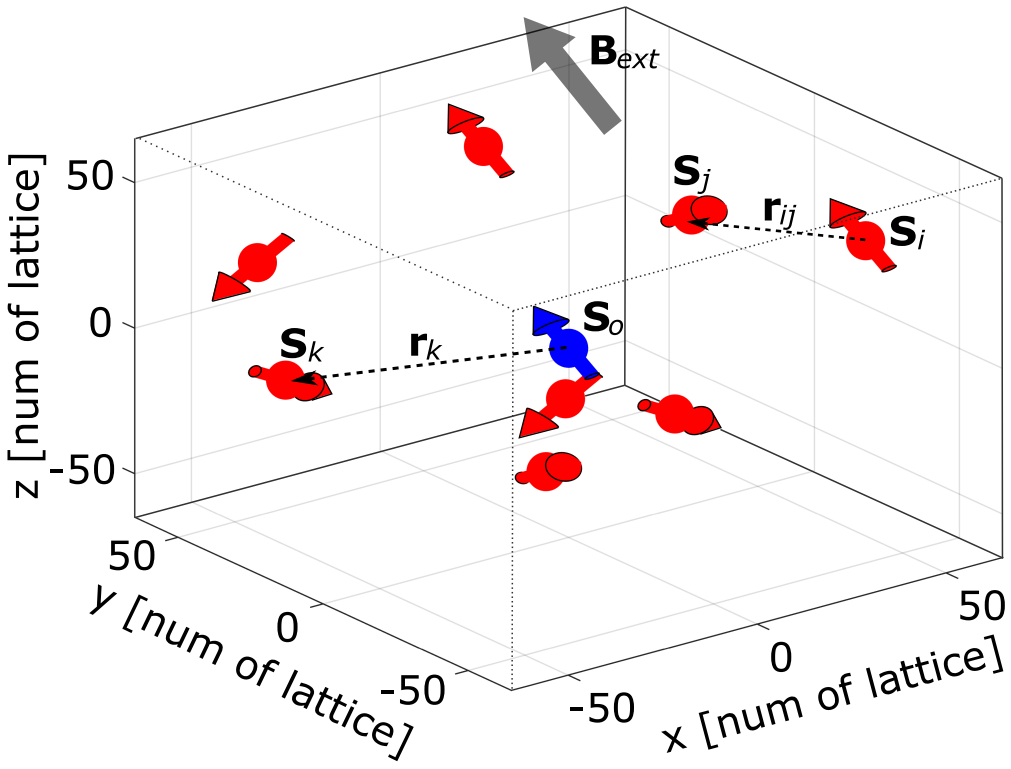
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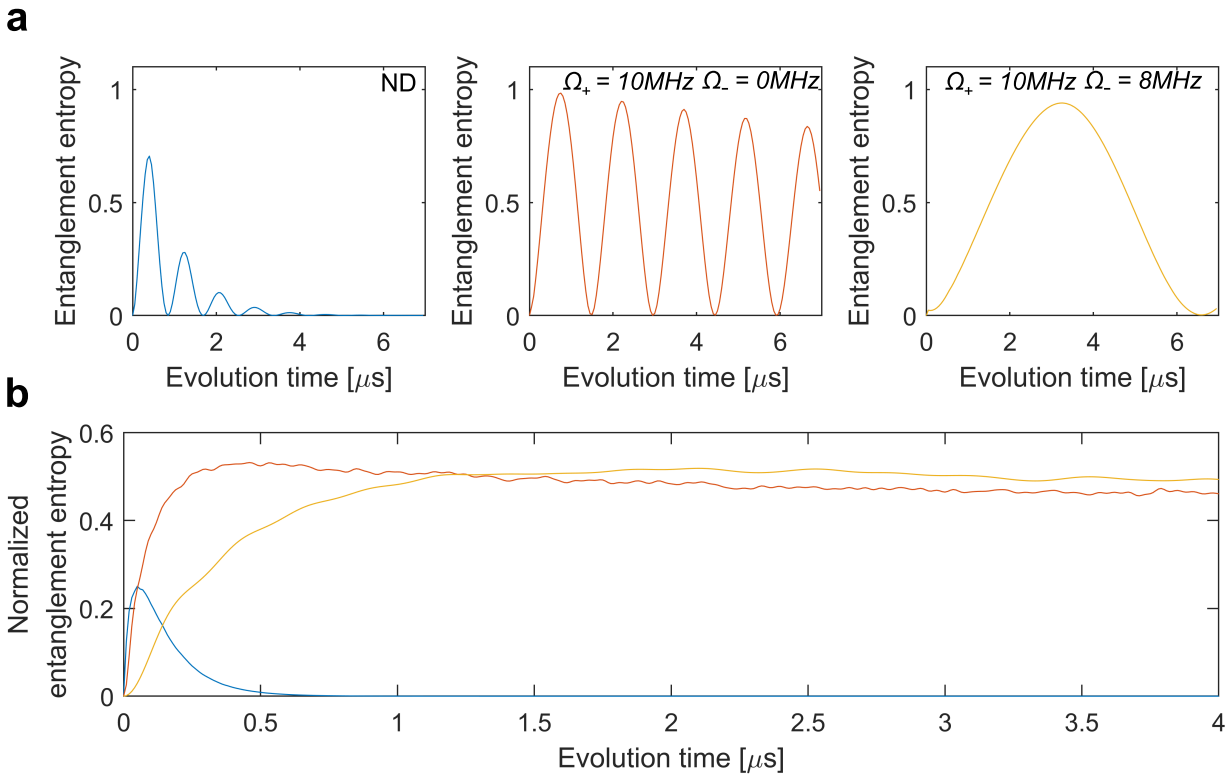
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Supplementary Figure 1. NV spin ensemble configuration with centre spin (blue) and bath NV spins (red). 50 lattice sites correspond to distance of ~5 nm (the average NV-NV separation for [NV] of 50 ppm)



Supplementary Figure 2. a, Time evolution of the von Neumann entropy of an NV spin pair with no driving field (blue), single driving field (red), and double driving field (yellow) applied. Bare dipolar coupling strength between two NV spins is set to 0.3 MHz. **b,** Time evolution of the normalized von Neumann entropy for the ensemble NV spin pairs with no driving field (blue), single driving field (red), and double driving field (yellow) applied.

