

## Supplemental Material: Collective quantum memory activated by a driven central spin

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Here we elaborate on the expressions and results presented in the main text.

### S1. EFFECTIVE LOW-ENERGY HAMILTONIAN FOR ELECTRON AND NUCLEI IN THE PRESENCE OF LATTICE STRAIN

As the starting point of the analysis, we shall consider a singly charged quantum dot subject to a magnetic field of strength  $B$ , perpendicular to the growth axis. We define the  $z$ -direction as the direction of the field and denote the Cartesian electron spin operators in this reference frame by  $S_\alpha$ ,  $\alpha = x, y, z$ . Along with the electron, the quantum dot contains  $N$  nuclei, each with the spin operators  $I_\alpha^j$ ,  $\alpha = x, y, z$ ;  $j = 1, \dots, N$ . In the presence of uniaxial material strain, the quadrupole moment of the nuclear spins will couple to the electric field gradient of the strained lattice, as described by the Hamiltonian [1, 2]

$$H_Q = \sum_j B_Q^j \left[ (I_x^j)^2 \sin^2 \theta^j + \frac{1}{2} (I_x^j I_z^j + I_z^j I_x^j) \sin 2\theta^j + (I_z^j)^2 \cos^2 \theta^j \right], \quad (\text{S1})$$

where  $\theta$  is the angle between the quadrupolar axis and the magnetic field and  $B_Q$  is the strength of the quadrupolar interaction. Furthermore, the electron and nuclei interact via the hyperfine interaction,  $H_{\text{hf}} = \sum_j 2A^j [S_z I_z^j + S_x I_x^j + S_y I_y^j]$ . For appreciable external magnetic field strength, the last two terms in  $H_{\text{hf}}$  describe electron–nucleus flip-flop processes that are perturbatively suppressed a factor of  $A^j / (\omega_Z^e - \omega_Z^n) \ll 1$ , for which reason these terms are typically neglected. In our case, this is well justified by the fact that the leading order perturbative processes governing the electron–nuclear energy exchange occur at a much higher rate, as we shall see. For these reasons, we take  $H_{\text{hf}} = \sum_j 2A^j S_z I_z^j$ . The total Hamiltonian for the electron and nuclear bath is then

$$H = \omega_Z^e S_z + \omega_Z^n \sum_j I_z^j + H_Q + H_{\text{hf}}, \quad (\text{S2})$$

with  $\omega_Z^e$  and  $\omega_Z^n$  the electron and nuclear Zeeman energies, respectively. A simple rotating frame transformation generated by the unitary  $U_R(t) = e^{-i\omega_Z^e S_z t}$  removes the term  $\omega_Z^e S_z$ .

From the quadrupolar interaction,  $H_Q$ , we now extract the contribution that commutes with  $I_z^j$  and thus does not couple different nuclear Zeeman eigenstates, which we denote by  $H_Q^0$ . The remainder,  $V_Q := H_Q - H_Q^0$  is then entirely off-diagonal in the nuclear Zeeman eigenbasis. Specifically, we have [3]

$$\begin{aligned} H_Q^0 &= \sum_j B_Q^j \left\{ \frac{1}{2} [(I_x^j)^2 + (I_y^j)^2] \sin^2 \theta^j + (I_z^j)^2 \cos^2 \theta^j \right\} \\ V_Q &= \sum_j \frac{1}{2} B_Q^j \left\{ [(I_x^j)^2 - (I_y^j)^2] \sin^2 \theta^j + [I_x^j I_z^j + I_z^j I_x^j] \sin 2\theta^j \right\} \end{aligned} \quad (\text{S3})$$

In strong field conditions where  $\omega_Z^n \gg B_Q^j$ , transitions between nuclear Zeeman eigenstates caused by  $V_Q$  are not energetically allowed to first order. To eliminate  $V_Q$  from  $\hat{H}$  and replace it with the appropriate corrections describing energetically allowed processes, we use a Schrieffer-Wolff transformation with generator

$$F = i \sum_j \frac{B_Q^j}{2\omega_Z^n} \left\{ \frac{1}{2} (I_x^j I_y^j + I_y^j I_x^j) \sin^2 \theta^j + (I_y^j I_z^j + I_z^j I_y^j) \sin 2\theta^j \right\}. \quad (\text{S4})$$

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Up to second order in the perturbation parameters  $B_Q^j/\omega_Z^n$  and  $A^j/\omega_Z^n$ , we find the transformed Hamiltonian  $H' = e^F \hat{H} e^{-F} \simeq \hat{H}_e + H'_n + H_{\text{hf}} + V'_Q$ , where [3]

$$H'_n = \omega_Z^n \sum_j I_z^j + H_Q^0 + [F, V_Q], \quad (\text{S5})$$

$$V'_Q = -S_z \sum_j \frac{A^j B_Q^j}{\omega_Z^n} \{ [(I_x^j)^2 - (I_y^j)^2] \sin^2 \theta^j + [I_x^j I_z^j + I_z^j I_x^j] \sin 2\theta^j \}. \quad (\text{S6})$$

Note that  $H'_n$  commutes with  $I_z^j$  and only leads to an anharmonic energy shift of the single-nucleus spin ladders, such that the Zeeman eigenstates of the  $j$ 'th nucleus,  $|m\rangle_j$ , have the energies  $m\omega_Z^n + m^2 \Delta_Q^j$ , where  $\Delta_Q^j = B_Q^j (\cos^2 \theta^j - \frac{1}{2} \sin^2 \theta^j)$ . In contrast,  $V'_Q$  describes a quadrupolar dressing of the hyperfine interaction that generates a noncollinear collective interaction between the electronic and nuclear spins.

### A. Hyperfine coupling distribution

The hyperfine coupling distribution,  $A^j$ , is highly non-uniform due to the inhomogeneous form of the electron wavefunction. For all practical purposes when calculating properties of the system, we obtain the hyperfine distribution numerically by taking the electron density Gaussian,

$$\rho_e(\mathbf{r}) = \prod_{\alpha=x,y,z} \frac{e^{-r_\alpha^2/(2L_\alpha^2)}}{\sqrt{2\pi L_\alpha^2}}, \quad (\text{S7})$$

and evaluating  $\rho_e$  in the points of a cubic lattice of size  $L_x \times L_y \times L_z$ . We have taken parameters for arsenic nuclei in GaAs and a quantum dot with  $L_x = L_y = 10$  nm,  $L_z = 1$  nm, consistent with e.g. Ref. [4].

