

Coherent control of a nuclear spin via interactions with a rare-earth ion in the solid-state

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Individually addressed Er^{3+} ions in solid-state hosts are promising resources for quantum repeaters, because of their direct emission in the telecom band and compatibility with silicon photonic devices. While the Er^{3+} electron spin provides a spin-photon interface, ancilla nuclear spins could enable multi-qubit registers with longer storage times. In this work, we demonstrate coherent coupling between the electron spin of a single Er^{3+} ion and a single $I = 1/2$ nuclear spin in the solid-state host crystal, which is a fortuitously located proton (^1H). We control the nuclear spin using dynamical decoupling sequences applied to the electron spin, implementing one- and two-qubit gate operations. Crucially, the nuclear spin coherence time exceeds the electron coherence time by several orders of magnitude, because of its smaller magnetic moment. These results provide a path towards combining long-lived nuclear spin quantum registers with telecom-wavelength emitters for long-distance quantum repeaters.

Optically interfaced solid-state atomic defects are promising for realizing quantum technologies, in particular quantum networks [1] and sensors [2]. In this approach, the optical transition of the defect provides a direct interface to the electronic spin [3]. However, enhanced functionality can be realized by coupling the electronic spin to one or more ancillary nuclear spins in the surrounding environment [4, 5]. For example, in NV centers in diamond, this approach has been used to realize long-lived nuclear spin quantum registers [6, 7], distribute entangled states across a quantum network [8], and implement multi-qubit operations such as quantum error correction protocols [9–12].

Individually addressed rare-earth ions (REIs) are an emerging platform [14–19] with several attractive features, including demonstrated single-shot spin readout [20, 21], and sub-wavelength addressing and control of many defects [22]. They can also be incorporated into a range of materials [23–25], which allows engineering of their environment to improve coherence and integration with devices [17, 18, 26]. Among REIs, Er^{3+} is particularly promising because its telecom-wavelength ($1.5\ \mu\text{m}$) optical transition may enable long-distance quantum repeaters without frequency conversion.

REIs are most commonly doped into yttrium-based host materials [27] [*e.g.*, Y_2SiO_5 (YSO), YVO_4 or $\text{Y}_3\text{Al}_5\text{O}_{12}$], which are magnetically noisy, harboring nu-

clear spins from Y (^{89}Y , with 100% abundance) and other elements (*e.g.*, ^{29}Si in YSO with 4.7% abundance). These form a bath that decoheres the electron spin. Recently, coupling between an REI electron spin (Ce^{3+}) and individual nuclear spins in the bath (^{89}Y , ^{29}Si) was observed in Ce:YSO [28]. Additionally, coherent coupling and quantum gate operations were demonstrated between a single $^{171}\text{Yb}^{3+}$ electron spin and collective states of a small ^{51}V nuclear spin ensemble in Yb:YVO_4 [29]. However, the operation of an ancilla nuclear spin qubit with a coherence time longer than the electron spin has not yet been demonstrated in REIs.

In this work, we demonstrate coherent coupling between the electron spin of a single Er^{3+} ion in YSO to a single nearby nuclear spin. The nuclear spin is controlled via microwave-driven, dynamical decoupling (DD) sequences on the electronic spin, realizing one-qubit and two-qubit gate operations that let us probe the coupling strength and coherent dynamics of the nuclear spin. We observe a nuclear spin T_2 time (with a Hahn echo) of 1.9 ms, about three orders of magnitude longer than the same quantity in the Er^{3+} electron spin alone. With a SWAP operation, we also demonstrate that the state of the nuclear spin in the computational basis survives electron spin readout and re-initialization. The properties of the nuclear spin suggest that it is a fortuitously located proton (^1H), whose location we determine by measuring the coupling strength for several magnetic field configurations. This work adds a key component to the toolkit of individually-addressed REIs, and also demonstrates a potentially powerful approach to engineering nuclear spin ancillae using non-native atomic species.

Our experimental platform consists of a silicon nanophotonic cavity (PC) bonded to a single crystal sample of YSO with a trace concentration of Er^{3+} ions (Fig. 1a). The cavity serves to enhance the emission rate

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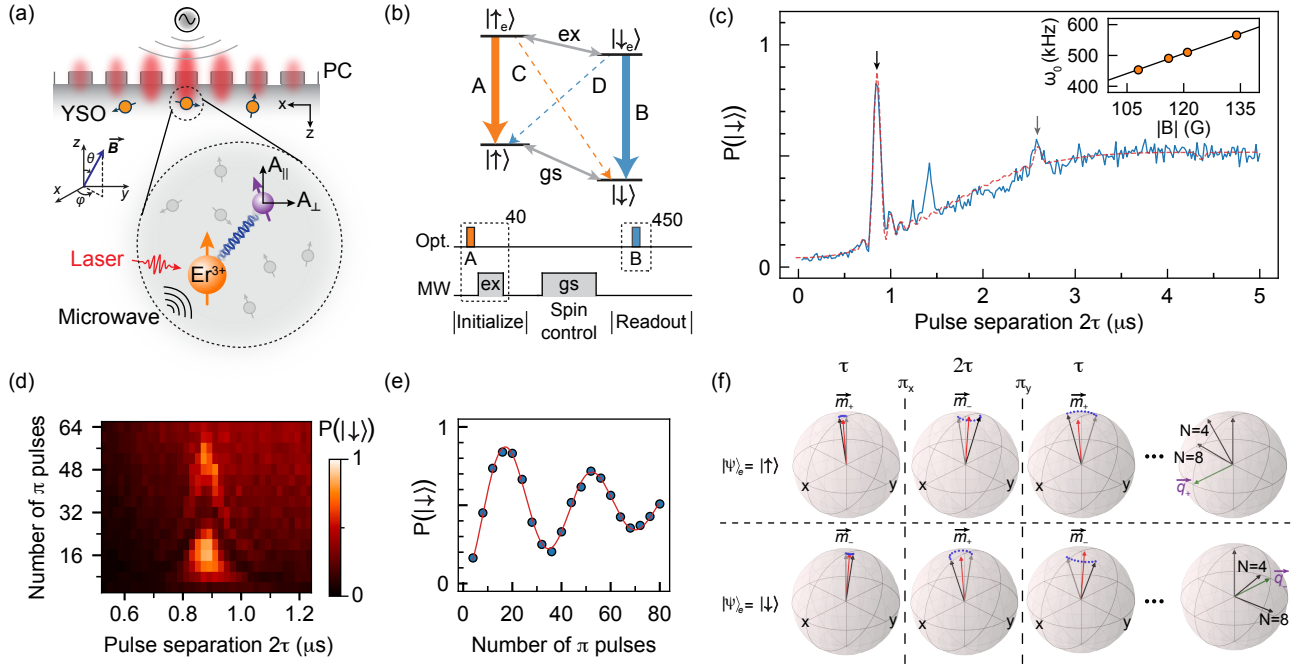


FIG. 1. **Detection and coherent control of a weakly-coupled nuclear spin using an Er^{3+} electronic spin.** (a) Measurement scheme. A single Er^{3+} spin, which is initialized, controlled, and readout via a combination of optical and microwave excitations, probes a nearby nuclear spin. (b) (top) Er^{3+} level structure; (bottom) Pulse scheme for initialization and single-shot readout of Er^{3+} spin state, interleaved with microwave pulses resonant with the ground state splitting for spin manipulation. Spin-conserving transitions (labeled A,B) are highly cyclic. (c) Evolution of an Er^{3+} spin under an XY-16 sequence as the separation between consecutive π -pulses (2τ) is varied (solid blue curve). Arrows indicate resonances at π/ω_0 and $3\pi/\omega_0$, based on the simulated signal (dashed red line) for the extracted interaction parameters and decay envelope with $T_{2,\text{XY16}} = 16.1 \pm 0.2 \mu\text{s}$. Inset: ω_0 as a function of field strength fits to a line with slope $4.3 \pm 0.1 \text{ kHz/G}$ and offset $-12 \pm 9 \text{ kHz}$. Experiments were performed at field angles $(\theta, \phi) = (90^\circ, 110^\circ)$ and $(90^\circ, 120^\circ)$ in a coordinate system defined by the YSO crystal structure: $(x, y, z) = (D_1, D_2, b)$ [13]. (d) The Er^{3+} spin population is plotted as a function of 2τ and number of π -pulses (N) in the XY- N sequence near the resonance centered at $2\tau_0 = 0.875 \mu\text{s}$. The “chevron” interference pattern originates from coherent interaction between the Er^{3+} spin and a nuclear spin (e) Coherent controlled rotation of the nuclear spin as a function of N at the 2τ resonance position. The data fits to a decaying sinusoidal (solid red line) with $\alpha = 10.2^\circ$ rotation per π -pulse. (f) Conditional evolution of a nuclear spin based on the Er^{3+} electronic spin state for one unit ($\tau - \pi_x - 2\tau - \pi_y - \tau$) of our decoupling sequence. Repeating this unit leads to a conditional x -rotation of the nuclear spin, around the effective axes $\mathbf{q}_\pm = \pm \hat{\mathbf{x}}$. All experiments were performed at the magnetic field configuration $(B, \theta, \phi) = (130 \text{ G}, 95^\circ, 110^\circ)$, corresponding to $(f_{\text{MW,gs}}, f_{\text{MW,ex}}) = (2.02, 1.32) \text{ GHz}$, unless indicated otherwise.

[17] and the cyclicity of the Er^{3+} optical transitions, enabling single-shot spin readout [20]. Additionally, microwave fields from a nearby antenna are used to drive spin rotations with a π pulse duration of approximately 31 ns. Additional details about the experimental geometry can be found in Ref. [30].

In a magnetic field, the spin-1/2 ground and excited states of the Er^{3+} ion become non-degenerate, giving rise to four distinct optical transitions (A-D) that can be used to control and measure the electron spin (Fig. 1b). We initialize the spin by alternately exciting the ion on the A transition and applying microwave π -pulses to the excited state, which polarizes the spin into $|\downarrow\rangle$ [22]. Spin readout is accomplished by repeatedly exciting the B transition, with a typical single-shot fidelity of 81%.

We probe the local magnetic environment of the Er^{3+} ion by performing microwave-driven DD sequences $[\text{XY-}N := (\tau - \pi_{x/y} - \tau)^N]$ on the Er^{3+} spin with vary-

ing inter-pulse separation 2τ [31–33]. On top of an overall coherence decay arising from the nuclear spin bath ($T_{2,\text{XY16}} = 16.1 \pm 0.2 \mu\text{s}$), the particular Er^{3+} ion studied in this work exhibits a sharp resonance at $2\tau_0 = 0.875 \mu\text{s}$ (Fig. 1c). At the resonance, varying the number of π pulses N in the XY- N sequence yields coherent oscillations of the Er^{3+} spin population (Fig. 1d,e). We attribute these features to a coherent interaction between the Er^{3+} electronic spin and a nearby nuclear spin.