Supplementary Methods

System Hamiltonian without Rabi driving fields — The electronic spin Hamiltonian for two off-axis NV spins without driving fields is given by

$$H = H_A + H_B + H_{int} \tag{1}$$

where H_A and H_B are the individual Hamiltonians for NV_A and NV_B, respectively, and H_{int} is the interaction Hamiltonian between the two NV spins. The individual Hamiltonians can be expanded as

$$\begin{aligned} H_A &= D(S_A^z)^2 + \gamma \mathbf{B_{ext}} \cdot \mathbf{S}_A + h_A \mathbf{S}_A \cdot \mathbf{I}_A \\ H_B &= D(S_B^z)^2 + \gamma \mathbf{B_{ext}} \cdot \mathbf{S}_B + h_B \mathbf{S}_B \cdot \mathbf{I}_B \end{aligned} \tag{2}$$

where D = 2.87 GHz is the zero-field splitting, γ is the NV electronic gyromagnetic ratio, $\mathbf{B_{ext}}$ is the external bias magnetic field, $\mathbf{S}_A$ and $\mathbf{S}_B$ are the NV electronic spin operators, and $h_{A,B}$ are hyperfine interactions between each NV electronic spin and its on-site ^{14}N nuclear spin (I=1). Since $D \gg h_{A,B}$, the secular terms associated with electron-nuclear spin flip-flop operators in hyperfine interactions with nearby nuclear spins may be disregarded when there is only a minor misalignment in the bias magnetic field[3]. The effect of non-secular hyperfine interaction terms can also be (partially) neglected for the Rabi drive frequency conditions used in the present experiment, except for energy-level splitting of the NV electronic spin sublevels into three hyperfine levels, $m_I = \pm 1, 0$, for the on-site ^{14}N nuclear spin (I=1), as observed in Ramsey measurements described in the main text. We assume that the external bias magnetic field $\mathbf{B_{ext}}$ is aligned with the NV_A spin, leaving only the $\gamma B_A S_A^z$ Zeeman term for NV_A. Then the Hamiltonian can be expressed in the basis of $|m_s^A = 0\rangle$ and $|m_s^A = \pm 1\rangle$. The bias magnetic field is misaligned with the NV_B quantization axis, and can thus cause spin state mixing for this NV via the transverse Zeeman term included in (2). However, for small magnetic field strength ($|\mathbf{B}_{ext}| \sim 40$ G $\ll |D/\gamma_e|$) as used in the present experiment, this effect can be neglected due to the large zero-field splitting of NV spin; we can then use $|m_s^B = 0\rangle$ and $|m_s^B = \pm 1\rangle$ as the basis states, and only the $\gamma B_B S_B^z$ Zeeman term remains for NV_B. The interaction Hamiltonian is described as a dipole-dipole interaction between the NV_A and NV_B spins,

$$H_{int} = -\frac{J_0}{r^3}(3(\mathbf{S}_A \cdot \hat{r})(\mathbf{S}_B \cdot \hat{r}) - \mathbf{S}_A \cdot \mathbf{S}_B) \tag{3}$$

where $J_0 = (2\pi)$ 52MHz·nm^3 and $\mathbf{r}$ is a relative displacement vector between the two spins. The interaction can be expressed in the basis of each NV spin's own quantization axis: i.e., $\mathbf{S}_{A,B}$ are decomposed in terms of the $(S_{A,B}^x, S_{A,B}^y, S_{A,B}^z)$ coordinate system, where $\hat{z}_{A,B}$ is the quantization axis for each NV spin. For two NV spins with different orientations and hence quantization axes, flip-flop terms in the interaction Hamiltonian $(S_A^x S_B^x + S_A^y S_B^y)$, $(S_A^x S_B^y - S_A^y S_B^x)$ oscillate fast with respect to $S_{A,B}^z$, and thus average out on timescales short compared to the effects studies here. Therefore only the Ising interaction term $S_A^z S_B^z$ remains[2], and the interaction Hamiltonian is simplified as

$$H_{int} \approx -\frac{J_0 q_{A,B}}{r^3} S_A^z S_B^z = \nu_{dip} S_A^z S_B^z \tag{4}$$

where $q_{A,B} = -(1 - 3\cos^2\theta)$ with $\cos\theta = \hat{z} \cdot \hat{r}$, and ν is the magnetic spin dipolar coupling strength between the two NVs. Note that other mixing terms (e.g., $S_A^x S_B^z$) can be neglected due to the bias magnetic field alignment with NV_A (along its z-axis) and the large zero field splitting relative to the Zeeman splitting[3].