## S2. PULSE SEQUENCE

By rotating the electron spin with a series of short pulses in conjunction with the free evolution of the system, it is possible to engineer the dynamics of the electron–nuclear system by enhancing and quenching various terms in the Hamiltonian controllably. Here, we adapt a pulse sequence developed for a central spin coupled a nuclear environment with a few, energetically separated spins [5] to our case of a mesoscopic bath with an energetically dense spectrum. The pulse sequence is described in detail in Ref. [5], but for completeness we present the central features here before showing how the sequence acts on the system studied in this work. The pulse cycle of the sequence can be written as

$$y_{\pi/2} \xrightarrow{\tau/4} x_{-\pi} \xrightarrow{\tau/4} y_{\pi/2} \xrightarrow{x_{\pi/2}} y_{\pi} \xrightarrow{\tau/4} x_{\pi/2}, \quad (\text{S8})$$

where  $q_\phi$  ( $q = x, y$ ) denotes a fast coherent rotation of the electron spin corresponding to the unitary operation  $e^{-i\phi S_q}$  and  $\xrightarrow{\tau/4}$  denotes free evolution of the system during the time interval  $\tau/4$ . The unitary evolution operator for a cycle of the sequence can then be expressed as

$$U_C = e^{-i\frac{\tau}{4} S_x} U_0 \left( \frac{\tau}{4} \right) e^{-i\pi S_y} U_0 \left( \frac{\tau}{4} \right) e^{-i\frac{\tau}{4} S_x} e^{-i\frac{\tau}{4} S_y} U_0 \left( \frac{\tau}{4} \right) e^{+i\pi S_x} U_0 \left( \frac{\tau}{4} \right) e^{-i\frac{\tau}{4} S_y}, \quad (\text{S9})$$

where  $U_0(t) = e^{-iH't}$  is the free evolution operator of the system. From Eq. (S9) it can be shown that the time evolution operator over two consecutive cycles can be written as the dynamics generated by a time dependent Hamiltonian,  $\mathcal{H}(t)$ , such that  $U_C^2 = \mathcal{T} e^{-i \int_0^{2\tau} ds \mathcal{H}(s)}$ , where  $\mathcal{T}$  is the chronological time-ordering operator. The pulse sequence Hamiltonian,  $\mathcal{H}(t)$ , is obtained as  $H'$  under the substitution  $S_z \rightarrow h_x(t) S_x + h_y(t) S_y$ , with the piecewise constant functions, which in the interval  $t \in [0, 2\tau]$  take the values

$$h_x(t) = \begin{cases} 0, & t \in [0, \tau/2[ \\ -1, & t \in [\tau/2, 3\tau/4[ \\ +1, & t \in [3\tau/4, \tau[ \\ 0, & t \in [\tau, 3\tau/2[ \\ +1, & t \in [3\tau/2, 7\tau/4[ \\ -1, & t \in [7\tau/4, 2\tau[ \end{cases} \quad (\text{S10})$$

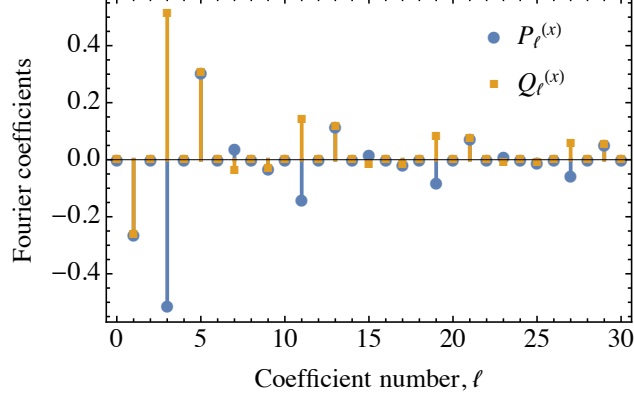


FIG. S1. Fourier coefficients for the pulse modulation function  $h_x$

with  $h_y(t) = h_x(t + \tau/2)$ . The time-dependent functions feature the periodic property  $h_x(t + 2\tau\ell) = h_x(t)$ ,  $h_y(t + 2\tau\ell) = h_y(t)$ , where  $\ell = 0, 1, \dots$ . Due to this periodicity, we can write the  $h_\alpha$ -functions in terms of their discrete Fourier series as

$$h_\alpha(t) = \sum_{\ell=0}^{\infty} P_\ell^{(\alpha)} \cos(\omega_\ell t) + Q_\ell^{(\alpha)} \sin(\omega_\ell t), \quad (\text{S11})$$

with  $\omega_\ell = \pi\ell/\tau$ . The Fourier components are calculated from  $h_\alpha$  as

$$\begin{aligned} P_\ell^{(\alpha)} &= \frac{1}{\tau} \int_0^{2\tau} h_\alpha(t) \cos(\omega_\ell t) dt \\ Q_\ell^{(\alpha)} &= \frac{1}{\tau} \int_0^{2\tau} h_\alpha(t) \sin(\omega_\ell t) dt, \end{aligned} \quad (\text{S12})$$

which due to the relation between  $h_x$  and  $h_y$  satisfy  $P_\ell^{(x)} = P_\ell^{(y)}$ ,  $Q_\ell^{(x)} = -Q_\ell^{(y)}$ . Furthermore, the coefficients are only nonzero for odd  $\ell$ . These coefficients are plotted in Fig. S1, which shows that the  $\ell = 3$  coefficients are largest. For these maximal coefficients, we have

$$P_3^{(x)} = -Q_3^{(x)} = -\frac{2+2\sqrt{2}}{3\pi} \simeq -0.51 \quad (\text{S13})$$

For now, we assume that the quadrupolar energy shift contained in  $H'_n$  is negligible compared to  $\omega_Z^n$ , which can be ensured if the angle of the strain axis relative to the magnetic field is close to  $\theta_0 = \arctan(\sqrt{2})$ . This assumption will be relaxed later to investigate the impact of arbitrary strain axes and strain inhomogeneities on the memory protocol. We can then write the pulse sequence Hamiltonian as  $\mathcal{H}(t) \simeq \mathcal{H}_0 + \mathcal{H}_I(t)$ ,

$$\mathcal{H}_0 = \omega_Z^n \sum_j I_z^j, \quad \mathcal{H}_I(t) = -[h_x(t)S_x + h_y(t)S_y] \left\{ \sum_j A^j I_z^j + \mathcal{A}_1(\Phi_1^+ + \Phi_1^-) + \mathcal{A}_2(\Phi_2^+ + \Phi_2^-) \right\}, \quad (\text{S14})$$

where  $\Phi_\zeta^+ = (\Phi_\zeta^-)^\dagger$  and

$$\Phi_1^- := \sum_j a_{1,j} (I_-^j I_z^j + I_z^j I_-^j), \quad \mathcal{A}_1 := \frac{1}{2} \sum_j \frac{A^j B_Q^j \sin 2\theta^j}{\omega_Z^n}, \quad a_{1,j} := \frac{1}{\mathcal{A}_1} \frac{A^j B_Q^j \sin 2\theta^j}{\omega_Z^n}, \quad (\text{S15})$$