To understand this phenomenon, we consider the interaction Hamiltonian between the electronic and nuclear spins in the rotating frame of the electronic spin under the secular approximation:

$$H/\hbar = 2S_z(A_{\parallel}I_z + A_{\perp}I_x) + \omega_L I_z. \quad (1)$$

Here, $\mathbf{S}$ is the pseudo spin-1/2 operator for the lowest Kramers’ doublet electronic state, $\mathbf{I}$ is nuclear spin oper-

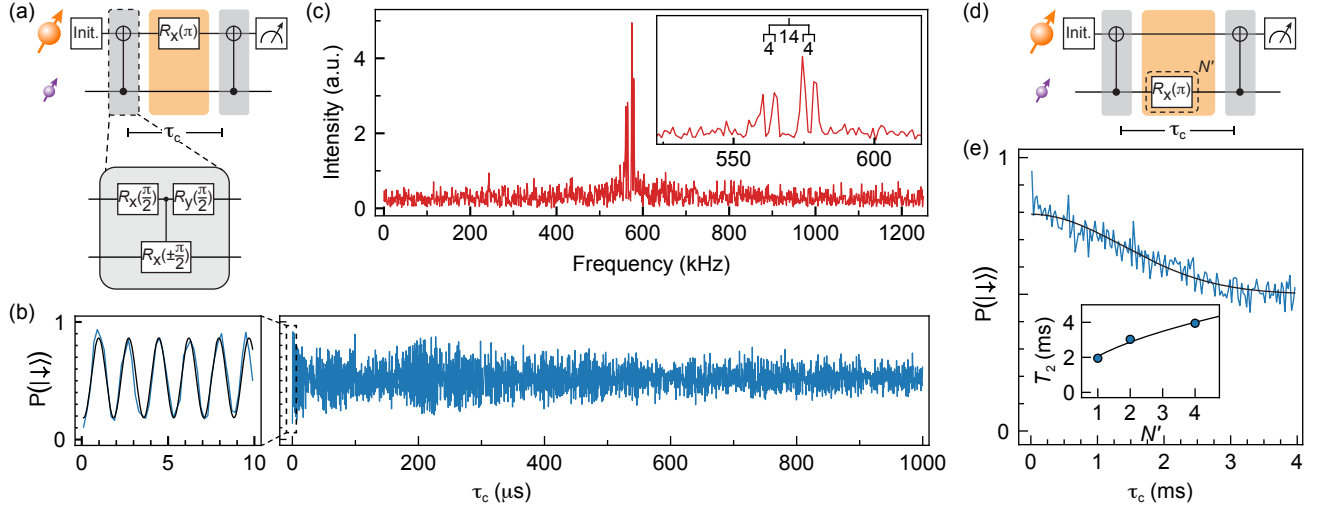


FIG. 2. **Probing coherence of the nuclear spin.** (a) Ramsey spectroscopy of the nuclear spin is performed by changing the delay τ_c between two C_nNOT_e operations and applying a π pulse on the electron at the halfway-point. (b) The spectroscopy reveals oscillations at ω_0 in the short time scale (solid black line) and a beating envelope over a millisecond. (c) FFT of the Ramsey signal reveals four distinct frequency components that are symmetric around ω_0 . (d) Hahn and CPMG sequences on the nuclear spin are performed by applying unconditional $R_x(\pi)$ operations. (e) The sequence yields $T_{2,\text{Hahn}} = 1.9 \pm 0.1$ ms and can be extended to $T_{2,\text{CPMG}} = 3.9 \pm 0.2$ ms (inset).

ator, $A_{\parallel}$ and $A_{\perp}$ are parallel and perpendicular hyperfine coupling parameters, and $\omega_L = \gamma_N B / \hbar$ is the (bare) Larmor frequency of the nuclear spin. In the spin Hamiltonian, we define the $\hat{z}$ -axis to be the direction of the external magnetic field, while the $\hat{x}$ axis is fixed by $A_{\perp}$. The orientation of the field in the reference frame of the YSO crystal axes is specified in Fig. 1. Rearranging terms in Eq. (1), the Hamiltonian simplifies to:

$$H/\hbar = |\uparrow\rangle\langle\uparrow| \otimes \omega_+ \mathbf{I} \cdot \mathbf{m}_+ + |\downarrow\rangle\langle\downarrow| \otimes \omega_- \mathbf{I} \cdot \mathbf{m}_-, \quad (2)$$

where $\omega_{\pm} = \sqrt{(\omega_L \pm A_{\parallel})^2 + A_{\perp}^2}$ is the effective Larmor frequency and $\mathbf{m}_{\pm} = (\pm A_{\perp}, 0, \pm A_{\parallel} + \omega_L) / \omega_{\pm}$ is the precession axis of the nuclear spin. From Eq. (2), it is evident that the nuclear spin rotation is conditional on the Er^{3+} electronic spin state: it precesses around the $\mathbf{m}_+$ ($\mathbf{m}_-$) axis at the frequency ω_+ (ω_-) when the electron is in the state $|\uparrow\rangle$ ($|\downarrow\rangle$).

In the strong magnetic field regime, $\omega_L \gg \sqrt{A_{\parallel}^2 + A_{\perp}^2}$, $\mathbf{m}_{\pm}$ are both nearly parallel to the magnetic field axis, $\hat{z}$. However, the small perpendicular component can be used to generate a conditional rotation of the nuclear spin. Specifically, toggling the electron spin using an XY- N sequence satisfying the resonance condition $2\tau_0 = \pi/\omega_0$ [$\omega_0 = (\omega_+ + \omega_-)/2 \sim \omega_L$] leads to a rotation of the nuclear spin around the effective precession axes $\mathbf{q}_{\pm}$, conditional on the initial electronic state (Fig. 1f). After N periods, the evolution of the nuclear spin is described by:

$$U_{\text{XY-}N} = |\uparrow\rangle\langle\uparrow| \otimes R_x(N\alpha) + |\downarrow\rangle\langle\downarrow| \otimes R_x(-N\alpha), \quad (3)$$

where $\alpha \sim 2A_{\perp}/\omega_L$ is the rotation angle per π pulse (Fig. 1e). In Fig. 1c (inset), we plot the observed resonance frequency of the XY-16 sequence as a function of

the magnetic field strength. It exhibits a slope of 4.3 ± 0.1 kHz/G, which is consistent with the gyromagnetic ratio of a proton (4.258 kHz/G) within the experimental uncertainty. Using measurement techniques described in the Supplemental Material [34], we can extract the full set of parameters $(|A_{\parallel}|, A_{\perp}, \omega_L) = (19.4, 50.5, 567.4)$ kHz for the magnetic field orientation used in Fig. 1c-e. With these parameters, $\alpha = 10.2^\circ$, such that XY-8 approximately implements a maximally entangling conditional- $R_x(\pm\pi/2)$ rotation on the nuclear spin.

We now study the coherence properties of the nuclear spin. Using additional rotations on the electronic spin, the conditional- $R_x(\pm\pi/2)$ operation is adapted to a C_nNOT_e operation, where the nuclear spin acts as a control (Fig. 2a). The operation is controlled by the nuclear spin state along the $\hat{x}$ -axis, which is roughly perpendicular to the precession axes $\mathbf{m}_{\pm}$, which are nearly parallel to the $\hat{z}$ -axis. Therefore, the sequence in Fig. 2a measures the precession of the nuclear spin states $|\pm x\rangle = (|\uparrow\rangle \pm |\downarrow\rangle)/\sqrt{2}$, equivalent to a standard Ramsey measurement of the nuclear spin coherence. The additional π -pulse on the electronic spin decouples the precession frequency from the (random) initial state of the nuclear spin, so that the average precession frequency ω_0 is observed at short times (Fig. 2b). However, at longer times, the precession shows a complex beating at several frequencies. A fast Fourier transform (FFT) of the Ramsey measurement reveals four symmetrically spaced peaks, where two subgroups of peaks are separated by 14 kHz and each subgroup is split by 4 kHz (Fig. 2c). This suggests the presence of additional spins in the nuclear spin environment, which we discuss below.

We measure the nuclear spin T_2 with a Hahn echo se-

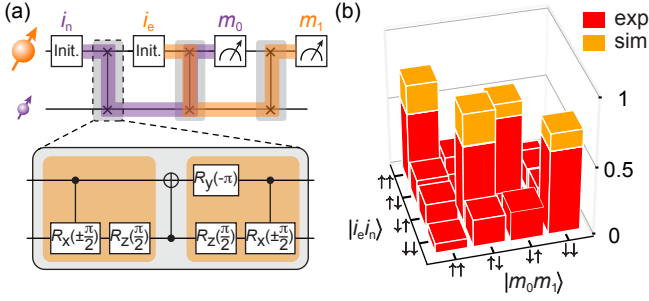


FIG. 3. **SWAP operation.** (a) Demonstration of SWAP operations between the electronic and nuclear spin using the indicated circuit, where the initialization $|i_e\rangle, |i_n\rangle \in \{|\uparrow\rangle, |\downarrow\rangle\}$ correspond to nuclear spin and electron spin initialization due to the action of SWAP. This is followed by readout of $|m_0\rangle$ and $|m_1\rangle$ also interleaved with SWAP. (b) Measurements $|m_0 m_1\rangle$ are plotted for each initialization $|i_e i_n\rangle$. The SWAP pattern is observed as $|i_n\rangle$ ($|i_e\rangle$) determines $|m_0\rangle$ ($|m_1\rangle$), after two SWAP operations. The corresponding SWAP fidelity in the computational basis is 87% (91%) for the experiment (simulation), where the simulation takes into account known error sources [34].

quence, by inserting a π -pulse on the nuclear spin during its precession (Fig. 2d). We perform the π pulse by applying the conditional- $R_x(\pm\pi/2)$ twice so that the nuclear spin rotates by π around the $\hat{x}$ -axis regardless of the electronic spin state. This cancels the discrete frequency shifts observed in the Ramsey experiment and yields a T_2 time of 1.9 ms, which can be extended to 3.9 ms with repeated applications of the nuclear spin π -pulse in a Carr-Purcell-Meiboom-Gill (CPMG) sequence (Fig. 2e). Crucially, this value is three orders of magnitude longer than the bare electronic spin coherence ($T_{2,e} = 2\mu\text{s}$ [22]), demonstrating that the smaller nuclear magnetic moment leads to extended coherence even in the magnetically noisy environment of YSO.

Next, we implement a SWAP operation to store the electronic state in the nuclear spin. The SWAP consists of conditional- $R_x(\pm\pi/2)$ rotations interleaved with single qubit rotations of the electronic spin and $\hat{z}$ -rotations of the nuclear spin, realized via its free precession for duration τ_0 (Fig. 3a) [34]. We apply the SWAP operation within a sequence that repeatedly initializes and measures the electronic spin in the $\hat{z}$ -basis. The first initialization $|i_n\rangle$ is swapped to the nuclear spin. After completing the second initialization $|i_e\rangle$, we obtain the two qubit state $|i_e i_n\rangle \in \{|\uparrow\uparrow\rangle, |\uparrow\downarrow\rangle, |\downarrow\uparrow\rangle, |\downarrow\downarrow\rangle\}$. Performing another SWAP operation before the first readout measures the state of the nuclear spin $|m_0\rangle$, followed by the state of the electronic spin $|m_1\rangle$. The populations measured for each two qubit readout outcome $|m_0 m_1\rangle$, for a given initialization $|i_e i_n\rangle$, yield a SWAP operation fidelity in the computational basis of 87% (Fig. 3b). This fidelity is consistent with a theoretical estimate incorporating the variation of the nuclear spin precession frequency (Fig. 2c), optical excitations of the Er^{3+} ion and a finite lifetime for

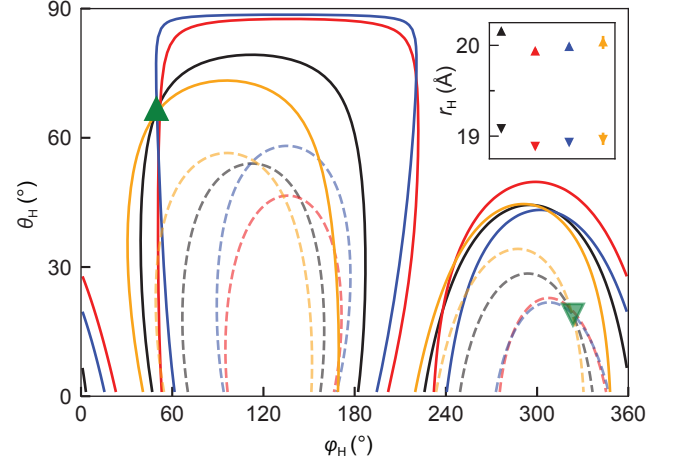


FIG. 4. **Locating the nuclear spin.** $A_{||}/A_{\perp}$ contour plots, where each contour line (black, red, blue, orange) corresponds to nuclear spin positions (θ_H, ϕ_H) that satisfy measurements at magnetic field orientations $(\theta, \phi) = (95, 110), (95, 140), (85, 140), (100, 90)$ respectively. The intersection of the contour lines, marked by a green triangle, represents a solution. Asserting $A_{||} > 0$ ($A_{||} < 0$), indicated by solid (dashed) contours, leads to a solution at $r_H, \theta_H, \phi_H = 20.0 \text{ \AA}, 66.7^\circ, 49.6^\circ$ ($r_H, \theta_H, \phi_H = 19.0 \text{ \AA}, 19.0^\circ, 323.6^\circ$). Inset: Calculated distances given $A_{||} > 0$ ($A_{||} < 0$) marked by upward (downward) triangles for each orientation at the indicated solution along with error bars obtained from a Monte-Carlo simulation of measurement uncertainty.

the nuclear spin [34]. This estimate lets us place a lower bound on the intrinsic nuclear spin lifetime of $T_1 > 0.6$ s. However, the changes in the magnetic moment of the electron spin due to excitations during readout generate significant dephasing of the nuclear spin, which precludes storage of arbitrary nuclear spin states during readout in the current configuration.