Double Electron-Electron Resonance signal — Using the simplified interaction Hamiltonian framework presented in Supp. Eqn. (4), we set up a unitary gate sequence for double electron-electron resonance (DEER) measurement given as below.

$$U_{sq}(t) = \left(U\left(\frac{\pi}{2}\right)_x \otimes \mathbb{I}\right) e^{-iH_{int}t/2} \; (U(\pi)_x \otimes U(\pi)_x) e^{-iH_{int}t/2} \left(U\left(\frac{\pi}{2}\right)_x \otimes \mathbb{I}\right) \tag{5}$$

By implementing the aforementioned gate sequence on the initial density matrix $\rho_i = |00\rangle\langle 00|$, and measuring the projection onto $\rho_0 = |00\rangle\langle 00|$ after a duration of time t, we obtain DEER signal in a single quantum basis ($|m_s = 0\rangle, |m_s = +1\rangle$).

$$Tr\left(\rho_0 U_{sq}(t) \rho_i U_{sq}^\dagger\right) = \frac{1}{4} e^{-\frac{1}{2}i\nu_{dip}t} \left(1 + e^{\frac{1}{2}i\nu_{dip}t}\right)^2 = \frac{1}{2}\left(1 + \cos\left(\frac{\nu_{dip}t}{2}\right)\right) \tag{6}$$

Now, setting up a unitary gate sequence for double-quantum DEER measurement using

$$U_{dq}(t) = \left(U\left(\frac{\pi}{2}\right)_{x,dq} \otimes \mathbb{I}\right) e^{-iH_{int}t/2} \; \left(U(\pi)_{x,dq} \otimes U(\pi)_{x,dq}\right) e^{-iH_{int}t/2} \left(U\left(\frac{\pi}{2}\right)_{x,dq} \otimes \mathbb{I}\right) \tag{7}$$

and for the initial density matrix $\rho_i' = |01\rangle\langle 10|$, and measuring the projection onto $\rho_0 = |00\rangle\langle 00|$ after a duration of time t yields DEER signal in a double quantum basis ($|B\rangle = \frac{1}{\sqrt{2}}(|+\rangle + |-\rangle), |D\rangle = \frac{1}{\sqrt{2}}(|+\rangle - |-\rangle)$.

$$Tr\left(\rho_0 U_{dq}(t)\rho_i' U_{dq}^{\dagger}\right) = \frac{1}{4}e^{-2i\nu_{dip}t}\left(1 + e^{2i\nu_{dip}t}\right)^2 = \frac{1}{2}\left(1 + \cos\left(2\nu_{dip}t\right)\right) \tag{8}$$

Ramsey measurement signal — Analogous to the derivation of the DEER measurement signal, a unitary gate sequence was established for the Ramsey measurement as presented below:

$$U_{sq}(t) = \left(U\left(\frac{\pi}{2}\right)_x \otimes \mathbb{I}\right) e^{-iH_{int}t} \left(U\left(\frac{\pi}{2}\right)_x \otimes \mathbb{I}\right) \tag{9}$$

Measured Ramsey signal for the initial density matrix $\rho_i' = |01\rangle\langle 10|$, and measuring the projection onto $\rho_0 = |00\rangle\langle 00|$ after a duration of time t gives below signal.

$$Tr\left(\rho_0 U_{sq}(t)\rho_i' U_{sq}^{\dagger}\right) = -\frac{1}{4}e^{-i\nu_{dip}t}\left(e^{i\nu_{dip}t} - 1\right)^2 = \frac{1}{2}\left(1 - \cos\left(\nu_{dip}t\right)\right) \tag{10}$$

Similarly, Ramsey signal derived in double quantum basis of $|B\rangle, |D\rangle$,

$$U_{dq}(t) = \left(U\left(\frac{\pi}{2}\right)_{x,dq} \otimes \mathbb{I}\right) e^{-iH_{int}t} \left(U\left(\frac{\pi}{2}\right)_{x,dq} \otimes \mathbb{I}\right) \tag{11}$$

is presented below.

$$Tr\left(\rho_0 U_{dq}(t)\rho_i' U_{dq}^{\dagger}\right) = \frac{1}{4}e^{-2i\nu_{dip}t}\left(1 + e^{2i\nu_{dip}t}\right)^2 = \frac{1}{2}\left(1 + \cos\left(2\nu_{dip}t\right)\right) \tag{12}$$

System Hamiltonian with double driving fields — Now we add Rabi transition driving fields into the system Hamiltonian and analytically calculate the change of effective dipolar coupling strengths due to the relative driving field strengths. The total Hamiltonian of the system is given as

$$\begin{aligned} H(t) = {} & D(S_A^z)^2 + \gamma B_A S_A^z + h_A S_A^z I_A^z + \Omega_+^A \cos(\omega_+^A t) S_{A,+}^x + \Omega_-^A \cos(\omega_-^A t)\, S_{A,-}^x \\ & + D(S_B^z)^2 + \gamma B_B S_B^z + h_B S_B^z I_B^z + \Omega_+^B \cos(\omega_+^B t) S_{B,+}^x + \Omega_-^B \cos(\omega_-^B t)\, S_{B,-}^x + \nu_{dip} S_A^z \otimes S_B^z \end{aligned} \tag{13}$$

where D is the zero-field splitting, v is the dipolar coupling strength between the two NV spins, and the $\Omega_{\pm}^{A,B}$ and $\omega_{\pm}^{A,B}$ terms are the Rabi frequencies and oscillating frequencies, respectively, of external microwave fields that resonantly drive the energy levels $|m_s = 0\rangle \leftrightarrow |m_s = -1\rangle$ and $|m_s = 0\rangle \leftrightarrow |m_s = +1\rangle$ for NV_A and NV_B. S_A^x and S_B^x operators are the Rabi driving terms, which transform the eigenstates into the dressed states. Gell-Mann operators[4] and Pauli matrices used in the above equation are

$$S^{x,}_{\{A,B\},+} = \begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \qquad S^{x}_{\{A,B\},-} = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & 1 & 0 \end{pmatrix}, \qquad S^{z}_{\{A,B\}} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & -1 \end{pmatrix} \tag{14}$$