$$\Phi_2^- := \sum_j a_{2,j} (I_-^j)^2, \quad \mathcal{A}_2 := \frac{1}{2} \sum_j \frac{A^j B_Q^j \sin^2 \theta^j}{\omega_Z^n}, \quad a_{2,j} := \frac{1}{\mathcal{A}_2} \frac{A^j B_Q^j \sin^2 \theta^j}{\omega_Z^n}, \quad (\text{S16})$$

such that  $\sum_j a_{\zeta,j} = 1$ . Note that in the free evolution between the Raman pulses, the drives are absent and the rotating frame can be defined with respect to an arbitrary frequency, which we set to  $\omega_Z^e$ , leading to  $\Delta = 0$ . Moving into the interaction picture set by  $\mathcal{H}_0$ , the interaction Hamiltonian is

$$\hat{\mathcal{H}}_I(t) = e^{i\mathcal{H}_0 t} \mathcal{H}_I(t) e^{-i\mathcal{H}_0 t} = -[h_x(t)S_x + h_y(t)S_y] \left\{ \sum_j A^j I_z^j + \mathcal{A}_1 (e^{+i\omega_Z^e t} \Phi_1^+ + e^{-i\omega_Z^e t} \Phi_1^-) + \mathcal{A}_2 (e^{+2i\omega_Z^e t} \Phi_2^+ + e^{-2i\omega_Z^e t} \Phi_2^-) \right\}. \quad (\text{S17})$$

At this point, we express  $h_\alpha(t)$  in terms of their Fourier expansions,

$$\hat{\mathcal{H}}_I(t) = - \sum_{\ell=1}^{\infty} \left\{ [P_\ell^{(x)} \cos(\omega_\ell t) + Q_\ell^{(x)} \sin(\omega_\ell t)] S_x + [P_\ell^{(x)} \cos(\omega_\ell t) - Q_\ell^{(x)} \sin(\omega_\ell t)] S_y \right\} \\ \times \left\{ \sum_j A^j I_z^j + \mathcal{A}_1 (e^{+i\omega_Z^e t} \Phi_1^+ + e^{-i\omega_Z^e t} \Phi_1^-) + \mathcal{A}_2 (e^{+2i\omega_Z^e t} \Phi_2^+ + e^{-2i\omega_Z^e t} \Phi_2^-) \right\}. \quad (\text{S18})$$

Furthermore, we assume that the delay time of the pulse sequence,  $\tau$ , is chosen such that one of the of the discrete Fourier frequencies,  $\omega_\ell$ , is close to either  $\omega_Z^e$  or  $2\omega_Z^e$ . We shall denote this particular  $\ell$ -index by  $\ell^*$  and say that  $\omega_{\ell^*} = \zeta\omega_Z^e$ , where  $\zeta$  is either 1 or 2. This resonance of the pulse sequence is obtained by setting  $\tau = \pi\ell^*/(\zeta\omega_Z^e)$ . As a result, the Fourier frequencies,  $\omega_\ell$ , are separated by  $\Delta\omega = \zeta\omega_Z^e/\ell^*$ . Provided that  $\ell^*$  is of order unity, all terms in  $\hat{\mathcal{H}}_I(t)$  that are not resonant with  $\zeta\omega_Z^e$  will average to zero in the dynamical evolution of the system. In particular, there does not exist a pair of odd indices  $(\ell_1, \ell_2)$  such that  $\ell_1\Delta\omega = \omega_Z^e$  and  $\ell_2\Delta\omega = 2\omega_Z^e$ . Since the Fourier coefficients  $P_\ell^{(\alpha)}$  and  $Q_\ell^{(\alpha)}$  are only nonzero for odd  $\ell$ , this means that the pulse sequence will never be resonant with both collective nuclear transitions simultaneously. Removing all rapidly rotating terms from Eq. (S18) leaves us with

$$\hat{\mathcal{H}}_I(t) \simeq -\mathcal{A}_{\zeta} \frac{1}{2} \Phi_{\zeta}^+ [P_{\ell^*}^{(x)} (S_x + S_y) + iQ_{\ell^*}^{(x)} (S_x - S_y)] + \mathcal{A}_{\zeta} \frac{1}{2} \Phi_{\zeta}^- [P_{\ell^*}^{(x)} (S_x + S_y) - iQ_{\ell^*}^{(x)} (S_x - S_y)]. \quad (\text{S19})$$

At this point, we choose  $\ell^* = 3$  which we found to have the largest Fourier component. Further, we rotate the electron spin coordinates by an angle of  $\pi/4$  in the  $xy$ -plane, such that  $S_x \rightarrow (S_x + S_y)/\sqrt{2}$ ,  $S_y \rightarrow (S_x - S_y)/\sqrt{2}$ , leading to

$$\hat{\mathcal{H}}_I \simeq \frac{\sqrt{2}+2}{3\pi} \mathcal{A}_{\zeta} (\Phi_{\zeta}^+ S_- + \Phi_{\zeta}^- S_+). \quad (\text{S20})$$

### S3. NUCLEAR CHAIN OF STATES UNDER PULSE SEQUENCE DYNAMICS

The pulse sequence interaction Hamiltonian,  $\hat{\mathcal{H}}_I$ , in Eq. (S20) describes the interaction between the electron and the nuclear spin bath. Due to its secular form, many of the terms in the expansion of the time evolution operator,  $\mathcal{U}$ , become zero, because  $S_+^2 = S_-^2 = 0$ . As a result, we can write  $\mathcal{U}$  as

$$\mathcal{U}(t) = e^{-i\hat{\mathcal{H}}_I t} = \mathbb{I} + \left( \frac{-i\mathcal{A}_{\zeta} t}{2} \right)^2 (\Phi_{\zeta}^+ S_- + \Phi_{\zeta}^- S_+) + \frac{1}{2!} \left( \frac{-i\mathcal{A}_{\zeta} t}{2} \right)^2 (\Phi_{\zeta}^- \Phi_{\zeta}^+ S_+ S_- + \Phi_{\zeta}^+ \Phi_{\zeta}^- S_- S_+) \\ + \frac{1}{3!} \left( \frac{-i\mathcal{A}_{\zeta} t}{2} \right)^3 (\Phi_{\zeta}^+ \Phi_{\zeta}^- \Phi_{\zeta}^+ S_- S_+ S_- + \Phi_{\zeta}^- \Phi_{\zeta}^+ \Phi_{\zeta}^- S_+ S_- S_+) + \dots \\ = \sum_{k=0}^{\infty} \frac{1}{(2k)!} \left( \frac{-i\mathcal{A}_{\zeta} t}{2} \right)^{2k} \{ (\Phi_{\zeta}^+ \Phi_{\zeta}^-)^k (S_- S_+)^k + (\Phi_{\zeta}^- \Phi_{\zeta}^+)^k (S_+ S_-)^k \} \\ + \sum_{k=0}^{\infty} \frac{1}{(2k+1)!} \left( \frac{-i\mathcal{A}_{\zeta} t}{2} \right)^{2k+1} \{ \Phi_{\zeta}^- (\Phi_{\zeta}^+ \Phi_{\zeta}^-)^k S_+ (S_- S_+)^k + \Phi_{\zeta}^+ (\Phi_{\zeta}^- \Phi_{\zeta}^+)^k S_- (S_+ S_-)^k \} \quad (\text{S21})$$