Finally, we probe the location and gyromagnetic ratio of the nuclear spin in the lattice by measuring the hyperfine parameters ($A_{||}, A_{\perp}, \omega_L$) at four angles of the external magnetic field. The average value of ω_L yields a gyromagnetic ratio $\gamma_N/\hbar = 4.26 \pm 0.04 \text{ kHz/G}$, which is consistent with a ^1H nuclear spin (4.258 kHz/G). The extracted hyperfine parameters at each field orientation constrain the nuclear spin position to lie on a 1D manifold. The intersection of these curves (Fig. 4) yields two possible positions of the nuclear spins, which could be further disambiguated by determining the sign of $A_{||}$, which we do not do in this work. For both possible locations, the distance between the electron and nuclear spin is around 1.9 – 2 nm. We are not able to confidently assign a position in the YSO crystal structure to the nuclear spin, because of uncertainties in the relative alignment of the magnetic axes of the Er^{3+} site with the crystallographic axes [13], which are not aligned because of the low C_1 symmetry of the Er^{3+} site in YSO.

We now turn to a discussion of several aspects of these

results. While hydrogen is not part of the YSO chemical formula, it is a ubiquitous impurity that is present during the growth and post-processing and able to unintentionally incorporate in to materials [35–37]. The Er^{3+} center studied in this work is one of six ions that we have probed in the same sample, and the only one to show a strong hydrogen nuclear spin coupling, which is consistent with a hydrogen concentration in the range of $0.3 - 3.9 \times 10^{18} \text{ cm}^{-3}$ [34], a plausible value [38]. Given that hydrogen is quite mobile in oxides, H atoms are also expected to diffuse easily and form defect complexes with large binding energies [39]. It is also noteworthy that the nuclear spin precession spectrum (Fig. 2a) shows four discrete frequency components. In the Supplemental Material, we extend our Ramsey measurements to show that the transitions between these frequencies occur as sudden jumps that can be observed with single-shot measurements, with a correlation time of seconds to minutes [34].

We rule out global magnetic field fluctuations and interactions with other nearby electronic spins because these would have a much larger impact on the Er^{3+} electron, which we do not observe. However, the jumps could be explained by coupling between the H and additional $I = 1/2$ nuclei, with Ising interaction strengths of 2 and 7 kHz. In contrast to the distinctively large gyromagnetic ratio of the hydrogen nuclear spin, nuclear spins with smaller gyromagnetic ratios may not be directly observed via the electronic spin within its finite coherence time.

Using comprehensive density functional theory calculations, we conclude that these coupling strengths cannot be explained by the native nuclei alone (^{89}Y and ^{29}Si). However, glow discharge mass spectroscopy (GDMS) analysis of a related YSO sample reveals the presence of several chemical impurities with $I = 1/2$ and ppm concentrations, including Cd and P. Typically, both interstitial hydrogen (H_i) and substitutional hydrogen (H_O) tend to act as donors in oxides, and are hence attracted to CdY and P_O , which act as acceptors, which may favor the formation of complexes of these impurities despite their low concentration. This hypothesis is further supported by comprehensive density functional theory calculations: a CdY-H_O complex and a $\text{P}_\text{O-H}_i$ complex, together with a nearby ^{29}Si , yield coupling strengths to H that are similar to the experimental values (structures and computational details are included in [34]). Although the observation of such a nuclear spin defect complex is not conclusive at this point, the possibility of engineering nuclear spin registers using this approach is potentially attractive and could be investigated further.

Lastly, we consider the prospects for several extensions to this work. First, driving both the electron and nuclear spins directly would allow the extension of these techniques to multiple nuclear spins [40, 41]. Second, to use the nuclear spin as an ancilla in a quantum network, it is necessary to preserve an arbitrary quantum state during a measurement of the electronic spin [42]. This can be achieved by minimizing the frequency shift of the nuclear spin when the electron is excited to the excited state, either through alignment of the magnetic field, using more weakly coupled nuclear spins, or encoding information in a decoherence-protected subspace of a pair of nuclear spins [43].

In conclusion, we demonstrate coherent interaction between a single Er^{3+} electronic spin and a nuclear spin. Using single and two-qubit operations based on DD sequences, we measure the coherence of this nuclear spin to be several orders of magnitude longer than that of the electronic spin. We further show that the nuclear spin state survives the initialization and readout of the electron in the computational basis by implementing a SWAP operation, and finally, determine its location with respect to the Er^{3+} ion. Combining the current work with our previous work on parallel measurement and control of multiple Er^{3+} ions coupled to the same nanophotonic cavity [22], we can ultimately envision a hybrid quantum register that is comprised of many telecom-compatible optically-interfaced solid-state spins with single particle control at each node, where each spin is individually coupled to long-lived nuclear spin registers.

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Supplementary Information for “Coherent control of a nuclear spin via interactions with a rare-earth ion in the solid-state”

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1 Device parameters

The silicon nanophotonic cavity used in this work has a measured quality factor $Q = 4.4 \times 10^4$ and a one-way coupling efficiency $\eta_{\text{cav}} = \kappa_{\text{wg}}/(\kappa_{\text{wg}} + \kappa_{\text{int}}) = 0.19$, where κ_{wg} and κ_{int} are waveguide and internal loss channels from the cavity, respectively. The particular Er^{3+} ion studied in this work has an optical excited state lifetime of around $60 \mu\text{s}$ (corresponding to a Purcell factor of about 190). Further details about the experimental setup and device fabrication can be found in Refs. [1, 2].

2 Electron-nucleus hyperfine interaction

2.1 Theoretical background

As stated in the main text, the two-body system of an electron spin and a nuclear spin is described by the secular hyperfine interaction Hamiltonian in the rotating frame of the electronic spin, $H = 2S_z(A_{\parallel} + A_{\perp}) + \omega_L I_z$ (where we set $\hbar = 1$ in the supplementary text). We can also express the interaction Hamiltonian in terms of nuclear spin Hamiltonians $H_{\pm}$, which are conditioned on the electronic spin state as:

$$\begin{aligned} H &= |\uparrow\rangle \langle\uparrow| \otimes H_+ + |\downarrow\rangle \langle\downarrow| \otimes H_- \\ H_{\pm} &= \omega_{\pm} \mathbf{I} \cdot \mathbf{m}_{\pm} = (\pm A_{\parallel} + \omega_L) I_z \pm A_{\perp} I_x, \end{aligned} \quad (1)$$

where $\mathbf{m}_{\pm} = (\pm A_{\perp}, 0, \pm A_{\parallel} + \omega_L)/\omega_{\pm}$ and $\omega_{\pm} = \sqrt{(\omega_L \pm A_{\parallel})^2 + A_{\perp}^2}$. A dynamical decoupling (DD) sequence can then be described by periods of free evolution under this Hamiltonian (Eq. 1) interleaved with periodically spaced π -pulses. A free evolution propagator for duration τ is given by $U = e^{-iH\tau} = \exp(-i|\uparrow\rangle \langle\uparrow| \otimes H_+ \tau) \cdot \exp(-i|\downarrow\rangle \langle\downarrow| \otimes H_- \tau)$. Expanding each exponential yields an intuitive simplification, expressing the nuclear spin evolution in terms of the Hamiltonians $H_{\pm}$ conditional on the electronic spin state:

$$\begin{aligned} U &= |\uparrow\rangle \langle\uparrow| \otimes e^{-iH_+\tau} + |\downarrow\rangle \langle\downarrow| \otimes e^{-iH_-\tau} = |\uparrow\rangle \langle\uparrow| \otimes U_+ + |\downarrow\rangle \langle\downarrow| \otimes U_- \\ &= |\uparrow\rangle \langle\uparrow| \otimes e^{-i\phi_+ \mathbf{m}_+ \cdot \mathbf{I}} + |\downarrow\rangle \langle\downarrow| \otimes e^{-i\phi_- \mathbf{m}_- \cdot \mathbf{I}} \end{aligned} \quad (2)$$

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As noted above, it is convenient to write the conditional free evolution of the nuclear spin, $U_{\pm} = e^{-iH_{\pm}\tau}$, as a rotation around the axes $\mathbf{m}_{\pm}$ by an angle $\phi_{\pm} = \omega_{\pm}\tau$ and express the DD sequence in terms of nuclear spin rotations. Evolution under the basic building block ($\tau - \pi - \tau$) of the DD sequence can be obtained from the free evolution propagator and the π -pulse $X \otimes I$, which acts only on the electronic spin, as:

$$\begin{aligned} V = U(X \otimes I)U &= |\uparrow\rangle \langle\downarrow| \otimes U_+U_- + |\downarrow\rangle \langle\uparrow| \otimes U_-U_+ = |\uparrow\rangle \langle\downarrow| \otimes V_+ + |\downarrow\rangle \langle\uparrow| \otimes V_- \\ &= |\uparrow\rangle \langle\downarrow| \otimes e^{-i\phi_{\mathbf{n}+} \cdot \mathbf{I}} + |\downarrow\rangle \langle\uparrow| \otimes e^{-i\phi_{\mathbf{n}-} \cdot \mathbf{I}}. \end{aligned} \quad (3)$$

Similar to the free propagation operators, $U_{\pm}$, we can express the product of rotations $V_{\pm} = U_{\pm}U_{\mp}$ of the nuclear spin as a rotation around an axis $\mathbf{n}_{\pm}$ by an angle ϕ . However, it is more convenient to work with a repetition of this block, $(\tau - \pi - 2\tau - \pi - \tau)$, which is composed of two π -pulses and diagonalized in the electronic spin basis:

$$\begin{aligned} W = V^2 &= |\uparrow\rangle \langle\uparrow| \otimes V_+V_- + |\downarrow\rangle \langle\downarrow| \otimes V_-V_+ = |\uparrow\rangle \langle\uparrow| \otimes W_+ + |\downarrow\rangle \langle\downarrow| \otimes W_- \\ &= |\uparrow\rangle \langle\uparrow| \otimes e^{-i(2\alpha)\mathbf{q}_+ \cdot \mathbf{I}} + |\downarrow\rangle \langle\downarrow| \otimes e^{-i(2\alpha)\mathbf{q}_- \cdot \mathbf{I}} \end{aligned} \quad (4)$$

Continuing the pattern of effective rotation, we express $W_{\pm} = V_{\pm}V_{\mp}$ as a rotation around the axes $\mathbf{q}_{\pm}$ by an angle 2α . This form has the advantage that any DD sequence $(\tau - \pi - 2\tau - \pi - \tau)^{N/2}$ with an even number of π -pulses, N , can be easily expressed as follows:

$$V^N = |\uparrow\rangle \langle\uparrow| \otimes e^{-i(N\alpha)\mathbf{q}_+ \cdot \mathbf{I}} + |\downarrow\rangle \langle\downarrow| \otimes e^{-i(N\alpha)\mathbf{q}_- \cdot \mathbf{I}}. \quad (5)$$

The aforementioned effective rotation axes and angles can be calculated using the following identity, $e^{-ia_1(\mathbf{p}_1 \cdot \boldsymbol{\sigma})}e^{-ia_2(\mathbf{p}_2 \cdot \boldsymbol{\sigma})} = e^{-ia(\mathbf{p} \cdot \boldsymbol{\sigma})}$ [$\boldsymbol{\sigma} = (\sigma_x, \sigma_y, \sigma_z)$ is the Pauli vector], where $\mathbf{p}$ and a is expressed as:

$$\begin{aligned} \cos a &= \cos a_1 \cos a_2 - \sin a_1 \sin a_2 (\mathbf{p}_1 \cdot \mathbf{p}_2) \\ \mathbf{p} \sin a &= \sin a_1 \sin a_2 (\mathbf{p}_1 \times \mathbf{p}_2) + \sin a_1 \cos a_2 \mathbf{p}_1 + \sin a_2 \cos a_1 \mathbf{p}_2. \end{aligned} \quad (6)$$

In order to obtain a maximally entangling operation, the effective rotation axes $\mathbf{q}_{\pm}$ must be antiparallel, such that $\mathbf{q}_+ \cdot \mathbf{q}_- = -1$. Based on Eq. 6, it can be shown that this condition implies $\cos \frac{\phi_+}{2} \cos \frac{\phi_-}{2} - \sin \frac{\phi_+}{2} \sin \frac{\phi_-}{2} \cos \gamma = 0$, where $\cos \gamma = \mathbf{m}_+ \cdot \mathbf{m}_-$ is the angle between the free-precession axes $\mathbf{m}_{\pm}$. When the equality holds, we obtain the following precession axes and angles:

$$\begin{aligned} \cos \alpha &= 1 - 2(\sin \frac{\phi_+}{2} \sin \frac{\phi_-}{2} \sin \gamma)^2 \\ \mathbf{q}_{\pm} \sin \alpha &= \pm 2(\sin \frac{\phi_+}{2} \sin \frac{\phi_-}{2} \sin \gamma) \hat{\mathbf{y}} \times (\sin \frac{\phi_+}{2} \cos \frac{\phi_-}{2} \mathbf{m}_+ + \sin \frac{\phi_-}{2} \cos \frac{\phi_+}{2} \mathbf{m}_-) \end{aligned} \quad (7)$$