Note that non-secular hyperfine interaction terms result in energy-level splitting of the $m_s = \pm 1$ sublevels of the NV spin, as observed in the Ramsey measurements presented in the main text. However, when a double driving field is applied to the NV spin, and under the condition of large Rabi drive frequency $\Omega_{\pm} \gg h_{\mathrm{A,B}}$, as used in our experiments, additional non-secular hyperfine detuning effects are suppressed. For further discussion on local detuning and broadening effects, refer to Section 1.7. The primary objective here is to derive an analytical form of the effective coupling strengths between two NV spins when one NV spin is in a doubly-dressed state. Throughout the derivation, we remain within a rotating frame aligned with the quantization axis of each NV spin and at a driving frequency corresponding to that axis. In order to go to the double rotating frame, we apply the unitary transformation[5]

$$V = \begin{pmatrix} e^{i\omega_+^A t} & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & e^{i\omega_-^A t} \end{pmatrix} \otimes \begin{pmatrix} e^{i\omega_+^B t} & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & e^{i\omega_-^B t} \end{pmatrix} \tag{15}$$

which commutes with $S_{A,B}^z$. Then the Hamiltonian becomes

$$\widetilde{H} = VH(t)V^{\dagger} - iV\frac{dV^{\dagger}}{dt}$$

$$= (\Omega_{+}^{A}\cos(\omega_{+}^{A}t) + \Omega_{-}^{A}\cos(\omega_{-}^{A}t))\begin{pmatrix} 0 & e^{i\omega_{+}^{A}t} & 0 \\ e^{-i\omega_{+}^{A}t} & 0 & e^{-i\omega_{-}^{A}t} \\ 0 & e^{i\omega_{-}^{A}t} & 0 \end{pmatrix} \otimes \mathbb{I}$$

$$+ \gamma B_A S_A^z \otimes \mathbb{I} + D(S_A^z)^2 \otimes \mathbb{I} + (-\omega_{+}^{A}|+1\rangle_A\langle+1|_A - \omega_{-}^{A}|-1\rangle_A\langle-1|_A) \otimes \mathbb{I}$$

$$+\mathbb{I} \otimes (\Omega_{+}^{B}\cos(\omega_{+}^{B}t) + \Omega_{-}^{B}\cos(\omega_{-}^{B}t))\begin{pmatrix} 0 & e^{i\omega_{+}^{B}t} & 0 \\ e^{-i\omega_{+}^{B}t} & 0 & e^{-i\omega_{-}^{B}t} \\ 0 & e^{i\omega_{-}^{B}t} & 0 \end{pmatrix}$$

$$+\mathbb{I} \otimes \gamma B_B S_B^z + \mathbb{I} \otimes D(S_B^z)^2 + \mathbb{I} \otimes (-\omega_{+}^{B}|+1\rangle_B\langle+1|_B - \omega_{-}^{B}|-1\rangle_B\langle-1|_B) + \nu_{dip} S_A^z \otimes S_B^z \quad (16)$$

and in matrix form, can be simplified as

$$\widetilde{H} \simeq \begin{pmatrix} D + \gamma B_A - \omega_{+}^{A} & \Omega_{+}^{A} & 0 \\ \Omega_{+}^{A} & 0 & \Omega_{-}^{A} \\ 0 & \Omega_{-}^{A} & D - \gamma B_A - \omega_{-}^{A} \end{pmatrix} \otimes \mathbb{I}$$
$$+\mathbb{I} \otimes \begin{pmatrix} D + \gamma B_A - \omega_{+}^{B} & \Omega_{+}^{B} & 0 \\ \Omega_{+}^{B} & 0 & \Omega_{-}^{B} \\ 0 & \Omega_{-}^{B} & D - \gamma B_B - \omega_{-}^{B} \end{pmatrix} + \nu_{dip} S_A^z \otimes S_B^z \quad (17)$$

where the rotating-wave approximation is used to simplify the equation. By setting $\omega_{+}^{A} = D + \gamma B_A - \delta\omega^A, \omega_{-}^{A} = D - \gamma B_A$, which is for a single quantum Ramsey in the basis of $|m_s = 0\rangle$, $|m_s = +1\rangle$, $\Omega_{+}^{A} = \Omega_{-}^{A} = 0$, assuming no spin driving for NV_A, and finally, letting $\omega_{+}^{B} = D + \gamma B_B, \omega_{-}^{A} = D - \gamma B_B$, we find:

$$\widetilde{H} = \begin{pmatrix} \delta\omega^A & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix} \otimes \mathbb{I} + \mathbb{I} \otimes \begin{pmatrix} 0 & \Omega_{+}^{B} & 0 \\ \Omega_{+}^{B} & 0 & \Omega_{-}^{B} \\ 0 & \Omega_{-}^{B} & 0 \end{pmatrix} + \nu_{dip} S_A^z \otimes S_B^z \quad (18)$$

Now, we want to analytically calculate the effective magnetic spin dipolar coupling strength when