At this point, we consider the initial nuclear state to be a classical mixture of product states. Denoting a general nuclear product state by  $|M\rangle = |m_1, \dots, m_n\rangle$ , the initial nuclear density operator can be written as  $\rho_n(0) = \sum_M p(M) |M\rangle\langle M|$ , which does not need to be internuclear factorisable, but can generally contain classical correlations, i.e. generally  $\rho_n(0) \neq \bigotimes \rho_j$ . For practical purposes, we can then calculate the dynamics for a given state  $|M\rangle$  and subsequently average over the distribution  $p(M)$ . More details about drawing samples from the initial distribution is given in Sec. S3 A. From Eq. (S21), we see that with  $|M\rangle$  as the

initial nuclear state, the evolution of the system will gradually populate the states  $\Phi_\zeta^\pm |M\rangle$ ,  $\Phi_\zeta^\mp \Phi_\zeta^\pm |M\rangle$ ,  $\Phi_\zeta^\pm \Phi_\zeta^\mp \Phi_\zeta^\pm |M\rangle$  and so forth. Our strategy for calculating the dynamics of the electron–nuclear system is to orthonormalise the set

$$\mathcal{S}_\pm(k^*) := \{(\Phi_\zeta^\mp \Phi_\zeta^\pm)^k |M\rangle, \Phi_\zeta^\pm (\Phi_\zeta^\mp \Phi_\zeta^\pm)^k |M\rangle | k \leq k^*\},$$

containing states generated by the evolution  $\mathcal{U}$  up to a certain truncation index,  $k^*$ . Note that by  $\mathcal{S}_+(k^*)$  and  $\mathcal{S}_-(k^*)$ , we understand two distinct sets, which we wish to orthonormalise separately. We shall define the normalised state  $|M_\pm^{(k)}; \zeta\rangle$  as

$$|M_\pm^{(k)}; \zeta\rangle = \begin{cases} \frac{(\Phi_\zeta^\mp \Phi_\zeta^\pm)^k |M\rangle}{\sqrt{\langle M | (\Phi_\zeta^\mp \Phi_\zeta^\pm)^{2k} | M \rangle}}, & k \text{ even} \\ \frac{\Phi_\zeta^\pm (\Phi_\zeta^\mp \Phi_\zeta^\pm)^{k-1} |M\rangle}{\sqrt{\langle M | (\Phi_\zeta^\mp \Phi_\zeta^\pm)^{2k-1} | M \rangle}}, & k \text{ odd,} \end{cases} \quad (\text{S22})$$

such that  $\bar{\mathcal{S}}_\pm(k^*) := \{|M_\pm^{(k)}; \zeta\rangle | k \leq k^*\}$  is simply the normalised form of  $\mathcal{S}_\pm(k^*)$ . The goal is now to perform Gram-Schmidt orthonormalisation to  $\bar{\mathcal{S}}_\pm(k^*)$  in order to derive the orthonormal set  $\hat{\mathcal{S}}_\pm(k^*)$ , whose elements we shall denote by  $|\hat{M}_\pm^{(k)}; \zeta\rangle$ . First, we see that  $|M\rangle = |M_\pm^{(0)}; \zeta\rangle$  and define  $|\hat{M}_\pm^{(0)}; \zeta\rangle := |M\rangle$ . The Gram-Schmidt orthonormalisation strategy now gives the recursive relation

$$|\hat{M}_\pm^{(1)}; \zeta\rangle = \frac{(\mathbb{I} - |M_\pm^{(0)}; \zeta\rangle \langle M_\pm^{(0)}; \zeta|) |M_\pm^{(1)}; \zeta\rangle}{\sqrt{\langle M_\pm^{(1)}; \zeta | (\mathbb{I} - |M_\pm^{(0)}; \zeta\rangle \langle M_\pm^{(0)}; \zeta|) |M_\pm^{(1)}; \zeta\rangle}}, \quad (\text{S23})$$

$$|\hat{M}_\pm^{(k)}; \zeta\rangle = \frac{(\mathbb{I} - \sum_{l=0}^{k-1} |\hat{M}_\pm^{(l)}; \zeta\rangle \langle \hat{M}_\pm^{(l)}; \zeta|) |M_\pm^{(k)}; \zeta\rangle}{\sqrt{\langle M_\pm^{(k)}; \zeta | (\mathbb{I} - \sum_{l=0}^{k-1} |\hat{M}_\pm^{(l)}; \zeta\rangle \langle \hat{M}_\pm^{(l)}; \zeta|) |M_\pm^{(k)}; \zeta\rangle}}. \quad (\text{S24})$$

The structure of the orthonormal set  $\hat{\mathcal{S}}(k^*)$  becomes clear when writing the states out explicitly. To do so, we first introduce the convenient shorthand notation

$$|M; (\Delta, j), (\Delta', j'), \dots\rangle := |m_1, \dots, (m_j + \Delta), \dots, (m_{j'} + \Delta'), \dots, m_n\rangle, \quad (\text{S25})$$

and the prefactors (not to be confused with the Fourier coefficients in Sec. S2)

$$\begin{aligned} P_\pm^{(1)}(m) &= (2m \pm 1) \sqrt{I(I+1) - m(m \pm 1)} \\ P_\pm^{(2)}(m) &= \sqrt{I(I+1) - m(m \pm 1)} \sqrt{I(I+1) - (m \pm 1)(m \pm 2)}, \end{aligned} \quad (\text{S26})$$

such that  $\Phi_\zeta^\pm |M\rangle = \sum_j a_{\zeta,j} P_\pm^{(\zeta)}(m_j) |M; (\pm\zeta, j)\rangle$ . Note that the prefactor  $P_\pm^{(\zeta)}(m)$  automatically becomes zero if the transition  $m \rightarrow m \pm \zeta$  is not allowed. The first four states in  $\hat{\mathcal{S}}_+(k^*)$  and  $\hat{\mathcal{S}}_-(k^*)$  can then be written as