In the strong magnetic field regime, $(\omega_L^2 \gg A_{\parallel}^2 + A_{\perp}^2)$, the free precession axes $\mathbf{m}_{\pm}$ are almost parallel to each other such that $\cos \gamma \sim 1$. This leads to a simpler resonant condition $\cos \frac{\phi_+ + \phi_-}{2} = 0$ or $2\tau = (\pi + m2\pi)/\omega_0$, where $\omega_0 = (\omega_+ + \omega_-)/2$. With further simplifications, we obtain the following:

$$\begin{aligned} \alpha &\simeq \gamma = \frac{2A_{\perp}\omega_L}{\omega_+\omega_-} \sim \frac{2A_{\perp}}{\omega_L} \\ \mathbf{q}_{\pm} &\simeq \pm(\hat{\mathbf{x}} + \frac{A_{\perp}A_{\parallel}}{\omega_L^2}\hat{\mathbf{z}}) \end{aligned} \quad (8)$$

For a geometrical description of the operation, it is worth noting that the amount of rotation per π -pulse, α , is simply equal to the angle γ between the free precession axes $\mathbf{m}_{\pm}$ in the strong magnetic field regime under the resonant conditions. The deviation of the effective rotation axes $\mathbf{q}_{\pm}$ from the $\hat{\mathbf{x}}$ -direction is on the order of $A_{\parallel}A_{\perp}/\omega_L^2 \sim 10^{-3}$ for our parameter regime, so we simply label the effective axes $\mathbf{q}_{\pm} = \hat{\mathbf{x}}_{\pm}$. With this understanding in place, we express the final form of a DD sequence with N pulses as $(R_x(\theta) = e^{-i\theta\sigma_x/2})$:

$$V^N = |\uparrow\rangle \langle\uparrow| \otimes R_x(N\alpha) + |\downarrow\rangle \langle\downarrow| \otimes R_x(-N\alpha) \quad (9)$$

2.2 Measuring hyperfine interaction parameters

We can use the above description of effective precession axes to obtain the conditional free precession frequencies ω_+ and ω_- of the nuclear spin. When combined with the knowledge of α , these three parameters are sufficient to solve for the Hamiltonian parameters ($A_{||}$, $A_{\perp}$, ω_L).

However, rather than measuring $\omega_{\pm}$ directly, we measure $\omega_0 = (\omega_+ + \omega_-)/2$ and $\omega_{\delta} = (\omega_+ - \omega_-)/2$. For the former, we perform the experiment described in Fig. 2a of the main text, where a free-precession of the nuclear spin occurs in between two C_nNOT_e operations, subject to one extra π -pulse on the electron spin during the free evolution, such that the effective precession frequency of the nuclear spin is an average of ω_+ and ω_- . To see this analytically, we first consider the free evolution during the experiment in Fig. 2a as:

$$U_{\pi_e}(\tau_c) = U(\tau_c/2)(X \otimes I)U(\tau_c/2) \quad (10)$$

Noting that this is equivalent to the unitary operator V for $\tau = \tau_c/2$ in Eq. 3, we can express it as an effective precession conditional on the electron spin state:

$$U_{\pi_e}(\tau_c) = |\uparrow\rangle\langle\downarrow| \otimes e^{-i\phi_{\mathbf{n}+}\cdot\mathbf{I}} + |\downarrow\rangle\langle\uparrow| \otimes e^{-i\phi_{\mathbf{n}-}\cdot\mathbf{I}}; \quad e^{-i\phi_{\mathbf{n}\pm}\cdot\mathbf{I}} = e^{-i\phi_{\pm}\mathbf{m}_{\pm}\cdot\mathbf{I}}e^{-i\phi_{\mp}\mathbf{m}_{\mp}\cdot\mathbf{I}}, \quad (11)$$

where the rotation around each free-precession axes, $\mathbf{m}_{\pm} = (\pm A_{\perp}, 0, \omega_L \pm A_{||})/\omega_{\pm}$, is given by $\phi_{\pm} = \omega_{\pm}\tau_c/2$. By defining $U_0 = C_nNOT_e U_{\pi_e}(\tau_c) C_nNOT_e$ and considering its action on the two-qubit initial state, $\rho_i = |\downarrow\rangle\langle\downarrow| \otimes I/2$, we can compute the signal, $s_0(\tau_c)$, for population measurement on the electron spin to first order in $A_{||}$:

$$s_0(\tau_c) = \text{Tr}[U_0 \rho_i U_0^{\dagger} P_{\downarrow}] \simeq 1 - \cos \frac{\phi^2}{2} - \left(\frac{A_{\perp}}{\omega_+ \omega_-} (\omega_0 \sin \frac{\phi_+ - \phi_-}{2} + 2A_{||} \sin \frac{\phi_+ + \phi_-}{2}) \right)^2 \quad (12)$$

In the strong magnetic field regime, where the free-precession axes $\mathbf{m}_{\pm}$ are approximately parallel such that $\cos \gamma \sim 1$, Eq. 6 leads to $\cos \frac{\phi}{2} = \cos \frac{\phi_+}{2} \cos \frac{\phi_-}{2} - \sin \frac{\phi_+}{2} \sin \frac{\phi_-}{2} = \cos(\phi_+ + \phi_-)/2$. This implies that, in this regime, the net rotation will simply be $\phi = \phi_+ + \phi_- = \omega_0 \tau_c$. Although the second term in Eq. 12 also includes frequencies $\omega_{\pm}/2$ and ω_{δ} , their magnitude is lower by $(A_{\perp}/\omega_L)^2 \sim 10^{-2}$. Neglecting this second term, the signal (Eq. 12) reduces to $s_0(\tau_c) \simeq 1 - \frac{1}{2} \cos \omega_0 \tau_c$.

In order to obtain ω_{δ} , we insert an additional π -pulse on the nuclear spin by using XY-16 operation as described in the main text:

$$U_{\pi_e, \pi_n}(\tau_c) = U(\tau_c/2)(X \otimes X)U(\tau_c/2) \quad (13)$$

In a similar fashion to Eq. 11, we can express this operation as a consecutive rotation of the nuclear spin around two axes, one of which is inverted due to the additional π -pulse on the nuclear spin:

$$U_{\pi_e, \pi_n}(\tau_c) = |\uparrow\rangle\langle\downarrow| \otimes e^{-i\phi'_{\mathbf{n}'}\cdot\mathbf{I}}X + |\downarrow\rangle\langle\uparrow| \otimes e^{-i\phi'_{\mathbf{n}'}\cdot\mathbf{I}}X; \quad e^{-i\phi'_{\mathbf{n}'\pm}\cdot\mathbf{I}} = e^{-i\phi_{\pm}\mathbf{m}_{\pm}\cdot\mathbf{I}}e^{-i\phi_{\mp}\mathbf{m}'_{\mp}\cdot\mathbf{I}}, \quad (14)$$

where $\mathbf{m}'_{\pm} = (\pm A_{\perp}, 0, -(\omega_L \pm A_{||})/\omega_{\pm})$ are the inverted axes for the nuclear spin. Defining $U_{\delta} = C_nNOT_e \cdot U_{\pi_e, \pi_n}(\tau_c) \cdot C_nNOT_e$ acting on the same initial state, we also compute the expected signal, $s_{\delta}(\tau_c)$:

$$s_{\delta}(\tau_c) = \text{Tr}[U_{\delta} \rho_i U_{\delta}^{\dagger} P_{\downarrow}] \simeq \cos \frac{\phi'^2}{2} + \left(\frac{A_{\perp}}{\omega_+ \omega_-} (\omega_0 \sin \frac{\phi_+ - \phi_-}{2} + 2A_{||} \sin \frac{\phi_+ + \phi_-}{2}) \right)^2 \quad (15)$$

Due to the additional π -pulse on the nuclear spin, the two precession axes are approximately anti-parallel such that $\mathbf{m}_{\pm} \cdot \mathbf{m}'_{\mp} = \cos \gamma' \sim -1$. Again based on Eq. 6, this yields $\cos \phi'/2 = \cos \frac{\phi_+}{2} \cos \frac{\phi_-}{2} + \sin \frac{\phi_+}{2} \sin \frac{\phi_-}{2} = \cos(\phi_+ - \phi_-)/2$, which implies that $\phi' = \phi_+ - \phi_- = \omega_{\delta} \tau_c$. Neglecting the second term, Eq. 15 reduces to $s_{\delta}(\tau_c) \simeq 1 + \frac{1}{2} \cos \omega_{\delta} \tau_c$.

Measuring α (Eq. 8), in addition to ω_0 and ω_{δ} , is sufficient to solve for the parameters ($A_{||}$, $A_{\perp}$, ω_L). Although the above analysis assumes perfect C_nNOT_e and $R_x(\pi)$ operations on the nuclear spin, we use the exact unitary operators obtained from the Hyperfine parameters for XY-8 and XY-16 sequences in simulation (Fig. S1). However, the decoherence mechanisms to be discussed for the SWAP operation are not included in these simulations. Finally, note that the signal $s_{\delta}(\tau_c)$ is agnostic to the sign of ω_{δ} , so that the sign of $A_{||}$ is undetermined.

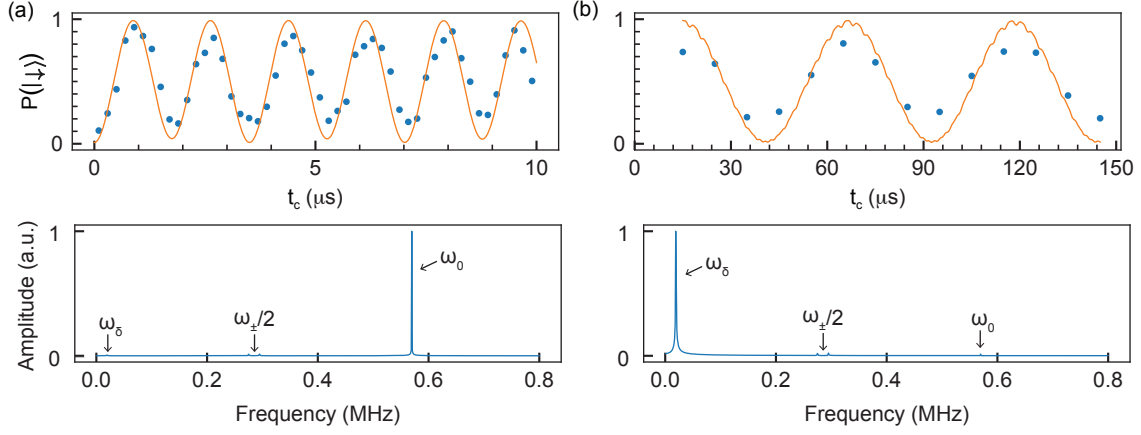


FIG. S1: **Observing ω_0 and ω_δ .** (a) Simulated $s_0(\tau_c)$ signal overlaid with experimental data and the FFT of the simulation, displaying the peak at ω_0 (b) Similarly plotting $s_\delta(\tau_c)$ and its FFT with a peak at ω_δ . The earliest data point in the top panel starts around 15 μs due to the finite length of the unconditional- $R_x(\pi)$ on the nuclear spin.

3 SWAP operation and nuclear spin memory

3.1 Description of SWAP

We can investigate error sources for the SWAP operation by looking into its constituents. From here on, we use the convention that $|\uparrow\rangle = |0\rangle$ and $|\downarrow\rangle = |1\rangle$ for both the electronic and nuclear spin bases. As noted in Fig. 3 of the main text, a SWAP operation can be constructed using a combination of $C_n\text{NOT}_e$ and single qubit rotations, where $C_n\text{NOT}_e$ is sketched in Fig. 2c. Analytically, the SWAP operation can be expressed as:

$$\begin{aligned} U_{\text{SWAP}} &= U_{\text{XY-8}} \cdot R_y(-\pi/2) \otimes R_z(\pi/2) \cdot U_{\text{XY-8}} \cdot R_x(\pi/2) \otimes R_z(\pi/2) \cdot U_{\text{XY-8}} \\ &= -e^{i\pi/4} (|00\rangle\langle 00| - i|01\rangle\langle 10| - |10\rangle\langle 01| - i|11\rangle\langle 11|) \end{aligned} \quad (16)$$

Here, $U_{\text{XY-8}} = |0\rangle\langle 0| \otimes R_x(\pi/2) + |1\rangle\langle 1| \otimes R_x(-\pi/2)$, is the conditional- $R_x(\pm\pi/2)$ unitary resulting from the XY-8 sequence (Eq. 9). This can be transformed into an iSWAP gate by using single qubit $\hat{\mathbf{z}}$ -rotations before and after U_{SWAP} :

$$\begin{aligned} \text{iSWAP} &= R_z(5\pi/4) \otimes R_z(0) \cdot U_{\text{SWAP}} \cdot R_z(\pi) \otimes R_z(\pi/4) \\ &= |00\rangle\langle 00| + i|01\rangle\langle 10| + i|10\rangle\langle 01| + |11\rangle\langle 11| \end{aligned} \quad (17)$$

As the $R_z(\pi/2)$ rotation on the nuclear spin is realized via its free-precession, the fidelity of U_{SWAP} is limited primarily by the T_2^* of the nuclear spin. In particular, the presence of four distinct nuclear spin precession frequencies, as shown in Fig. 2c, should be considered when estimating the fidelity of the operation.