NV_B is prepared in the doubly-dressed state. For the NV_A spin to be controlled on resonance, Supp. Eqn. (18) can be re-written as

$$\widetilde{H} = \mathbb{I} \otimes \begin{pmatrix} 0 & \Omega_+ & 0 \\ \Omega_+ & 0 & \Omega_- \\ 0 & \Omega_- & 0 \end{pmatrix} + \nu_{dip} S_A^z \otimes S_B^z \tag{19}$$

in the double rotating frame and with $\Omega_+^B = \Omega_+, \Omega_-^B = \Omega_-$. We diagonalize the Rabi driving term by tilting the full Hamiltonian with the rotation matrix R, which is the set of eigenvectors of the Rabi driving term, only on the NV_B spin:

$$\widetilde{H}_R = (\mathbb{I} \otimes R^{-1})\widetilde{H}(\mathbb{I} \otimes R) = \mathbb{I} \otimes \begin{pmatrix} \sqrt{\Omega_+^2 + \Omega_-^2} & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & -\sqrt{\Omega_+^2 + \Omega_-^2} \end{pmatrix} + \nu_{dip} S_A^z \otimes (R^{-1} S_B^z R)$$

$$= \sqrt{\Omega_+^2 + \Omega_-^2}\left(\mathbb{I} \otimes S_{B,R}^z\right) + \nu_{dip} S_A^z \otimes \begin{pmatrix} \frac{\Omega_+^2 - \Omega_-^2}{2(\Omega_+^2 + \Omega_-^2)} & \frac{\sqrt{2}\Omega_+\Omega_-}{\Omega_+^2 + \Omega_-^2} & \frac{\Omega_+^2 - \Omega_-^2}{2(\Omega_+^2 + \Omega_-^2)} \\ \frac{\sqrt{2}\Omega_+\Omega_-}{\Omega_+^2 + \Omega_-^2} & -\frac{\Omega_+^2 - \Omega_-^2}{\Omega_+^2 + \Omega_-^2} & \frac{\sqrt{2}\Omega_+\Omega_-}{\Omega_+^2 + \Omega_-^2} \\ \frac{\Omega_+^2 - \Omega_-^2}{2(\Omega_+^2 + \Omega_-^2)} & \frac{\sqrt{2}\Omega_+\Omega_-}{\Omega_+^2 + \Omega_-^2} & \frac{\Omega_+^2 - \Omega_-^2}{2(\Omega_+^2 + \Omega_-^2)} \end{pmatrix} \tag{20}$$

where R is given as

$$R = \begin{pmatrix} \frac{\Omega_+}{\sqrt{2}\sqrt{\Omega_+^2 + \Omega_-^2}} & \frac{\Omega_-}{\sqrt{\Omega_+^2 + \Omega_-^2}} & \frac{\Omega_+}{\sqrt{2}\sqrt{\Omega_+^2 + \Omega_-^2}} \\ \frac{1}{\sqrt{2}} & 0 & -\frac{1}{\sqrt{2}} \\ \frac{\Omega_-}{\sqrt{2}\sqrt{\Omega_+^2 + \Omega_-^2}} & -\frac{\Omega_+}{\sqrt{\Omega_+^2 + \Omega_-^2}} & \frac{\Omega_-}{\sqrt{2}\sqrt{\Omega_+^2 + \Omega_-^2}} \end{pmatrix} \tag{21}$$

and $S_{B,R}^z$ is a spin-1 Pauli z-matrix in a tiled frame. Doubly-dressed states have basis states:

$$|+\rangle_d = \frac{1}{\sqrt{2}}\left(\frac{\Omega_+}{\sqrt{\Omega_+^2 + \Omega_-^2}}|+\rangle + \frac{\Omega_-}{\sqrt{\Omega_+^2 + \Omega_-^2}}|-\rangle + |0\rangle\right),$$
$$|0\rangle_d = \left(\frac{\Omega_-}{\sqrt{\Omega_+^2 + \Omega_-^2}}|+\rangle - \frac{\Omega_+}{\sqrt{\Omega_+^2 + \Omega_-^2}}|-\rangle\right),$$
$$|-\rangle_d = \frac{1}{\sqrt{2}}\left(\frac{\Omega_+}{\sqrt{\Omega_+^2 + \Omega_-^2}}|+\rangle + \frac{\Omega_-}{\sqrt{\Omega_+^2 + \Omega_-^2}}|-\rangle - |0\rangle\right) \tag{22}$$

For $\Omega_{+,-} \gg \nu_{dip}$, off diagonal terms in tilted Hamiltonian $\widetilde{H}_R$ are rapidly oscillating in time, and therefore can be neglected. Furthermore, the $|0\rangle_B\langle 0|$ operator does not contribute to the effective coupling Hamiltonian. Therefore, $\widetilde{H}_R$ can be approximated as below:

$$\widetilde{H}_R \approx \sqrt{\Omega_+^2 + \Omega_-^2}\left(\mathbb{I} \otimes S_{B,R}^z\right) + \nu_{dip} S_A^z \otimes \begin{pmatrix} \frac{\Omega_+^2 - \Omega_-^2}{2(\Omega_+^2 + \Omega_-^2)} & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & \frac{\Omega_+^2 - \Omega_-^2}{2(\Omega_+^2 + \Omega_-^2)} \end{pmatrix}$$

$$= \sqrt{\Omega_+^2 + \Omega_-^2}\left(\mathbb{I} \otimes S_{B,R}^z\right) + \frac{\Omega_+^2 - \Omega_-^2}{2(\Omega_+^2 + \Omega_-^2)} \nu_{dip} S_A^z \otimes \left(S_{B,R}^z\right)^2 \quad (23)$$

From the above Hamiltonian in the rotating frame, we can deduce the effective magnetic spin dipolar interaction strength ν_{eff} used in the main text as the following:

$$\nu_{eff} \approx \frac{1}{2} \frac{(\Omega_+^2 - \Omega_-^2)}{(\Omega_+^2 + \Omega_-^2)} \nu_{dip} \quad (24)$$