$$\begin{aligned} |M_\pm^{(0)}; \zeta\rangle &= |M\rangle \\ |\hat{M}_\pm^{(1)}; \zeta\rangle &= \frac{1}{\mathcal{N}_\pm^{(1)}(M; \zeta)} \sum_j a_{\zeta,j} P_\pm^{(\zeta)}(m_j) |M; (\pm\zeta, j)\rangle, \\ |\hat{M}_+^{(2)}; \zeta\rangle &= |\hat{M}_-^{(2)}; \zeta\rangle = \frac{1}{\mathcal{N}_+^{(2)}(M; \zeta)} \sum_{\langle j_1 j_2 \rangle} a_{\zeta, j_1} a_{\zeta, j_2} P_+^{(\zeta)}(m_{j_1}) P_-^{(\zeta)}(m_{j_2}) |M; (+\zeta, j_1), (-\zeta, j_2)\rangle, \\ |\hat{M}_+^{(3)}; \zeta\rangle &= \frac{1}{\mathcal{N}_+^{(3)}(M; \zeta)} \left\{ \sum_{\langle j_1 j_2 j_3 \rangle} a_{\zeta, j_1} a_{\zeta, j_2} a_{\zeta, j_3} P_+^{(\zeta)}(m_{j_1}) P_-^{(\zeta)}(m_{j_2}) P_+^{(\zeta)}(m_{j_3}) |M; (+\zeta, j_1), (-\zeta, j_2), (+\zeta, j_3)\rangle \right. \\ &\quad \left. + \sum_{\langle j_1 j_2 \rangle} a_{j_1}^2 a_{j_2} P_+^{(\zeta)}(m_{j_1}) P_+^{(\zeta)}(m_{j_1} + \zeta) P_-^{(\zeta)}(m_{j_2}) |M; (+2\zeta, j_1), (-\zeta, j_2)\rangle \right\}, \\ |\hat{M}_-^{(3)}; \zeta\rangle &= \frac{1}{\mathcal{N}_-^{(3)}(M; \zeta)} \left\{ \sum_{\langle j_1 j_2 j_3 \rangle} a_{\zeta, j_1} a_{\zeta, j_2} a_{\zeta, j_3} P_+^{(\zeta)}(m_{j_1}) P_-^{(\zeta)}(m_{j_2}) P_+^{(\zeta)}(m_{j_3}) |M; (+\zeta, j_1), (-\zeta, j_2), (-\zeta, j_3)\rangle \right. \\ &\quad \left. + \sum_{\langle j_1 j_2 \rangle} a_{j_1} a_{j_2}^2 P_+^{(\zeta)}(m_{j_1}) P_-^{(\zeta)}(m_{j_2}) P_-^{(\zeta)}(m_{j_2} - \zeta) |M; (+\zeta, j_1), (-2\zeta, j_2)\rangle \right\}, \end{aligned} \quad (\text{S27})$$

where  $\mathcal{N}_\pm^{(k)}(M; \zeta)$  is a normalisation factor and  $\sum_{\langle j_1 \dots j_n \rangle}$  denotes a summation over the  $n$  indices (each running from 1 to  $N$ ) that only includes terms where no pair of indices are equal. Extending this sequence of states to higher values of  $k$  is a tedious, but straightforward task. For the special case of  $I = 3/2$ , the situation vastly simplifies due to the identity  $P_\pm^{(\zeta)}(m)P_\pm^{(\zeta)}(m \pm 1) = 0$ , thus eliminating the possibility of multiple noncollinear excitations of the same nuclear spin. As a result, for  $I = 3/2$ , we can write any state in  $\hat{\mathcal{S}}(k^*)$  in a general form as

$$|\hat{M}_\pm^{(k)}; \zeta\rangle = \frac{\sum_{\langle j_1 \dots j_k \rangle} a_{\zeta,1} \dots a_{\zeta,k} P_\pm^{(\zeta)}(m_{j_1}) P_\mp^{(\zeta)}(m_{j_2}) \dots P_{\pm\lambda_k}^{(\zeta)}(m_{j_k}) |M; (\pm\zeta, j_1), (\mp\zeta, j_2), \dots, (\pm\lambda_k \zeta, j_k)\rangle}{\sqrt{\sum_{\langle j_1 \dots j_k \rangle} [a_{\zeta,1} \dots a_{\zeta,k} P_\pm^{(\zeta)}(m_{j_1}) P_\mp^{(\zeta)}(m_{j_2}) \dots P_{\pm\lambda_k}^{(\zeta)}(m_{j_k})]^2}}, \quad (\text{S28})$$

where  $\lambda_k = (-1)^{k+1}$ . In general, we find that for all even values of  $k$ ,  $|\hat{M}_+^{(k)}; \zeta\rangle = |\hat{M}_-^{(k)}; \zeta\rangle$ , and we might thus drop the  $\pm$ -index on  $|\hat{M}_\pm^{(k)}; \zeta\rangle$  for even  $k$ .

For the purpose of calculating the dynamics, we are generally interested in knowing how the interaction Hamiltonian,  $\hat{\mathcal{H}}_I$ , couples the different states in  $\hat{\mathcal{S}}_\pm(k^*)$ . The general structure of  $\hat{\mathcal{S}}(k^*)$  as presented in Eqs. (S27) and (S28), leads to the selection rule

$$\langle \hat{M}_\alpha^{(k)}; \zeta | \Phi_\zeta^\beta | \hat{M}_\gamma^{(k')}; \zeta \rangle = 0 \text{ if } |k - k'| \neq 1, \quad (\text{S29})$$

meaning that we only need to evaluate transition matrix elements between neighbouring states in  $\hat{\mathcal{S}}_\pm(k^*)$ . For general  $I$ , we find the elements between the states in Eq. (S27)

$$\begin{aligned} \langle \hat{M}_\pm^{(1)}; \zeta | \Phi_\zeta^\pm | M^{(0)}; \zeta \rangle &= \sqrt{\sum_j [a_{\zeta,j} P_\pm(m_j)]^2} =: \Omega_\pm(M; \zeta), \\ \langle \hat{M}_\pm^{(1)}; \zeta | \Phi_\zeta^\mp | M^{(0)}; \zeta \rangle &= 0, \\ \langle \hat{M}_\pm^{(2)}; \zeta | \Phi_\zeta^\pm | \hat{M}_\pm^{(1)}; \zeta \rangle &= 0, \\ \langle \hat{M}_\pm^{(2)}; \zeta | \Phi_\zeta^\mp | \hat{M}_\pm^{(1)}; \zeta \rangle &= \frac{1}{\Omega_\pm(M; \zeta)} \sqrt{\sum_{\langle j_1 j_2 \rangle} [a_{\zeta,j_1} a_{\zeta,j_2} P_+(m_{j_1}) P_-(m_{j_2})]^2}, \\ \langle \hat{M}_\pm^{(3)}; \zeta | \Phi_\zeta^\mp | \hat{M}_\pm^{(2)}; \zeta \rangle &= 0, \\ \langle \hat{M}_+^{(3)}; \zeta | \Phi_\zeta^+ | \hat{M}_+^{(2)}; \zeta \rangle &= \left\{ \sum_{\langle j_1 j_2 \rangle} [a_{\zeta,j_1} a_{\zeta,j_2} P_+(m_{j_1}) P_-(m_{j_2})]^2 \right\}^{-1/2} \left\{ \sum_{\langle j_1 j_2 j_3 \rangle} [a_{\zeta,j_1} a_{\zeta,j_2} a_{\zeta,j_3} P_+^{(\zeta)}(m_{j_1}) P_-^{(\zeta)}(m_{j_2}) P_+^{(\zeta)}(m_{j_3})]^2 \right. \\ &\quad \left. + \sum_{\langle j_1 j_2 \rangle} [a_{j_1}^2 a_{j_2} P_+^{(\zeta)}(m_{j_1}) P_+^{(\zeta)}(m_{j_1} + \zeta) P_-^{(\zeta)}(m_{j_2})]^2 \right\} \\ \langle \hat{M}_-^{(3)}; \zeta | \Phi_\zeta^- | \hat{M}_-^{(2)}; \zeta \rangle &= \left\{ \sum_{\langle j_1 j_2 \rangle} [a_{\zeta,j_1} a_{\zeta,j_2} P_+(m_{j_1}) P_-(m_{j_2})]^2 \right\}^{-1/2} \left\{ \sum_{\langle j_1 j_2 j_3 \rangle} [a_{\zeta,j_1} a_{\zeta,j_2} a_{\zeta,j_3} P_+^{(\zeta)}(m_{j_1}) P_-^{(\zeta)}(m_{j_2}) P_-^{(\zeta)}(m_{j_3})]^2 \right. \\ &\quad \left. + \sum_{\langle j_1 j_2 \rangle} [a_{j_1}^2 a_{j_2} P_-^{(\zeta)}(m_{j_1}) P_-^{(\zeta)}(m_{j_1} - \zeta) P_+^{(\zeta)}(m_{j_2})]^2 \right\}. \end{aligned} \quad (\text{S30})$$