3.2 Description of error channels during the SWAP experiment

The primary sources of error are: (1) modulation of the nuclear spin precession frequency; and (2) variation in the hyperfine interaction strength when the electron is in the excited state (during initialization and readout). These can be represented as trace-preserving quantum channels acting on the two-qubit density matrix, ρ . Accounting for these errors in the SWAP experiment (Fig. 3), we can calculate a lower bound for the nuclear spin T_1 .

First, let's incorporate the effects of precession frequency modulation using a quantum channel description. The basic building block of the SWAP operation is the free evolution of our two-body system for

the duration τ_0 , realizing both the XY-8 sequence and the $\hat{\mathbf{z}}$ -rotation of the nuclear spin. We label the corresponding quantum channel as $S_{\tau_0}^i(\rho)$, where the superscript i indicates that the nuclear spin precesses at one of the four frequencies, $\omega_{L,i} = \omega_L + \Delta_i$ ($\Delta_i \in \{\pm 5, \pm 9\}$ kHz), as identified in Fig. 2c. This basic channel is calculated by evolving ρ under the hyperfine Hamiltonian (Eq. 1) and electron spin dephasing (Fig. 1c) for duration τ_0 under the Linblad master equation. Corresponding SWAP channels, $\mathcal{S}_{\text{SWAP}}^i(\rho)$, can be obtained by interleaving $S_{\tau_0}^i(\rho)$ with unitary channels to apply single-qubit rotations on the electron spin as described in Sec. 3.1. The final SWAP channel, $\mathcal{S}_{\text{SWAP}}(\rho)$, averages over the four precession frequencies for the nuclear spin:

$$\mathcal{S}_{\text{SWAP}}(\rho) = \frac{1}{4} \sum_{i=1}^4 \mathcal{S}_{\text{SWAP}}^i(\rho) \quad (18)$$

On the other hand, errors due to variation in hyperfine interaction strength can also be accounted for using a similar quantum channel description. The action of a spin-selective excitation pulse can be understood as an infinitesimal sum over unitary operations, corresponding to the variable decay time of the electron from the excited state back to the ground state. First, we give a quantum channel description of the readout scheme sketched in Fig. 1b. An optical excitation pulse projects the electron spin to the states $|1\rangle$ and $|0\rangle$, via the projectors P_1 and P_0 , respectively. If the electron is in the $|1\rangle$ state, it is excited and undergoes a unitary evolution under the excited state Hamiltonian H_e^i for time t and the ground state Hamiltonian H_g^i for the remainder of the photon collection window t_W (120 μs), where i stands for the index of $\omega_{L,i}$ in each Hamiltonian. If the electron is in the $|0\rangle$ state, it remains unaffected by the optical excitation and simply evolves under H_g^i for duration t_W . Finally, if the electron has not decayed within this time, it will evolve under H_e^i for duration t_W . As with the SWAP channel (Eq. 18), we also consider the variation in the nuclear spin Larmor frequency, but here, we ignore electron spin dephasing as the electron is projected to its computational basis after readout. We provide below a definition of the excitation channel, $\mathcal{S}_{\text{excite}}^1(\rho)$, for the $|1\rangle$ transition used in the readout:

$$\begin{aligned} \mathcal{S}_{\text{excite}}^1(\rho) &= S(0, P_0 \rho P_0) + p_R S(t_W, P_1 \rho P_1) + \int_0^{t_W} dt p(t) S(t, P_1 \rho P_1) \\ S(t, \rho) &= \frac{1}{4} \sum_i U^i(t) \rho U^{i\dagger}(t); \quad U^i(t) = e^{-iH_g^i(t_W-t)} e^{-iH_e^i t} \\ p(t) &= \frac{1}{T_{1,\text{op}}} e^{-t/T_{1,\text{op}}}; \quad p_R = 1 - \int_0^{t_W} p(t) dt \end{aligned} \quad (19)$$

Note that the excitation channel description for initialization, $\mathcal{S}_{\text{excite}}^0(\rho)$, can be obtained similarly by swapping the projectors P_0 and P_1 . We measure the optical lifetime $T_{1,\text{op}}$ (60 μs) in an independent measurement and calculate H_e^i based on the reported g-tensor [3] and estimated position for the nuclear spin relative to the Er^{3+} ion (Fig. 4). As discussed in Sec. 3.4 with a simple example, in the strong magnetic field regime, the effect of the excitation channel is primarily a dephasing of the nuclear spin due to the uncertainty in phase accumulated by the nuclear spin during the excitation and decay of the electron spin.

Using Eq. 19 as the basic block, we can also construct the full quantum channel description of the readout and initialization processes, consisting of 450 and 40 optical pulses, respectively (Fig. 1c). Importantly, we take into account the finite cyclicity of the optical transition to induce spin-flips, $p_f = 0.2\%$ and the excited state MW π -pulse during the initialization. This means that the electron spin will only be excited until it is optically pumped to the other spin state.

3.3 Estimate of nuclear spin T_1

With the channels described above, we can simulate the SWAP experiment (Fig. 3), consisting of multiple rounds of initialization, SWAP, and readout. To estimate the nuclear spin T_1 , we extract the fidelity of the SWAP from the experiment and compute the same quantity based on our theoretical model. The difference

between the two lets us put a lower bound on the nuclear spin T_1 . To extract the fidelity, we consider the initialization i_n and readout m_0 , shown in Fig. 3, where the SWAP operation is applied twice for storage and retrieval of the computational basis states $|0\rangle$ ($|\uparrow\rangle$) and $|1\rangle$ ($|\downarrow\rangle$). An important implementation detail is that the steps for initialization, SWAP, and readout are looped multiple times during the experiment. Therefore, Fig. 3 can be equivalently sketched as Fig. S2. The Fig. 3 histogram indices can then be re-labeled as $(i_e, i_n) \equiv (i_{k+1}, i_k)$ and $(m_0, m_1) \equiv (m_{k+1}, m_{k+2})$, keeping in mind the modular structure.

After accounting for all known sources of errors described in Sec. 3.2, we attribute the remaining difference in simulation and experiment fidelity to the nuclear spin T_1 , as in $F_{\text{exp}} = e^{-T_{\text{store}}/T_1} F_{\text{sim}}$, where $T_{\text{store}} = 59.2$ ms is the time between the initialization i_k and readout m_{k+1} , while $F_{\text{exp}} = 0.76$ and $F_{\text{sim}} = 0.84$ are the experiment and simulation fidelities for retrieval of the initialization i_k at m_{k+1} . Attributing this remaining difference, to the nuclear spin lifetime allows us to put a lower bound on the T_1 of 0.63 seconds. However, the difference could also originate from other sources of error that is not included in the simulations. Finally, considering that the SWAP acts twice during the computation of the fidelities ($F_{\text{exp}}, F_{\text{sim}}$), we can extract a SWAP fidelity in the computational basis of $F_{\text{SWAP,exp}} = 87\%$ and $F_{\text{SWAP,sim}} = 91\%$ for the experiment and simulation, respectively.

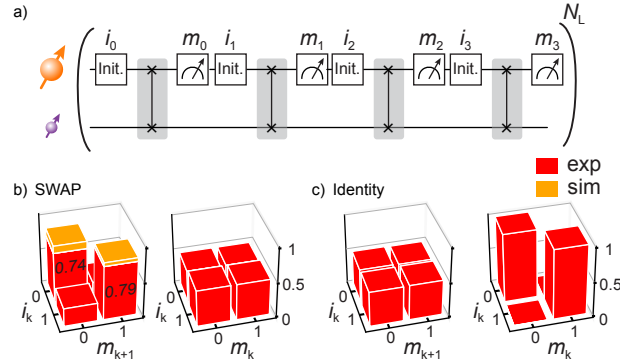


FIG. S2: **SWAP experiment.** (a) Implementation of the SWAP experiment, where a SWAP operation is interleaved between each initialization i_k and subsequent readout m_k . A sequence of initialization $(i_0, i_1, i_2, i_3) = (0, 0, 1, 1)$ is repeated $N_L = 1500$ times in a loop such that all instances $(i_k, i_{k+1}) = \{00, 01, 10, 11\}$ are realized. This allows for all initialization combinations of the electron-nuclear spin pair in the computational basis to be generated. (b) The histogram verifies that the initialization i_k is readout at m_{k+1} , while being uncorrelated with m_k . The average success rate for retrieving the information in experiment (simulation) is 76% (84%). (c) A control experiment, where the SWAP operation is replaced by an identity, shows that i_k is fully correlated with m_k and uncorrelated with m_{k+1} as expected.

3.4 Discussion on storing arbitrary states

Although we demonstrate information storage and retrieval in the computational basis, it is of greater interest to store an arbitrary state in the nuclear spin memory. To consider this plausibility, we discuss a simple problem of nuclear spin dephasing under optical excitation of the electron. To obtain a simple analytical expression, we only consider changes in the parallel hyperfine term $A_{||}$, which largely accounts for the dephasing in the strong field regime, using the ground and excited state Hamiltonians, $H_g = 2A_g S_z I_z + \omega_L I_z$ and $H_e = 2A_e S_z I_z + \omega_L I_z$, respectively. Limiting ourselves to the subspace where the electron is in the $|1\rangle$ state and moving to a rotating frame of the nuclear spin at $\omega_L - A_g$, we have the nuclear spin Hamiltonians conditional on the electron excitation state:

$$H_g = 0, \quad H_e = \delta_A I_z, \quad \delta_A = A_g - A_e \quad (20)$$

Starting with the nuclear spin state in an equal superposition, $|\psi_n\rangle = 1/\sqrt{2}(|0\rangle + |1\rangle)$ (which is most sensitive to dephasing), we can time evolve the state as:

$$\rho(t) = |\psi_n(t)\rangle \langle \psi_n(t)| = \frac{1}{2}(|0\rangle \langle 0| + |1\rangle \langle 1| + e^{i\delta_A t'} |0\rangle \langle 1| + e^{-i\delta_A t'} |1\rangle \langle 0|), \quad (21)$$

where the electron was excited at $t = 0$ and decayed at $t = t'$. Assuming a standard probability distribution for the excited state decay, $P(t) = \gamma e^{-\gamma t}$, we can calculate the density matrix for the nuclear spin after one decay of the electron as:

$$\rho_n^{(1)} = \int_0^\infty dt' P(t') \rho(t') = \frac{1}{2}(|0\rangle \langle 0| + |1\rangle \langle 1| + c|0\rangle \langle 1| + c^*|1\rangle \langle 0|) \quad c = \frac{1/\tau}{1/\tau - i\delta_A} \quad (22)$$

The nuclear spin density matrix after N excitations will simply result from propagating the density matrix through repeated decays in the same manner:

$$\rho_n^{(N)} = \frac{1}{2}(|0\rangle \langle 0| + |1\rangle \langle 1| + c^N |0\rangle \langle 1| + c^{*N} |1\rangle \langle 0|) \quad (23)$$

Therefore, the purity of the density matrix can be expressed as the length of the Bloch vector magnitude, where $\delta_f = \delta_A/(2\pi)$:

$$F = \sqrt{2\text{Tr}[\rho_n^{(N)2}] - 1} = \left(\frac{1}{\sqrt{1 + 4\pi^2 \delta_f^2 / \gamma^2}} \right)^N \quad (24)$$

Based on our parameters, $\delta_f/\gamma = 0.56$, the Bloch vector length for a superposition state reduces to $F = 0.3$ after a single excitation. In order to achieve $F = 0.9$ after 450 excitations (typically used in single shot readout), we need $\delta_f/\gamma = 0.0034$. The required factor of ~ 150 improvement in the ratio can be plausibly obtained by a combination of using more weakly coupled nuclear spins and choosing a magnetic field direction to minimize δ_f or increasing the decay rate, γ , via stronger Purcell enhancement of the optical transition. As commented on Ref. [3], the $\text{Er}^{3+}:\text{YSO}$ g-tensors for the ground state ($^4I_{15/2}$) and excited state ($^4I_{13/2}$) allows for choosing magnetic field directions such that the interaction of the electronic spin with the bath is unperturbed by an excitation. This can then allow information to remain protected in the nuclear spin during a spin-photon entanglement generation attempt that requires repeated excitations of the electron spin.