Hartmann-Hahn cross-polarization signal — To gain analytical insights in the cross-polarization dynamics under the double driving scheme, we now incorporate the Rabi driving term for NV_A:

$$\widetilde{H} = \begin{pmatrix} 0 & \Omega^A & 0 \\ \Omega^A & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix} \otimes \mathbb{I} + \mathbb{I} \otimes \begin{pmatrix} 0 & \Omega_+^B & 0 \\ \Omega_+^B & 0 & \Omega_-^B \\ 0 & \Omega_-^B & 0 \end{pmatrix} + \nu_{dip} S_A^z \otimes S_B^z \quad (25)$$

We diagonalize the Rabi driving terms by tilting the full Hamiltonian with the rotation matrix $R_A \otimes R_B$, which is the set of eigenvectors of each Rabi driving term:

$$\widetilde{H}_{DR} = \Omega^A\left(S^z_{A,+} \otimes \mathbb{I}\right) + \sqrt{(\Omega^B_+)^2 + (\Omega^B_-)^2}\left(\mathbb{I} \otimes S^z_{B,+}\right)$$
$$+\nu_{dip}\begin{pmatrix} \frac{1}{2} & \frac{1}{2} & 0 \\ \frac{1}{2} & \frac{1}{2} & 0 \\ 0 & 0 & -1 \end{pmatrix} \otimes \begin{pmatrix} \frac{\Omega_+^2 - \Omega_-^2}{2(\Omega_+^2 + \Omega_-^2)} & \frac{\Omega_+^2 - \Omega_-^2}{2(\Omega_+^2 + \Omega_-^2)} & \frac{\sqrt{2}\Omega_+\Omega_-}{\Omega_+^2 + \Omega_-^2} \\ \frac{\Omega_+^2 - \Omega_-^2}{2(\Omega_+^2 + \Omega_-^2)} & \frac{\Omega_+^2 - \Omega_-^2}{2(\Omega_+^2 + \Omega_-^2)} & \frac{\sqrt{2}\Omega_+\Omega_-}{\Omega_+^2 + \Omega_-^2} \\ \frac{\sqrt{2}\Omega_+\Omega_-}{\Omega_+^2 + \Omega_-^2} & \frac{\sqrt{2}\Omega_+\Omega_-}{\Omega_+^2 + \Omega_-^2} & -\frac{\Omega_+^2 - \Omega_-^2}{\Omega_+^2 + \Omega_-^2} \end{pmatrix} \tag{26}$$

where R_A and R_B are given as

$$R_A = \begin{pmatrix} \frac{1}{\sqrt{2}} & \frac{1}{\sqrt{2}} & 0 \\ \frac{1}{\sqrt{2}} & -\frac{1}{\sqrt{2}} & 0 \\ 0 & 0 & 1 \end{pmatrix}, R_B = \begin{pmatrix} \frac{\Omega_+}{\sqrt{2}\sqrt{\Omega_+^2 + \Omega_-^2}} & \frac{\Omega_+}{\sqrt{2}\sqrt{\Omega_+^2 + \Omega_-^2}} & \frac{\Omega_-}{\sqrt{\Omega_+^2 + \Omega_-^2}} \\ \frac{1}{\sqrt{2}} & -\frac{1}{\sqrt{2}} & 0 \\ \frac{\Omega_-}{\sqrt{2}\sqrt{\Omega_+^2 + \Omega_-^2}} & \frac{\Omega_-}{\sqrt{2}\sqrt{\Omega_+^2 + \Omega_-^2}} & -\frac{\Omega_+}{\sqrt{\Omega_+^2 + \Omega_-^2}} \end{pmatrix} \tag{27}$$

For $\Omega_{+,-} \gg \nu$, the $S^z_{A,\pm}S^x_{B,\pm}$, $S^x_{A,\pm}S^z_{B,\pm}$ terms can be almost neglected, then Supp. Eqn. (26) can be simplified to

$$\widetilde{H}_{DR} \approx \Omega^A\left(S^z_{A,+} \otimes \mathbb{I}\right) + \sqrt{(\Omega^B_+)^2 + (\Omega^B_-)^2}\left(\mathbb{I} \otimes S^z_{B,+}\right)$$
$$+\nu_{dip}\frac{1}{2}\, S^x_{A,+} \otimes \left(\frac{\Omega_+^2 - \Omega_-^2}{2(\Omega_+^2 + \Omega_-^2)} S^x_{B,+} + \frac{\sqrt{2}\Omega_+\Omega_-}{\Omega_+^2 + \Omega_-^2}\left(S^x_{B,-} + S^x_{B,pm}\right)\right) \tag{28}$$

where S^x_{pm} is a Pauli x-matrix between the $|+\rangle, |-\rangle$ basis states. When the Hartmann-Hahn matching condition is met, i.e., $\Omega^A = \sqrt{(\Omega^B_+)^2 + (\Omega^B_-)^2}$, only the term $S^x_{A,+}S^x_{B,+}$ remains for a coherent interaction. Consequently, the total Hamiltonian can be further simplified to:

$$\widetilde{H}_{DR} \approx \Omega^A\left(S^z_{A,+} \otimes \mathbb{I}\right) + \sqrt{(\Omega^B_+)^2 + (\Omega^B_-)^2}\left(\mathbb{I} \otimes S^z_{B,+}\right)$$
$$+\nu_{dip}\frac{\Omega_+^2 - \Omega_-^2}{4(\Omega_+^2 + \Omega_-^2)}\, S^x_{A,+} \otimes S^x_{B,+} \tag{29}$$