Because  $\Phi^- = (\Phi^+)^\dagger$ , we only need to find the matrix elements for one of the two operators, since the elements of the other will be given thereby. Here we find the elements of  $\Phi^+$ .

For  $N \gg 1$  and  $k \ll N$ , we find the approximation

$$\begin{aligned} \langle \hat{M}_\pm^{(k'+2)} | \Phi^+ | \hat{M}_\pm^{k+2} \rangle &\simeq \langle \hat{M}_\pm^{(k')} | \Phi^+ | \hat{M}_\pm^k \rangle \\ \langle \hat{M}_\pm^{(k'+1)} | \Phi^+ | \hat{M}_\pm^{k+1} \rangle &\simeq \langle \hat{M}_\mp^{(k')} | \Phi^+ | \hat{M}_\mp^k \rangle \end{aligned} \quad (\text{S31})$$

For the realistic situations studied in the present work, the relative error of this approximation is below  $10^{-4}$ . Under the approximation (S31), the matrix elements can be generalised as

$$\langle \hat{M}_+^{(k')} | \Phi^+ | \hat{M}_+^{(k)} \rangle = \begin{cases} 0 & k' \text{ even} \\ \Omega_+(M; \zeta) \delta_{k', k+1} + \Omega_-(M; \zeta) \delta_{k', k-1} & k' \text{ odd} \end{cases} \quad (\text{S32})$$

$$\langle \hat{M}_-^{(k')} | \Phi^+ | \hat{M}_-^{(k)} \rangle = \begin{cases} \Omega_+(M; \zeta) \delta_{k', k+1} + \Omega_-(M; \zeta) \delta_{k', k-1} & k' \text{ even} \\ 0 & k' \text{ odd} \end{cases} \quad (\text{S33})$$

The interaction Hamiltonian can then be expanded on the derived basis in the form

$$\hat{\mathcal{H}}_I = S_- \sum_n \mathcal{G}_- |\hat{M}_+^{(2n-1)}\rangle \langle \hat{M}_+^{(2n)}| + \mathcal{G}_+ |\hat{M}_+^{(2n+1)}\rangle \langle \hat{M}_+^{(2n)}| + \mathcal{G}_+ |\hat{M}_-^{(2n)}\rangle \langle \hat{M}_-^{(2n-1)}| + \mathcal{G}_- |\hat{M}_-^{(2n)}\rangle \langle \hat{M}_-^{(2n+1)}| + \text{H.c.} \quad (\text{S34})$$

where the coupling rates are given by  $\mathcal{G}_\pm := \frac{2+\sqrt{2}}{3\pi} \mathcal{A}_\zeta \Omega_\pm$ , suppressing explicit dependence of  $\Omega_\pm$  on  $M$  and  $\zeta$ .

### A. Sampling from initial nuclear distributions

When performing calculations on the dynamics of the system, it is necessary to draw samples of initial states,  $|M\rangle$ , from a distribution,  $p(M)$ . In this work, we take this distribution to be thermal at inverse temperature  $\beta = 1/(k_B T)$ , such that the initial nuclear density operator is  $\rho_n(0) = \bigotimes_j e^{-\beta \omega_z^n I_z^j} / \text{Tr} \left[ e^{-\beta \omega_z^n I_z^j} \right]$ . Here, we have ignored the effect of the quadrupolar interaction on the thermal populations, assuming  $\omega_z^n \ll B_Q$ . The probability distribution  $p(M)$  is then given by

$$p(M) = \langle M | \rho_n(0) | M \rangle = \prod_j p(m_j), \quad (\text{S35})$$

where

$$p(m_j) = \frac{e^{-\beta \omega_z^n m_j}}{\sum_{m=-I_j}^{I_j} e^{-\beta \omega_z^n m}}. \quad (\text{S36})$$

A sample  $M$  is then obtained by drawing  $N$  single-nucleus quantum numbers  $\{m_j\}$  from the distribution  $p(m_j)$ . The ensemble polarisation is linked to the temperature through the relation  $P = \frac{1}{I} \sum_{m=-I}^I m p(m)$ .

## S4. QUADRUPOLAR INHOMOGENEITIES

In the presence of quadrupolar inhomogeneities, the term  $H_Q^0 = \sum_j \Delta_Q^j (I_z^j)^2$  in Eq. (S5) will be non-zero, and the quadrupolar shift,  $\Delta_Q^j$  will be described by a statistical distribution over all the nuclei. This term will be carried onto the interaction Hamiltonian in Eq. (S20), which then becomes  $\hat{\mathcal{H}}_I' = \hat{\mathcal{H}}_I + H_Q^0$ . To study the effect of this, we consider a fully polarised initial nuclear state, such that the dynamics in the absence of inhomogeneities is spanned by the nuclear states  $|\mathbf{0}\rangle$  and  $|\mathbf{1}\rangle := |\hat{\mathbf{0}}_+^{(1)}; \zeta\rangle$ . The ground state,  $|\mathbf{0}\rangle$  is an eigenstate of  $H_Q^0$ , but the collective excitation  $|\mathbf{1}\rangle$  is not. The effect of  $H_Q^0$  is then to rotate  $|\mathbf{1}\rangle$  into a set of orthogonal collective excitations, which do not interact with the electron through  $\mathcal{H}_I$ . To demonstrate this, we define an orthonormal basis for  $\mathbb{C}^N$ ,  $\{v_\alpha | \alpha = 1, \dots, N\}$ , such that  $\sum_j v_{\alpha,j}^* v_{\beta,j} = \delta_{\alpha\beta}$ . We choose the first vector to be  $v_{1,j} = a_j / \sqrt{\sum_j a_j^2}$ . These vectors can then be mapped onto a complete basis of spin waves with Zeeman energy  $\zeta \omega_z^n$ ,