4 Nuclear spin search

4.1 Obtaining hyperfine parameters from magnetic dipole Hamiltonian

In order to locate the nuclear spin with respect to the electron, we can study the position dependence of the hyperfine parameters ($A_{||}, A_{\perp}, \omega_L$). The hyperfine Hamiltonian (Eq. 1), expressed in terms of these parameters, can be obtained from the magnetic dipole Hamiltonian between the Er^{3+} spin and the nuclear spin:

$$H_{\text{dip}} = -\frac{\mu_0}{4\pi r^3} \mu_B \mu_N g_n \left(\vec{g} \cdot \mathbf{S} \cdot \mathbf{I} - 3(\vec{g} \cdot \hat{\mathbf{r}})(\mathbf{I} \cdot \hat{\mathbf{r}}) \right), \quad (25)$$

where μ_B is the Bohr magneton, μ_N is the nuclear magneton, μ_0 is the magnetic permeability, g_n is the nuclear spin g-factor, $\vec{g}$ is the electron spin g-tensor in the solid state host, r is the distance between the electron and nuclear spin and $\hat{\mathbf{r}}$ is the direction. Under a static magnetic field, each spin also obtains a Zeeman Hamiltonian:

$$H_Z = \mu_B \mathbf{B} \cdot \vec{g} \mathbf{S}' - \mu_N g_n \mathbf{B} \cdot \mathbf{I}', \quad (26)$$

where the magnetic field vector $\mathbf{B}$ and electron (nuclear) spin vector $\mathbf{S}'$ ($\mathbf{I}'$) are in the lab-frame. The S_z and I_z operators are defined using these terms:

$$S_z = \mathbf{B} \cdot \vec{g} \mathbf{S}' / |\mathbf{B} \vec{g}|, \quad I_z = \mathbf{B} \cdot \mathbf{I}' / |\mathbf{B}| \quad (27)$$

This yields the electron and nuclear spin Larmor frequencies as $\omega_{L,e} = \mu_B |\mathbf{B} \vec{g}|$ and $\omega_{L,n} = \mu_N g_n |\mathbf{B}| = \gamma_N |\mathbf{B}|$ (γ_N being the gyromagnetic ratio), yielding $H_Z = \omega_{L,e} S_z + \omega_{L,n} I_z$.

As described in the main text, we work in the frame rotating with $\omega_{L,e} S_z$ and under the secular approximation such that terms that do not commute with S_z are discarded. The Hamiltonian in Eq. 25 then reduces to $H_{\text{dip,RWA}} = \sum_i 2A_i S_z I_i$, where $A_i = \text{Tr}[\{H_{\text{dip}}, S_z I_i\}]$ can be extracted via anti-commutation relations. We can further simplify the interaction terms A_i into $A_{\parallel} = A_z$ and $A_{\perp} = \sqrt{A_x^2 + A_y^2}$, since I_x and I_y operators can always be rotated such that A_y is set to 0 and $A_x \geq 0$. This reduces $H_{\text{dip,RWA}}$ to a simple form of $H = 2S_z(A_{\parallel} I_z + A_{\perp} I_x) + \omega_L I_z$, which is provided in the main text and is defined in terms of only three parameters.

Then, the only unknown variables in determining the parameters ($A_{\parallel}, A_{\perp}, \omega_L$) are γ_N , r , and $\hat{\mathbf{r}}$, which tell us the species and location of the nuclear spin. The problem is made simpler by observing that the ratio $A_{\parallel}/A_{\perp}$ only depends on $\hat{\mathbf{r}}$ since both the numerator and denominator of the ratio varies proportionately with γ_N/r^3 . γ_N can also be determined independently from the Larmor frequency ω_L . The remaining unknown, r , can then be determined from $\sqrt{A_{\parallel}^2 + A_{\perp}^2}$ values, given $\hat{\mathbf{r}}$ and γ_N . We use this approach to roughly determine the location and constrain the volume considered in the χ^2 calculation to report the most likely position.

4.2 Determining location

We parameterize the χ^2 calculation in terms of the nuclear spin location $\mathbf{r}(\equiv r\hat{\mathbf{r}})$ with respect to the Er^{3+} ion. We directly compare the experimental measurements of $(\omega_0, \omega_{\delta}, \alpha)$, discussed in Sec. 2.2, to model estimates of the same measurements as a function of $\mathbf{r}$:

$$\chi^2(\mathbf{r}) = \sum_i \frac{(x_{\text{obs},i} - x_{\text{mod},i}(\mathbf{r}))^2}{\sigma_{\text{obs},i}^2 + \sigma_{\text{mod},i}^2(\mathbf{r})} \quad (28)$$

Here, $x_{\text{obs},i}$ and $x_{\text{mod},i}$ correspond to the observed and estimated values of ω_0 , ω_{δ} and α at four different magnetic field orientations as indicated in Fig. 4 of the main text, such that there are twelve data points that are compared in total and three parameters, (r_H, θ_H, ϕ_H) , to optimize. $\sigma_{\text{obs},i}$ and $\sigma_{\text{mod},i}$ correspond to the errors in each data point, where the former arises due to experimental uncertainties and the latter is obtained from Monte-Carlo simulations as explained below.

Uncertainty in the model estimates originates from errors in the static magnetic field orientation. We estimate the orientation uncertainty by allowing a shift of the intended field to first order, as in $(B + \Delta B, \theta + \Delta\theta, \phi + \Delta\phi)$, where B is the field magnitude, and (θ, ϕ) is the magnetic field orientation with respect to the (D_1, D_2, b) axes of the crystal [3] (Fig. 1a). We choose $(\Delta B, \Delta\theta, \Delta\phi)$ such that the measured splitting for the ground and excited state spin levels using optically detected magnetic resonance (ODMR) are minimized with respect to the predicted values at the field settings used. This yields $\Delta B = 3.99 \pm 0.76$ G, $\Delta\theta = 0.89^\circ \pm 0.34^\circ$ and $\Delta\phi = 0.79^\circ \pm 0.44^\circ$. The correction yields a field strength of $B = 134$ G for the four measurements indicated in Fig. 4 of the main text. Based on the ω_L values obtained at each orientation, we estimate a gyromagnetic ratio γ_N of 42.6 ± 0.4 MHz/T. Since this value is consistent with the Hydrogen gyromagnetic ratio, 42.58 MHz/T, we set the Hydrogen g-factor as a constant in the χ^2 calculations. To calculate the uncertainty in the model estimates, we propagate the uncertainty in the magnetic field, via a Monte Carlo simulation, for the model estimates $x_{\text{obs},i}$ corresponding to ω_0 , ω_{δ} and α . Minimizing χ^2 as a function of $\mathbf{r}$ near the expected locations leads us to the final estimates of the nuclear spin position, indicated in Fig. 4. At these locations, we find a reduced chi-square of $\min_{\mathbf{r}}(\chi^2)/\nu = 2.7$ for both signs of $A_{\parallel}$, where the degree of freedom $\nu = 12 - 3 = 9$ is based on the number of measurement variables (12) and fit parameters (3). The obtained value is not far from a reduced chi-square of 1 and the remaining inconsistency may be due two reasons: first, the correction to the magnetic field is only a first order correction, which does not entirely resolve the discrepancy between the expected and measured splitting values; and second, uncertainties in the reported g-tensor itself [3] are not taken into account in our calculations.

4.3 Hydrogen concentration estimate

To consider the plausibility of the presence of hydrogen (H) in the YSO crystal, we provide an estimate of H concentration. A confidence interval on the H concentration can be calculated based on our observation of H in the vicinity of one out of six total Er^{3+} ions studied in this sample. The purpose of this calculation is to provide a rough estimate of the concentration given available information.

The probability to observe some number k of hydrogen atoms within a given volume V and concentration ρ is described by the Poisson distribution: $P(k; \lambda) = (\lambda^k e^{-\lambda})/k!$, where $\lambda = \rho V$ is the expected number of H atoms in a given volume. Then, the probability of not observing hydrogen within the same volume is $P_0(\rho) = e^{-\rho V}$ and observing at least one H is $P_1(\rho) = 1 - P_0(\rho)$. Therefore, the probability to observe an H atom in the vicinity of n out of m Er^{3+} ions is: $P(O|\rho) = \binom{n}{m} P_0(\rho)^{m-n} P_1(\rho)^n$. The probability distribution for the Hydrogen concentration, constrained only by our observation, is then given by:

$$p(\rho|O) = \frac{P(O|\rho)}{\int_0^\infty d\rho' P(O|\rho')}, \quad (29)$$

where we assume that prior probability for the hydrogen concentration is uniform, i.e. $P(\rho) = P(\rho')$. The probability for the concentration to lie within a given range, (ρ_1, ρ_2) , is then obtained by integrating the above expression: $P(\rho_1 \leq \rho \leq \rho_2|O) = \int_{\rho_1}^{\rho_2} p(\rho|O) d\rho$.

With 1 observation out of 6 trials ($n = 1, m = 6$), we obtain the probability distribution: $p(\rho|O) = 30V(e^{-5\rho V} - e^{-6\rho V})$. We define the observable volume, $V = 4/3\pi r_{\text{obs}}^3$, to be a sphere around the Er^{3+} ion. The observation of an H nuclear spin ~ 2 nm away from the Er^{3+} ion resulted in a near full contrast peak in the XY-16 sequence. However, farther H with weaker signals could also be observed provided that they are above the noise floor by a margin. To be 5σ above the noise floor at the resonant time, τ_0 , we find that a conditional rotation of $N\alpha \simeq \pi/4$ is sufficient, in contrast to $N\alpha \simeq \pi$ for the observed interaction at $N = 16$ pulses for XY-16. The corresponding reduction in interaction strength, by a factor of ~ 4 , allows for a hydrogen located farther by a factor of ~ 1.5 or about 3 nm away from the Er^{3+} ion. Therefore, we set $r_{\text{obs}} = 3$ nm for the observable volume. Based on the probability distribution, we find that the Hydrogen concentration is in the range $0.3 - 3.9 \times 10^{18} \text{ cm}^{-3}$ with 68% confidence, with the likeliest concentration at $1.6 \times 10^{18} \text{ cm}^{-3}$ (Fig. S3).

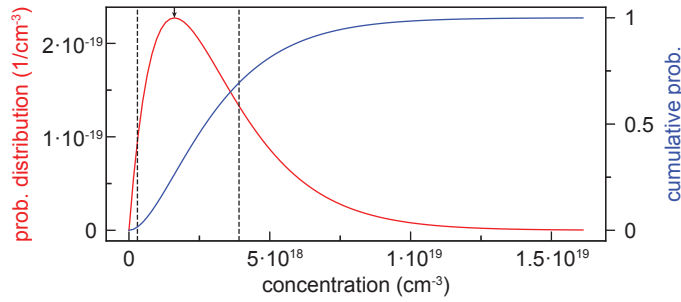


FIG. S3: **Estimating Hydrogen concentration.** The probability distribution for the Hydrogen concentration, $p(\rho|O)$ (red), and the cumulative probability function $P(\rho|O) = \int_0^\rho p(\rho'|O) d\rho'$ (blue), as a function of concentration in cm^{-3} units. The dashed lines indicate the 68% confidence range from $0.3 \times 10^{18} \text{ cm}^{-3}$ to $3.9 \times 10^{18} \text{ cm}^{-3}$. The probability distribution has a maximum at $1.6 \times 10^{18} \text{ cm}^{-3}$.