The form of this equation is almost identical to that for a two spin-1/2 Hartmann-Hahn cross-polarization transfer[6]. Following a similar derivation, we obtain the cross-polarization transfer rate.

$$R_{pol} \approx \frac{1}{2} \frac{(\Omega_+^2 - \Omega_-^2)}{(\Omega_+^2 + \Omega_-^2)} \nu_{dip} \tag{30}$$

Effect of temperature drift on the effective dipolar coupling strength — Typically, phase accumulation measurements in the double quantum basis are considered to be more resilient to temperature drift since the associated NV spin phase noise is common mode noise in both $|\pm 1\rangle$ states and is thus canceled out during a sensing measurement. However, in a doubly-dressed state scheme, the zero-field detuning δD caused by temperature drift ($\delta D(S^z)^2$) shifts the Hartmann-Hahn (HH) matching condition to the following relation:

$$\Omega_{\delta D}^A \approx \sqrt{\left(\frac{\delta D}{2}\right)^2 + (\Omega_+^B)^2 + (\Omega_-^B)^2} \tag{31}$$

which may broaden the HH resonance condition, e.g., as in the measurements shown in main Fig. 4b. This heating or system drift issue can be mediated by generating a sufficiently large Rabi frequency, with the driving amplitude surpassing the detuning caused by heating ($\partial D/\partial T = -74$ kHz K^{-1}) on the NV spin. As one example of such an approach, an NV spin ensemble within a nano-beam structure and using a small Omega loop waveguide with an inner diameter of 20 μm provided Rabi frequencies[2] up to (2π) 100 MHz.

Effect of local detuning on the effective dipolar coupling strength — Local detuning of an NV spin from the global Rabi driving resonance frequency can arise from various effects, including many-body interactions with bath spins (e.g., ^{13}C nuclear spins and P1 electronic spins), non-secular hyperfine interactions, and variations in the zero-field splitting due to strain in the diamond. Nominally, such NV detuning is suppressed with high Rabi driving frequencies, as used in the present experiment. However, a large local detuning still affects the effective dipolar coupling strength, which can degrade the fidelity of dressed-state control. In the rotating frame, a local detuning on a driven spin shifts the energy gap between the dressed states to $\sqrt{\Omega^2 + \Delta_i^2}$, where Δ_i is the local detuning for the i^{th} spin. Under the strong drive approximation, the effective dipolar coupling with the local detuning can be rewritten as below:

$$\nu_{eff} \approx \left(\frac{\Omega_+^2 - \Omega_-^2}{\Omega_+^2 + \Omega_-^2 + 2\Delta_i^2} \right) \frac{\nu_{dip}}{2} \tag{32}$$

We can use this expression to estimate the effective many-body induced detuning on the coupling strength for realistic measurement conditions. The local disorder potential $\Delta_i \approx (2\pi)$ 4 MHz for [NV] = 50 ppm, as in the present experiment[1]. For typical Rabi driving frequencies $\Omega_+ \approx (2\pi)$ 20 MHz and $\Omega_- \approx (2\pi)$ 10 MHz, ν_{eff} is estimated to change by only 7 % with and without the Δ_i detuning in Supp. Eqn. (32). Recalling that the distribution of the effective dipolar coupling strength has FWHM/R_{dd} > 2 (main Fig. 5c), the effect of detuning from many-body interactions on the distribution of the effective dipolar coupling is therefore expected to be modest. A similar argument applies to the effects from non-secular hyperfine interaction terms and CVD diamonds with typical local strain homogeneity.

Supplementary Discussion

Doubly-dressed state scheme is robust to inhomogeneous Rabi driving field amplitudes — For the doubly-dressed state scheme, the distribution of the effective NV-NV coupling strength is not affected by the Rabi driving field magnitude inhomogeneity. As the Rabi frequency is linearly proportional to the applied oscillating magnetic field strength, the ratio $r = \Omega_+/\Omega_-$ between the Rabi frequencies of the two transitions used in the doubly-dressed state scheme ($\Omega_+ : |0\rangle \leftrightarrow |+\rangle$ and $\Omega_- : |0\rangle \leftrightarrow |-\rangle$) is linearly proportional to the ratio between the double driving field strengths, which are generated from a signal source. One of the advantages of this scheme is that once $\Omega_\pm \gg \nu_{dip}$ is satisfied, the effective NV-NV dipolar coupling strength ν_{eff} only depends on the ratio r. Recalling that the main source of inhomogeneity of an NV Rabi driving frequency arises from the microwave field strength being inversly proportional to the distance between an NV spin and the signal source, and also noting that the NV-NV distance in the present experiment is much less than this distance to the microwave signal source, the ratio *r* between the two driving field used in the doubly-dressed state scheme is robust to any practical inhomogeneity in field amplitudes, given that the signal source for the fields is identical.

No phase-matching condition required for doubly-dressed state scheme — The doubly-dressed state scheme presented in this work does not require any phase-matching condition, since the eigenvalue of the relevant (below) Hamiltonian is always $\sqrt{\Omega_+^2 + \Omega_-^2} S^z$:

$$H = \Omega_+ S_+^x + \Omega_- (\cos(\phi)\, S_-^x + \sin(\phi)\, S_-^y) \tag{33}$$

We also note that the concept of "no phase-locking required" can be interpreted as the effective spin number averaging being solely determined by the relative amplitude of the driving, without the need for a specific phase relation.