$$|\alpha\rangle := \sum_j v_{\alpha,j} |\mathbf{0}; (\zeta, j)\rangle, \quad (\text{S37})$$

such that  $|\mathbf{1}\rangle$  corresponds to  $|\alpha = 1\rangle$ . Of all these spin waves, only  $|\mathbf{1}\rangle$  is coupled to  $|\mathbf{0}\rangle$  via  $\mathcal{H}_I$ :

$$\langle \mathbf{0} | \Phi_\zeta^- | \alpha \rangle = P_-^{(\zeta)} (-I + \zeta) \sum_j a_j v_{\alpha,j} = \delta_{\alpha,1} P_-^{(\zeta)} (-I + \zeta) \sqrt{\sum_j a_j^2}. \quad (\text{S38})$$

and  $\langle \mathbf{0} | \Phi_\zeta^+ | \alpha \rangle = \langle \beta | \Phi_\zeta^\pm | \alpha \rangle = 0$ . The diffusion rate from  $|\mathbf{1}\rangle$  into this dark subspace,  $\gamma$ , is approximated by calculating the time evolution of  $|\mathbf{1}\rangle$  under  $H_Q^0$  and projecting back onto  $|\mathbf{1}\rangle$ :

$$\langle \mathbf{1} | e^{-iH_Q^0 t} | \mathbf{1} \rangle = \frac{\sum_j a_j^2 e^{-i\Delta_Q^j \zeta^2 t}}{\sum_j a_j^2}. \quad (\text{S39})$$

Assuming statistical independence of  $a_j$  and  $\Delta_Q^j$  and taking the ensemble distribution of  $\Delta_Q^j$  as the normal distribution  $p(\Delta_Q) = e^{-\Delta_Q/(2\sigma^2)} / \sqrt{2\pi\sigma^2}$ , we find

$$\langle \mathbf{1} | e^{-iH_Q^0 t} | \mathbf{1} \rangle \simeq \int_{-\infty}^{\infty} d\Delta_Q p(\Delta_Q) e^{-i\Delta_Q \zeta^2 t} = e^{-\frac{1}{2}(\zeta^2 \sigma)^2 t^2}, \quad (\text{S40})$$

such that the  $1/e$  decay rate for the population,  $|\langle \mathbf{1} | e^{-iH_Q^0 t} | \mathbf{1} \rangle|^2$ , is  $\gamma = \zeta^2 \sigma$ .

To include this effect in the dynamical evolution when assessing the read/write error, we note that the electron-nuclear state  $|\phi\rangle|\alpha\rangle$  with  $|\alpha\rangle \neq |\mathbf{1}\rangle$  is an eigenstate of the transfer-generating interaction,  $\hat{\mathcal{H}}_I$  and thus fully equivalent to  $|\phi\rangle|\mathbf{0}\rangle$  when studying the retrieved state of the electron [6].

### A. Collective refocusing sequence

Here we briefly demonstrate, by adapting the analysis in Ref. [7], how the collective memory excitation can be refocused to compensate for quadrupolar inhomogeneities during storage. We start out by considering the nuclear spin bath immediately after the electron state has been transferred to it. Its initial state is then  $|\psi_0\rangle = \alpha|\mathbf{0}\rangle + \beta|\mathbf{1}\rangle$ , where  $|\mathbf{0}\rangle$  and  $|\mathbf{1}\rangle$  are defined in the main text. Assuming that we wish to read out the nuclear state at time  $t = T$ , we start out by letting the system evolve until time  $t = T/2$ , where the state is

$$|\psi(T/2)\rangle = \alpha|\mathbf{0}\rangle + \beta \sum_j a_j e^{-i\zeta^2 \Delta_Q^j(T/2)} |-I, \dots, (-I+\zeta), \dots, -I\rangle, \quad (\text{S41})$$

where the zero-point energy for the  $j^{\text{th}}$  nucleus has been defined as the energy of  $|-I\rangle_j$ . At this time, a  $\pi$ -rotation within the  $(-I, -I+\zeta)$  subspace is performed using nuclear magnetic resonance (with two pulses needed for  $\zeta = 2$ ) [8], and the state becomes

$$|\psi(T/2+0^+)\rangle = \alpha |-I+\zeta, \dots, -I+\zeta\rangle + \beta \sum_j a_j e^{-i\zeta^2 \Delta_Q^j(T/2)} |-I+\zeta, \dots, (-I)_j, \dots, -I+\zeta\rangle. \quad (\text{S42})$$

After this rotation, the state evolves until time  $t = T$ ,

$$|\psi(T)\rangle = \alpha e^{-iE_0(T/2)} |-I+\zeta, \dots, -I+\zeta\rangle + \beta \sum_j a_j e^{-i\zeta^2 \Delta_Q^j(T/2)} e^{-iE_0^j(T/2)} |-I+\zeta, \dots, (-I)_j, \dots, -I+\zeta\rangle, \quad (\text{S43})$$

where  $E_0 := \zeta^2 \sum_j \Delta_Q^j$  and  $E_0^j := \zeta^2 \sum_{i \neq j} \Delta_Q^i$ . Since  $E_0^j + \zeta^2 \Delta_Q^j = E_0$ , the evolution phase factors out as a global phase. Furthermore, once again performing a  $\pi$ -rotation yields the state

$$|\psi(T+0^+)\rangle = e^{-iE_0(T/2)} (\alpha|\mathbf{0}\rangle + \beta|\mathbf{1}\rangle), \quad (\text{S44})$$

which is equal to the initial state up to a global phase.

## S5. DECOUPLING FROM OTHER NUCLEAR SPECIES

In realistic quantum dots, the nuclear spin environment comprises several nuclear spin species with different Zeeman energies and quadrupolar parameters. However, if the transition energies are sufficiently well separated, the electron spin state can be selectively transferred to only one of the species. This selectivity is provided by the pulse sequence, which averages out interactions that are not resonant with the targeted spin wave. To demonstrate that the pulse sequence decouples the unwanted nuclear species, we use a simple few-spin model, containing a single nucleus of each species, which can be solved numerically [9]. By introducing a collective enhancement factor, we expect this few-spin model to phenomenologically describe the collective nature of the nuclear excitations. The Hamiltonian of this multi-species model is  $H = \sum_j \omega_Z^j I_z^j + H_{\text{hf}} + H_Q^0 + V_Q'$ , where the  $j$ -summations run over different nuclear species. To account for the collective enhancement of the interaction, the non-collinear interaction rates,  $A^j B_Q^j / \omega_Z^j$ , in  $V_Q'$  have been scaled up with a factor of  $\sqrt{N^j}$ , where  $N^j$  is the number of nuclei of the species  $j$ . The nuclear Zeeman energies are related to the magnetic field,  $B$ , through their gyromagnetic ratios,  $\omega_Z^j = g_j B$ . The hyperfine interaction strength is taken as an average over the ensemble,  $A^j = \mathcal{A}^j / N$ , where  $\mathcal{A}^j$  is the hyperfine constant of the nuclear species  $j$ .