5 Nuclear spin environment

5.1 Shifts in precession frequency

When measuring the free precession of the nuclear spin (Fig. 2b,c of the main text), we observe discrete shifts in the precession frequency of the nuclear spin. To explore this further, let's look at an example

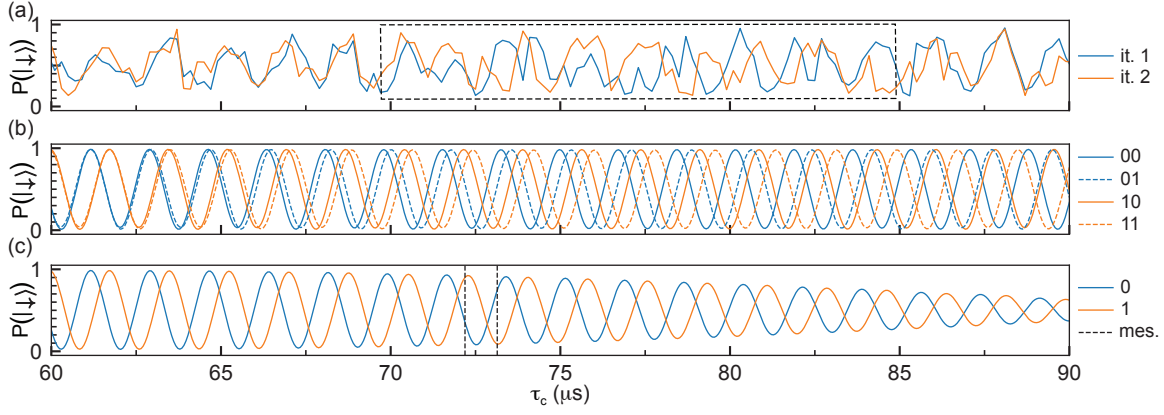


FIG. S4: **Observing frequency shifts.** (a) The free precession signal $s_0(\tau_c)$, which is expected to oscillate at ω_0 , displays discrete shifts in frequency. This is manifested as a phase difference at the time scale above for two different iterations of the experiment. (b) Simulated precession signal $s_0(\tau_c)_{ij}$ for each scenario of the dark-spin pair initialization. (c) Simulated precession signal corresponding only to the d_1 state, after averaging over the d_2 states. Both readout locations (dashed lines) allow for distinguishing between d_1 states but with opposite populations.

provided in Fig. S4a of the same experiment, where two different iterations of the experiment are out of phase with respect to each other, for the same precession times.

The frequencies are resolved in the FFT of the precession signal (Fig. 2c) and can be summarized as $\omega = \{\omega_0 + (-1)^i A_1 + (-1)^j A_2 : i, j \in (0, 1)\}$ where ω_0 is the mean precession frequency, $A_1 = 2.25$ kHz and $A_2 = 7.18$ kHz are shifts in frequency. This motivates a simple model, where the nuclear spin interacts with two ‘dark spins’, in the form $H_{\text{dark}} = 2I_z(A_1 I_{z,d_1} + A_2 I_{z,d_2})$, where the precession frequency of the nuclear spin is shifted by $\pm A_1$ and $\pm A_2$ depending on the state of dark spins d_1 and d_2 . Adding H_{dark} terms to the hyperfine interaction Hamiltonian, we get the following four-body Hamiltonian:

$$\tilde{H} = 2S_z(A_{\parallel}I_z + A_{\perp}I_x) + (\omega_L + 2A_1 I_{z,d_1} + 2A_2 I_{z,d_2})I_z \quad (30)$$

Using the above Hamiltonian, we can simulate the expected signal $s_0(\tau_c)_{ij}$ for a Ramsey experiment (Eq. 12), where the indices (i, j) corresponds to the dark spin states which shift the precession frequency. In particular, we observe that curves with opposite d_2 states are roughly in phase with each other, while the d_1 state translates to a large phase offset, at the beginning of the time window in Fig. S4b. This can be understood in terms of the phase difference accumulated due to d_1 and d_2 as a function of τ_c , $\phi_i(\tau_c) = 2A_i(\tau_c + 2N\tau_0)$, ($N = 8$) where the second term is a correction due to the finite duration of the $C_n \text{NOT}_e$ operation, during which the frequency of the nuclear spin is still shifted. This tells us that $\phi_2(\tau_c) = 2\pi$ at $\tau_c = 62.6 \mu\text{s}$, such that the any population readout near this time window will be insensitive to the state of d_2 .

Furthermore, the phase accumulated due to d_1 at $\tau_{c,1} = 72.19 \mu\text{s}$ and $\tau_{c,2} = 73.14 \mu\text{s}$ is as large as $\phi_1(\tau_c) \sim 0.7\pi$, such that the population is very sensitive to the d_1 state. Although larger phases can be accumulated for d_1 at later times, the d_2 state also becomes significant. Based on this model, we perform readout of d_1 by repeatedly measuring $s_0(\tau_c)$ at $\tau_{c,1}$ and $\tau_{c,2}$. In Fig. S5a, we repeatedly perform the Ramsey experiment at times $\tau_{c,1}$ and $\tau_{c,2}$. At these times, we observe anti-correlated jumps in the signal that we attribute to quantum jumps of the d_1 state, as expected from Fig. S4c. By thresholding based on a histogram of Fig. S5b, we extract 98% readout fidelity for d_1 and extract its lifetime by looking at the frequency of jump events (Fig. S5c). The measurement time (29 seconds) between consecutive data points in the Fig. S5a was small enough to observe d_1 jumps occurring with a lifetime of 5 mins.

In order to observe d_2 , we work at a value of τ_c that accumulates a phase for d_2 of $\phi_2(\tau_c) \sim 1.2\pi$, which is also uncorrelated with the d_1 state. Although d_1 already contributes an appreciable phase here, there still exist points in time where oscillations with a phase-offset will intersect such that the population is insensitive

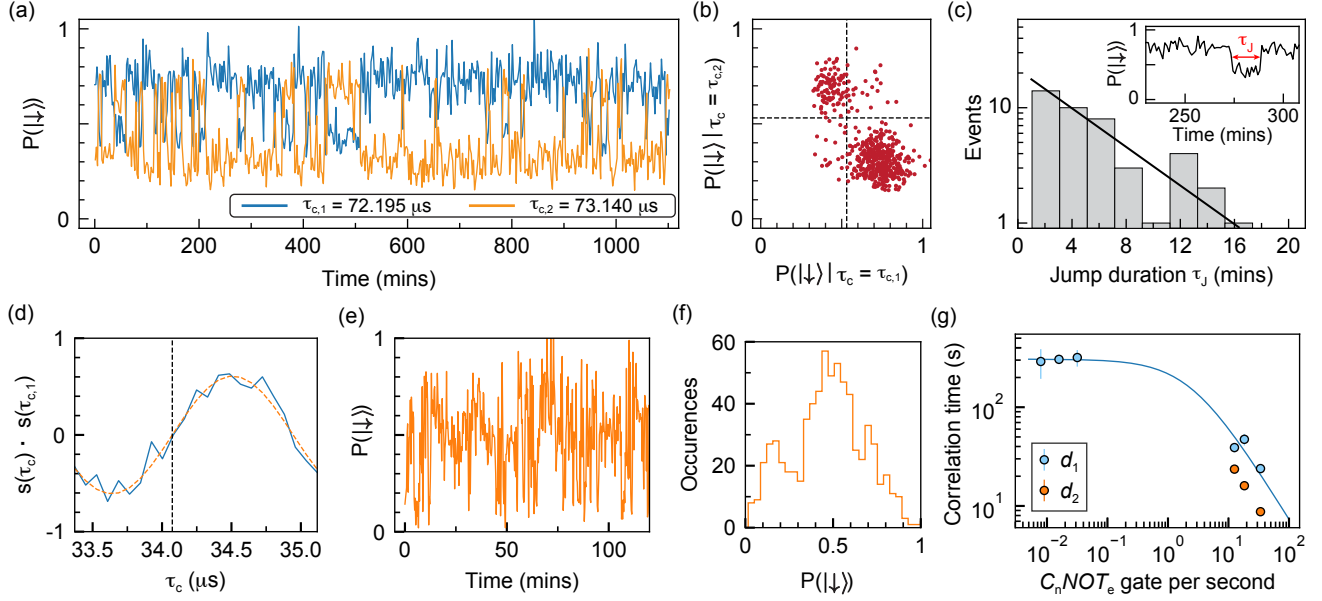


FIG. S5: **Dark spin readout.** (a) Repeated Ramsey measurements at two different τ_c reveal quantum jumps of d_1 . (b) Distribution of the measured populations in (a) are unequally weighted bi-modal and anti-correlated at the two τ_c . (c) Duration of the quantum jumps shown in (a) is consistent with a lifetime of 5.19 ± 0.55 mins (black line is the fitted exponential decay). Inset: A representative jump event from (a). (d) Cross-correlation measurement comparing time traces equivalent to (a) at τ_c with the same at $\tau_{c,1}$ (where we observe d_1 jumps) in order to find a time point that is not correlated with the d_1 state. The vertical line indicates the point of no-correlation with the d_1 state $\tau_c = 34.07 \mu\text{s}$. (e) Repetitive Ramsey measurements for d_2 performed at $\tau_c = 34.07$, displaying weaker amplitude jumps. (f) Population histogram of d_2 extracted from (d). (g) Measured spin relaxation times at varying repetition rates of the $C_n\text{NOT}_e$ operation. At low repetition rates, the spin relaxation time approaches an intrinsic value $T_{1,\text{dark}} = 5.12 \pm 0.13$ mins for d_1 .

to the d_1 state. The cross-correlation of time-traces $s(\tau_c)$ and $s(\tau_{c,1})$, yields a point of no correlation with the d_1 state, which we use to readout d_2 (Fig. S5d). By performing faster measurements (every 5.9 seconds), we were able to observe quantum jumps associated with d_2 , revealing a bi-modal distribution (Fig. S5e,f). The lifetime associated with d_1 and d_2 reveal a dependency on the rate of $C_n\text{NOT}_e$ operations applied (Fig. S5g), which suggests that d_1 and d_2 are perturbed by the probing of the nuclear spin.

As discussed in the main text, a direct interaction between the electron spin and the dark spins was investigated using a DEER sequence [4] under the assumption that these were electron spins with $2 < g < 14$, but the spectroscopy did not reveal such interactions. We can verify that the shift in frequency is not due to a global effect, such as static magnetic field fluctuations, given the stable ground and excited spin resonance frequency of the Er^{3+} ion. Here, we propose two mechanisms to explain the frequency shift. d_1 and d_2 can be spin-1/2 nuclear spins in the YSO lattice: the hydrogen atom interacts with two nearby nuclear spins with interaction strengths of 2 kHz and 7 kHz, respectively. Alternatively, d_1 and d_2 could be represented by a single nuclear spin coupled to a tunneling hydrogen atom, such that the interaction strength modulates between 5 kHz and 9 kHz, due to relative changes in their position. In order to explore the origin of the local environment factor that cause the coupling of H with nearby nuclear spins, we perform density functional theory (DFT) computation of H-related defect complexes in YSO and propose possible defects that can explain the frequency shift.

5.2 Density functional theory study of the origin of frequency shifts

Hydrogen is a ubiquitous impurity in semiconductors and insulators, as many growth and post-growth processes introduce H. Here we perform a systematic study of the structure and formation energies of hydrogen-related centers in YSO. Hydrogen can be present as hydrogen interstitials or as substitutional H (H_O), and can also form complexes with impurities or with vacancies. We consider impurities that have nonzero nuclear spin and that based on chemical analysis are expected to be present in the sample: applying these criteria, we focus on Cd_Y and P_O , which act as acceptors and can form complexes with H.

Our density functional theory (DFT) calculations are performed using the projector augmented wave method implemented in the Vienna *Ab-initio* Simulation Package (VASP) [5, 6]. The plane-wave cutoff energy is 500 eV. The hybrid exchange-correlation functional of Heyd, Scuseria, and Ernzerhof (HSE) [7] is used with 25% mixing of Hartree-Fock exchange, which yields the direct band gap of 6.49 eV, in reasonable agreement with the experimental band gap (6.14 eV) [8]. The computed lattice constants of YSO are $a = 14.40 \text{ \AA}$, $b = 6.75 \text{ \AA}$, $c = 10.44 \text{ \AA}$, $\beta = 122.1^\circ$, in very good agreement with the experimental values ($a = 14.37 \text{ \AA}$, $b = 6.71 \text{ \AA}$, $c = 10.40 \text{ \AA}$, $\beta = 122.2^\circ$) [9].