Ensemble simulation — To simulate the NV spin ensemble and its spin-spin interaction dynamics when NV spins are prepared in dressed states, we first configure the diamond tetrahedral lattice and adopt a central spin model by placing an NV spin at the origin. The NV spin density is set to 50 ppm, which gives an average NV-NV separation ~5 nm (~50 unit lattice sites); and the bath NV spins are placed at random lattice sites with one of the four NV spin orientations. The size of the simulated NV spin bath is determined to be ~100 unit lattice sites, where the distance from the centre NV spin to the furthest NV spin is comparable to the average NV-NV distance. We let the external magnetic field of 100 G be aligned with the centre NV spin orientation (See Supp. Fig. 1).

For a given spin configuration, we first calculate the centre NV spin to the bath NV spins' overall magnetic dipolar coupling $\Delta^2 = \sum_k \nu_k^2$. We pair the centre NV spin to the k^{th} bath NV spin, with each pair interacting with the spin-spin dipolar interaction Hamiltonian $H_{0k}^{dd} = \frac{-J_0}{r_k^3}(3(\mathbf{S_0} \cdot \hat{\boldsymbol{r}}_k)(\mathbf{S_k} \cdot \hat{\boldsymbol{r}}_k) - \mathbf{S_0} \cdot \mathbf{S_k})$, where $J_0 = (2\pi)$ 52 MHz·nm^3, $\mathbf{S_0}$ is the centre NV spin, and $\mathbf{r_k}$ is a relative displacement vector between the centre NV spin and the k^{th} NV spin. We only consider bath NV spins with different spin orientations to the centre NV spin since spin exchange terms are highly suppressed in the intra-group interaction in the dressed state[2]. For an off-axis pair of NV spins,

$$H_{int} \approx \frac{J_0(1 - 3\cos^2\theta)}{r_k^3} S_0^z S_k^z = \nu_k S_0^z S_k^z \tag{34}$$

where $cos\theta = \hat{z} \cdot \hat{r}_k$ and ν_k is the bare dipolar coupling strength. Note that the other mixing terms (e.g., $S_0^x S_k^x$, etc.) can be neglected due to the external magnetic field being aligned with the centre NV spin and the comparatively large zero-field splitting of 2.87 GHz.

We then pair the bath NV spins and assess pairwise interaction strengths $max(\nu_{ij}) = R_{dd}$, adopting Supp. Eqn. (34). Here, we choose the i^{th} NV bath spin to be aligned with the external magnetic field, and pair it with the nearest neighboring j^{th} off-axis NV bath spin (with the strongest dipolar coupling strength). 50,000 different bath configurations are simulated and contribute to statistical distributions for Δ and R_{dd}.

Next, to simulate the NV ensemble spin bath interaction with the doubly-dressed state scheme, we add double transition driving fields into the total two spin Hamiltonian, and numerically calculate the effective dipolar coupling strengths by varying the Rabi driving strength

parameters. For the calculation of Δ, we numerically solve each spin pair Hamiltonian of the centre NV spin and the k^{th} bath NV spin to extract the effective dipolar coupling strength when the double driving field is applied to the k^{th} bath NV spin. In the double rotating frame, the total Hamiltonian can be simplified as

$$H_{tot} = \Omega_+ I \otimes S_{k,+}^x + \Omega_- I \otimes S_{k,-}^x + \nu_i S_0^z \otimes S_k^z \tag{35}$$

Similar to the previous bare spin bath R_{dd} calculation, we choose the i^{th} NV bath spin to be aligned with the external magnetic field, and pair it with the nearest neighboring j^{th} off-axis NV bath spin in the presence of the double driving fields. Again, for 50,000 different bath configurations, by varying the relative strengths of the double transition driving fields, we extract effective dipolar coupling strengths, which contribute to statistical distributions for Δ and R_{dd}.

Enhanced NV ensemble spin entanglement entropy for the doubly-dressed state scheme — To quantitatively estimate the effect of the doubly-dressed state scheme on improved fidelity of non-classical state generation, we first simulate the time evolution of the total density matrix σ of two NV spins prepared in a superposition state with the dipolar coupling strength given as ν_{dip}, both without and with the double driving field applied[3]. We assume [NV] = 50 ppm for the spin ensemble system; and adopt the NV spin decoherence rate for Hahn-echo and continuous Rabi driven spin-lock (SL) measurements from *Kucsko et. al.*[2]. For a pair of interacting NV spins, we extract the von Neumann entropy at each time step of 50 ns, with the result shown in Supp. Fig. 2a. We use the following equation to calculate the von Neumann entropy:

$$E(\sigma||\rho) = \min_{\rho \in disentangled} Tr(\sigma \ln \sigma/\rho)/\ln 2 \tag{36}$$

where E = 1 indicates a maximally entangled state and E = 0 indicates no entanglement[6]. We confirm that the entanglement entropy of each NV spin pair is modulated with a frequency that is linearly dependent on the dipolar coupling strength ν_{dip}. Furthermore, we find that the entanglement entropy survives longer for Rabi driven cases, due to the extended spin coherence provided by the SL measurement.

Next, to estimate the fidelity of non-classical state generation within the ensemble NV spins, we randomly choose spin pair configurations from the statistical distribution of spin

interactions for different driving conditions; and sum the time evolution of von Neumann entropies over 10,000 spin pairs. Then we normalize the result with the total number of spin pairs. As shown in Supp. Fig. 2b, for the undriven case, the normalized ensemble entanglement entropy is below 0.3 with limited lifetime of < 0.5 μs. However, for the single (Ω_+ = 10 MHz) and double driven (Ω_+ = 10 MHz, Ω_- = 8 MHz) cases, the entanglement entropy almost reaches 0.5 with a significantly extended entanglement lifetime. The doubly-dressed state scheme is thus considered to be more promising for conserving the ensemble spin entanglement lifetime.

Supplementary References

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