The electron is initialised in the  $|+1/2\rangle$  state and the nuclei are initialised in the ground state,  $\bigotimes_j |-I^j\rangle$ . We consider a uniformly strained GaAs quantum dot [10] with quadrupolar interaction strength consistent with maximum external strain applied in Ref. [11], where the pulse sequence is chosen to be resonant with the  $|-3/2\rangle \leftrightarrow |-1/2\rangle$  transition of As,  $\tau = 3\pi / [\omega_Z^{\text{As}} + (\zeta^2 - 2I^{\text{As}}\zeta)\Delta_Q^{\text{As}}]$ . To understand the effects of the presence of Ga nuclei, we present in Fig. S2a the spectrum of nuclear transitions that are available from the ground state. Each transition is represented by a Lorentzian line whose width and height are given by the transition matrix element  $\langle -I^j + \zeta | V_Q' | -I^j \rangle$ , where the  $\zeta = 1$  transitions are indicated with solid lines



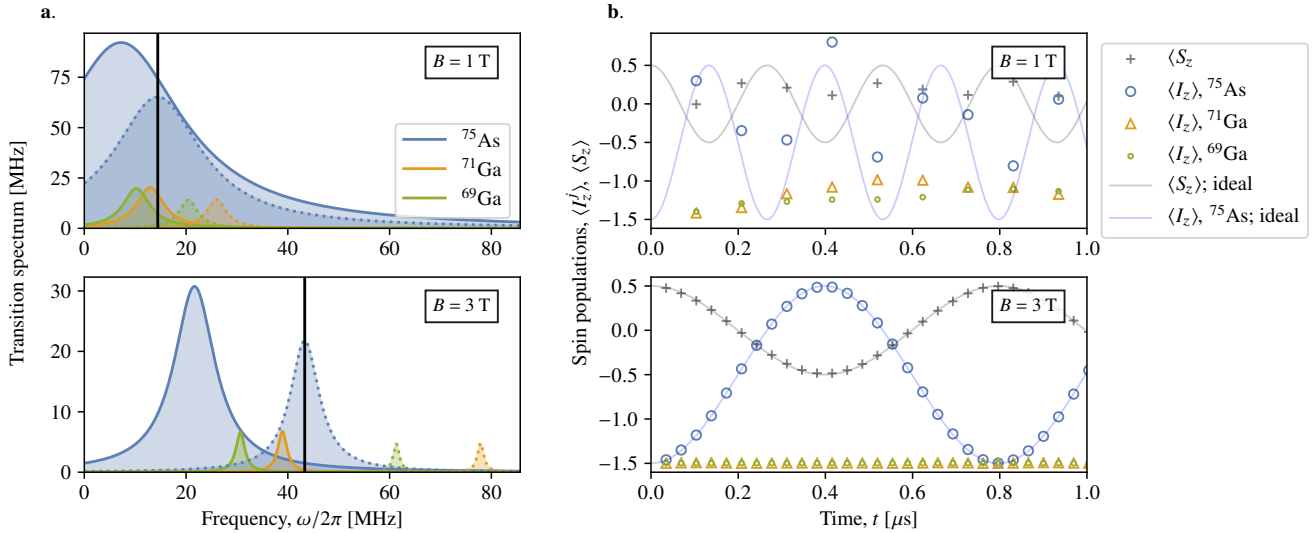


FIG. S2. **a.** Transition spectra, at magnetic fields of 1 T (upper) and 3 T (lower), of the three nuclear species present in GaAs when initialised in the ground state. Solid lines signify the transition  $|-3/2\rangle \leftrightarrow |-1/2\rangle$ , and dotted lines signify  $|-3/2\rangle \leftrightarrow |+1/2\rangle$ . **b.** Time evolution of multiple nuclear spin species under the pulse sequence when resonant with the  $|-3/2\rangle \leftrightarrow |-1/2\rangle$  transition of  $^{75}\text{As}$ . Data points indicate spin populations calculated numerically, and the solid lines indicate the ideal time evolution of the electron and As nuclei as in Eq. (2) of the main text. **Parameters:** For  $^{75}\text{As}$ :  $I = 3/2$ ,  $B_Q = 2\pi \times 1 \text{ MHz}$ ,  $g = 2\pi \times 7.22 \text{ MHz/T}$ ,  $\mathcal{A} = 2\pi \times 10.3 \text{ GHz}$ ; for  $^{69}\text{Ga}$ :  $I = 3/2$ ,  $B_Q = 2\pi \times (-0.5) \text{ MHz}$ ,  $g = 2\pi \times 10.22 \text{ MHz/T}$ ,  $\mathcal{A} = 2\pi \times 8.8 \text{ GHz}$ ; and for  $^{71}\text{Ga}$ :  $I = 3/2$ ,  $B_Q = 2\pi \times (-0.5) \text{ MHz}$ ,  $g = 2\pi \times 12.98 \text{ MHz/T}$ ,  $\mathcal{A} = 2\pi \times 11.5 \text{ GHz}$ . The total number of nuclei is taken as  $N = 5 \times 10^4$ . The As concentration is 0.5 and the concentrations of the two Ga isotopes are both taken as 0.25. The quadrupolar angle has been taken as  $\arctan(\sqrt{2})$ .

and  $\zeta = 2$  with dotted lines. The resonance frequency of the pulse sequence is indicated by a black line. At low magnetic field strengths ( $B \sim 1 \text{ T}$ ), the pulse sequence will pick up interactions with several nuclear transitions, but when the field is sufficiently strong to separate the peaks ( $B \gtrsim 3 \text{ T}$ ), a single nuclear transition can be chosen. This is confirmed in Fig. S2b, where the time evolution of the system under the pulse sequence is calculated and compared to the ideal evolution given in Eq. (2) of the main text. At  $B = 1 \text{ T}$ , the Ga nuclei and the  $^{75}\text{As}$   $\zeta = 1$  nuclear spin wave interfere with the targeted  $\zeta = 2$  spin wave in  $^{75}\text{As}$  and de-coheres the desired interaction, while at  $B = 3 \text{ T}$ , the electron polarisation is transferred ideally to the  $\zeta = 2$  spin wave of the  $^{75}\text{As}$  nuclei.

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