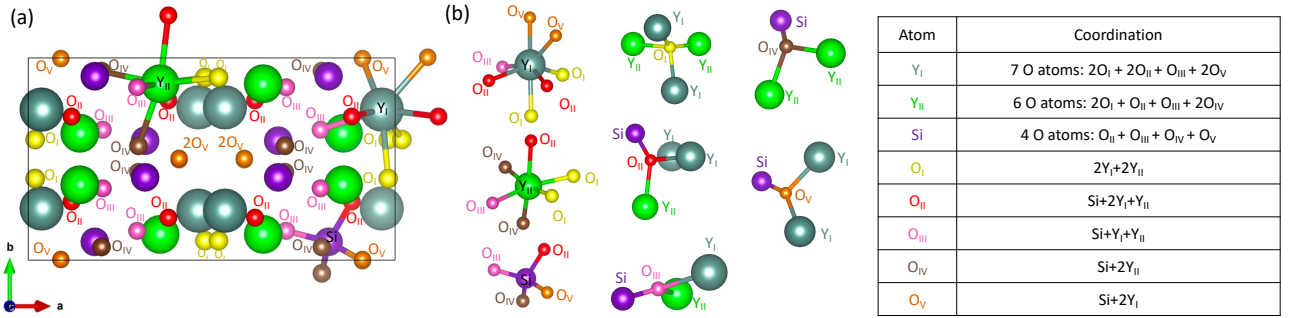


FIG. S6: (a) Structure of Y_2SiO_5 (YSO). The unit cell is indicated and contains 64 atoms. (b) Coordination of Y, Si, and O atoms in Y_2SiO_5

The crystal structure of YSO (with $\text{C}/2\text{m}$ space group symmetry) is sketched in Fig. S6a. YSO has two types of Y atoms (Y_I and Y_II), one type of Si atom, and five types of O atoms (O_I , O_II , O_III , O_IV , and O_V). Fig. S6b summarizes the coordination of Y, Si, and O atoms. To calculate defect structures, we construct a supercell with dimensions $a \times 2b \times c$, containing 128 atoms. We consider the following defect structures that could involve H interacting with cations that have nuclear spin 1/2: H interstitials (H_i), substitutional hydrogen (which can also be viewed as an oxygen vacancy–H complex) (H_O), Si vacancy–H complex (V_Si –H), Y vacancy–H complex (V_Y –H), substitutional Cd–substitutional H complex (Cd_Y –H), and substitutional P–H complex (P_O –H).

The formation energy $E^\text{f}(D^q)$ of a point defect D in charge state q is calculated as:

$$E^\text{f}(D^q) = E(D^q) - E_\text{bulk} + \sum \mu_i n_i + qE_\text{F} + \Delta_\text{corr}. \quad (31)$$

$E^\text{f}(D^q)$ is the total energy of the supercell containing defect D in the charge state q . E_bulk is the total energy of the perfect supercell. $|n_i|$ is the number of atoms added ($n_i < 0$) or removed ($n_i > 0$) from the system. μ_i is the chemical potential of species i and E_F is the Fermi level, which is referenced to the valence band maximum (VBM). Δ_corr is a term that corrects for the finite size of charged supercells.

We define $\Delta\mu_i$ as the deviation of the chemical potential from the reference states: $\mu_i = \mu_{i,\text{ref}} + \Delta\mu_i$. For Y (Si), $\mu_{i,\text{ref}}$ is the energy of bulk Y (Si), and for oxygen, the reference is an O_2 molecule. Assuming thermodynamic equilibrium, the $\Delta\mu_i$ are related by

$$2\Delta\mu_\text{Y} + \Delta\mu_\text{Si} + 5\Delta\mu_\text{O} = \Delta H^\text{f}(\text{Y}_2\text{SiO}_5) \quad (32)$$

$\Delta H^f(\text{Y}_2\text{SiO}_5)$ is the formation enthalpy of YSO. The $\Delta\mu_i$ are also limited by the formation of other compounds: $2\Delta\mu_{\text{Y}} + 3\Delta\mu_{\text{O}} < \Delta H^f(\text{Y}_2\text{O}_3)$, $\Delta\mu_{\text{Si}} + 2\Delta\mu_{\text{O}} < \Delta H^f(\text{SiO}_2)$, and $2\Delta\mu_{\text{H}} + \Delta\mu_{\text{O}} < \Delta H^f(\text{H}_2\text{O})$. $\Delta H^f(\text{Y}_2\text{O}_3) = -19.23$ eV, $\Delta H^f(\text{SiO}_2) = -8.97$ eV, $\Delta H^f(\text{H}_2\text{O}) = -2.65$ eV and are the formation enthalpy of Y_2O_3 , SiO_2 , and H_2O . The constraints imposed by Y_2O_3 , SiO_2 , and H_2O define a stability region for YSO, which is shown in Fig. S7a. We chose to use the O-rich, Si-rich (Fig. S7b) and O-poor, Si-rich (Fig. S7c) conditions to present the formation energies.

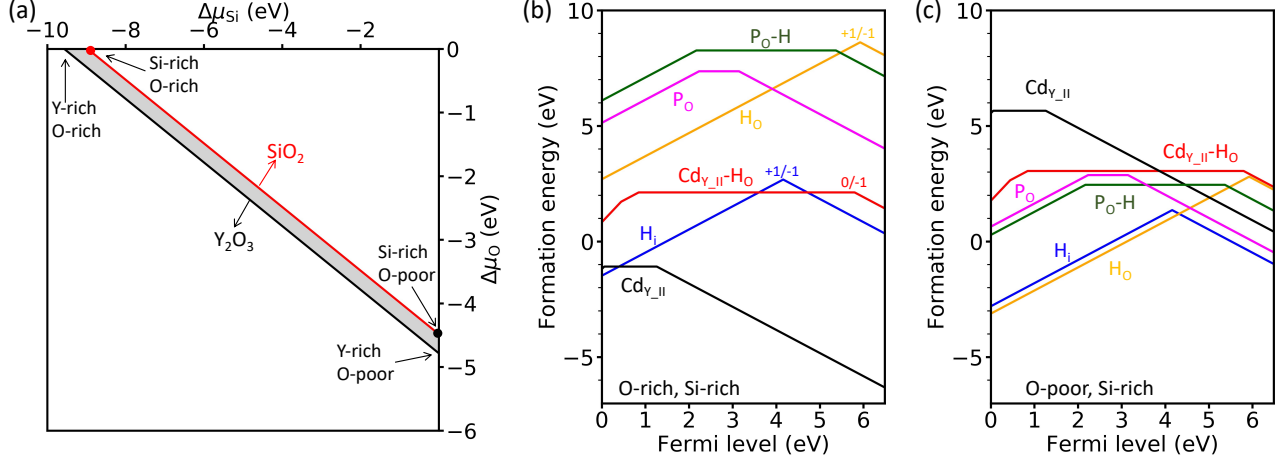


FIG. S7: (a) Stability region for Y_2SiO_5 shaded in grey. The red (O-rich, Si-rich) and black (O-poor, Si-rich) dots represent the choice of chemical potentials in this study. (b)-(c) Formation energy diagram of defects under (b) O-rich, Si-rich and (c) O-poor, Si-rich (c) conditions

We then focused on identifying hydrogen positions that would be consistent with the observed frequency shifts of 2 kHz and 7 kHz resulting from interaction with a spin-1/2 nucleus. From the two-body magnetic dipole interaction Hamiltonian, we calculate the frequency shift as

$$A = \frac{1}{2} \frac{\mu_0 \mu_N^2 g_1 g_2}{4\pi r^3} (1 - 3\cos^2\theta) \quad (33)$$

μ_N is the nuclear magneton, μ_0 is the magnetic constant, g_1 and g_2 are the nuclear spin g -factors, r is the distance between the two nuclei, and θ is the angle between the bond and the magnetic field. The maximum shift is obtained when the magnetic field is aligned with the bond direction.

First, we examined whether interstitial or substitutional hydrogen interacting with host atoms (Y or Si) could be responsible for the observed frequency shifts. The bottleneck, caused by the small nuclear spin g -factor of Y, is identifying a location where H can be close enough to an Y atom to yield a 2 kHz frequency shift. After examining many configurations, we found that the shortest H–Y distance is found in H_0 in a +1 charge state (H_0^+) (Fig. S8a). The corresponding H–Si distance is 1.5 Å. The H–Si distance of 1.5 Å can lead to a 7 KHz frequency shift; but even the short H–Y distance (2.4 Å) leads to a frequency shift of only 0.4 kHz. Hydrogen interacting with Y and Si atoms thus cannot explain the observed frequency shifts.

We therefore consider interactions of hydrogen with unintentional impurities, for which Cd and P are prime candidates. After extensive exploration of possible configurations, we propose the following two defect complexes that yield magnetic coupling strengths in the vicinity of observed values: $\text{Cd}_{\text{Y}}\text{-H}_0$ (Fig. S8b) and $\text{P}_0\text{-H}_\text{I}$ (Fig. S8c).

$\text{Cd}_{\text{Y}}\text{-H}_0$ (Fig. S8b) with a Y_{II} atom substituted by a Cd atom and an O_{IV} atom substituted by a H atom is stable in the neutral charge state over a wide range of the Fermi level. The formation energy is 2.12 eV under (O-rich, Si-rich) conditions, and 3.05 eV under (O-poor, Si-rich) conditions. The binding energy, calculated as $E_{\text{bind}} = E^f(\text{Cd}_{\text{Y}_{\text{II}}}) + E^f(\text{H}_{\text{O}_{\text{IV}}}) - E^f(\text{Cd}_{\text{Y}}\text{-H}_0)$ is positive (0.76 eV), which indicates that the defect complex is more stable than spatially separated $\text{Cd}_{\text{Y}_{\text{II}}}^-$ and H_0^+ . The H–Cd distance is 2.4 Å and the H–Si distance is 1.5 Å. The largest frequency shifts that these distances allow, assuming that the

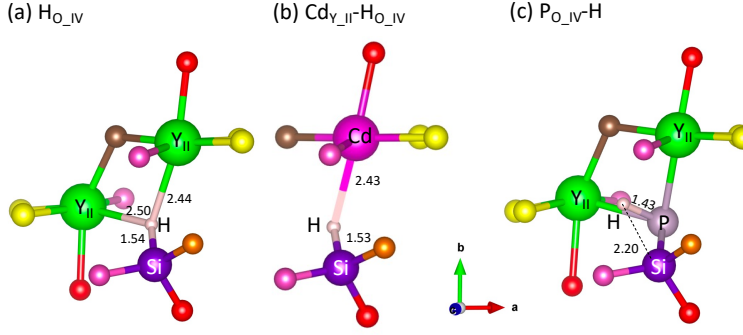


FIG. S8: Structures of defect complexes (a) $\text{H}_{\text{O}_{\text{IV}}}$ at $q = +1$, (b) $\text{Cd}_{\text{Y}}\text{-H}_{\text{O}_{\text{IV}}}$, and (c) $\text{P}_{\text{O}}\text{-H}_{\text{i}}$.

field is aligned with the bonds, is 2 kHz for $\text{H-}^{111}\text{Cd}$ coupling and 7 kHz for $\text{H-}^{29}\text{Si}$ coupling. However, the Cd-H-Si bond angle of 120° implies that the magnetic field cannot be simultaneously aligned with each bond, yielding smaller interaction strengths for a given field direction. For instance, if we assume that the angle between the magnetic field and the H-Cd bond is 35° and 155° for the H-Si bond, then the H-Cd coupling is 1.7 kHz and the H-Si coupling is 4.9 kHz.

$\text{P}_{\text{O}}\text{-H}_{\text{i}}$ (Fig. S8c) is also stable in the neutral charge state over a wide range of the Fermi level. The formation energy is 2.45 eV under (O-poor, Si-rich) conditions. The defect complex is also more stable than separated $\text{P}_{\text{O}_{\text{IV}}}^-$ and H_{i}^+ , with $E_{\text{bind}} = 0.85$ eV. The H-Si distance is 2.2 Å, leading to a maximum coupling strength of 2 kHz when the field is aligned with the bond. The H-P distance is 1.4 Å and due to the g -factor of P being much larger than that of Si, this leads to a maximum coupling strength of 17 kHz. Since the P-H-Si angle is 69° , taking the alignment of the magnetic field into account similarly lowers H-P coupling. For instance, assuming that the angle between the magnetic field and bond is 10° for the H-Si bond and 79° for the H-P bond, the H-Si coupling is still 2 kHz but the H-P coupling is 7 kHz. Therefore, a field direction exists such that the $\text{P}_{\text{O}}\text{-H}_{\text{i}}$ complex yields the coupling strengths observed in the experiment. However, the required magnetic field direction does not match the field direction set in the experiment within error. At our field direction, we expect this complex to yield an H-Si coupling of 0.5 kHz and H-P coupling of 7.5 kHz.

We note that hydrogen can often occupy several inequivalent positions in the vicinity of the impurity with similar energies; some of these positions could lead to alignments or distances that yield coupling strengths with even closer agreement. Our calculations for the $\text{Cd}_{\text{Y}}\text{-H}_{\text{O}}$ and $\text{P}_{\text{O}}\text{-H}_{\text{i}}$ complexes show that the observed frequency shifts can be plausibly due to interactions within such defect complexes.

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