

Engineering Quantum Resources for Quantum Networking using Single Rare-Earth Ions inside Crystals

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ABSTRACT

Quantum networks are foundational components of quantum technology, enabling transformative applications in secure communication, distributed quantum computation, and enhanced sensing. Rare-earth ions in solid-state hosts represent a leading platform for building such networks due to their exceptional optical and spin coherence properties. This thesis details the experimental realization of a quantum network node using single $^{171}\text{Yb}^{3+}$ ions in YVO_4 coupled to nanophotonic crystal cavities. We demonstrate the fundamental building blocks of quantum networks and develop multiple advanced capabilities, including multiplexing, protected nuclear spin storage, and high-dimensional qudit control, to expand the platform’s power and versatility.

Using this platform, we demonstrate heralded remote entanglement between two physically separate devices. A key innovation is a novel entanglement distribution protocol that employs real-time feedforward to cancel spectral diffusion on timescales slower than a single experiment by rephasing the optical transition based on photon arrival time. We also apply real-time phase compensations to entangle ^{171}Yb ions with different optical frequencies. By combining this novel protocol with multiple spectrally distinguishable ions, we demonstrate heralding of a three-ion W state and implement multiplexed remote entanglement. This multiplexing approach increases the entanglement rate by nearly a factor of two, showcasing a scalable pathway to mitigate network overhead.

Beyond establishing remote entanglement, we explore the local nuclear spin environment of ^{171}Yb as an integrated quantum resource. We harness the four symmetrically located ^{51}V nuclear spins to generate multi-qubit Greenberger–Horne–Zeilinger states. Critically, we identify and experimentally verify a decoherence-protected subspace within these states that exhibits insensitivity to common-mode magnetic field noise. By developing a sequence to transfer quantum information into this protected subspace, we establish the ^{51}V nuclear ensemble as an integrated, noise-resilient quantum memory.

To further expand the platform’s capabilities, we demonstrate coherent control over the four-level ground state of the ^{171}Yb ion, operating it as a qudit. Through development of a new device architecture that enables microwave driving of all transitions and comprehensive characterization of their coherence properties, this

work establishes the foundation for higher-dimensional quantum communication protocols that offer significant advantages in network capacity and efficiency.

Collectively, these results establish the $^{171}\text{Yb}:\text{YVO}_4$ system as a uniquely versatile platform and demonstrate the feasibility of building scalable quantum networks using single rare-earth ions in crystals.

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TABLE OF CONTENTS

Acknowledgements	iii
Abstract	v
Published Content and Contributions	vii
Table of Contents	viii
List of Illustrations	x
List of Tables	xxvi
Chapter I: Introduction	1
1.1 Structure of the Thesis	1
1.2 Quantum Networks and Quantum Repeaters	2
1.3 Different Experimental Platforms for Quantum Networking	5
1.4 Rare-Earth Ions for Quantum Networking	6
Chapter II: $^{171}\text{Yb}:\text{YVO}_4$ as a Quantum Network Node	11
2.1 Energy Level Structure and System Hamiltonian	11
2.2 Device Platform	13
2.3 Single Ion Properties	22
2.4 Nuclear Spin Coupling	30
2.5 Summary	38
Chapter III: Multiplexed Quantum Networking using $^{171}\text{Yb}:\text{YVO}_4$	41
3.1 Remote Entanglement Protocol	41
3.2 Experimental Setup	45
3.3 Ion Properties	51
3.4 Bipartite Entanglement Herdaling	53
3.5 Tripartite Entanglement Herdaling	64
3.6 Multiplexed Remote Entanglement	73
3.7 Outlook	77
Chapter IV: Creating Decoherence-Protected Greenberger–Horne–Zeilinger State using Nuclear Spins	79
4.1 Central Spin Qubits and Nuclear Spin Controls	80
4.2 Yb-V GHZ State and ^{51}V Number Sensing	83
4.3 Sensing the Decoherence Protected Subspace	88
4.4 V GHZ State Storage and Applications	93
4.5 Simulation of GHZ States	99
4.6 Summary and Outlook	101
Chapter V: $^{171}\text{Yb}:\text{YVO}_4$ as a Qudit System for Quantum Networking	103
5.1 System Hamiltonian and Device Requirements	104
5.2 Experimental Setup	105
5.3 Qudit Characterization and Coherence	106
5.4 Nuclear Spin Control	110
5.5 Outlook	111

Chapter VI: Conclusion and Outlook	113
6.1 Nuclear Spin Storage Enhancement	113
6.2 Improving Entanglement Rate and Fidelity	114
6.3 Long-Distance Networking Capabilities	114
6.4 Future Research Directions	114
Bibliography	116
Appendix A: Hybrid Quantum Photonic Interface for $^{171}\text{Yb}^{3+}:\text{YVO}_4$	132
A.1 Device Design and Fabrication	133
A.2 Detection of Single Yb Ions in YVO_4	135
A.3 Coherent Optical Control of a Single ^{171}Yb Ion	136
A.4 Conclusion and Outlook	138

LIST OF ILLUSTRATIONS

<i>Number</i>	<i>Page</i>
1.1 Schematic diagram of quantum repeaters. Left: direct entanglement generation between two end nodes is subject to exponential photon loss. Right: A quantum repeater chain divides the distance into shorter segments. Entanglement is first generated over these elementary links and then extended via entanglement swapping at the intermediate nodes.	4
2.1 Yb environment and energy level structure. (a) Schematic illustration of ^{171}Yb embedded in YVO_4 . The ^{171}Yb ion (yellow, center) is depicted as a central spin, surrounded by the dominant nuclear spin contributors (^{51}V nuclei, blue). (b) Energy level diagram of ^{171}Yb in YVO_4 . Red arrows indicate optical transitions along the crystal c axis, while blue arrows represent transitions along the a axis. Dotted gray lines mark forbidden transitions. The figure also highlights microwave transitions along the crystal c axis: f_g (ground state) at approximately 675 MHz and f_e (excited state) at approximately 3.37 GHz.	13
2.2 YVO_4 boule and coplanar waveguides. (a) YVO_4 boule grown by Gamdan Optics. (b) Gold coplanar waveguide fabricated on a YVO_4 chip, imaged under an optical microscope.	16
2.3 Design and fabrication of photonic crystal cavities. (a) Simulated electric field profile of the localized cavity mode in a one-dimensional photonic crystal nanobeam; the color scale indicates normalized electric field strength. (b) SEM image of a fabricated nanophotonic crystal cavity milled in YVO_4 . (c) Photo of the assembled device, showing the chip mounted on a copper stub and wire-bonded to a PCB for measurement. (d) Experimental reflection spectrum of the fabricated photonic crystal cavity, exhibiting a quality factor (Q) of approximately 10,000 and an input coupling ratio $\kappa_{\text{in}}/\kappa = 0.12$	18
2.4 Device setups inside a ^3He cryostat. Two device setups (“Alice” and “Bob”) are shown. Different components are labelled in the figure. .	20

2.5	Optical and microwave experimental setup. Laser beams are combined and directed to the photonic crystal cavity. Light emitted from the device is collected through a fiber coupler and detected with a superconducting nanowire single-photon detector (SNSPD). Microwave and RF signals for coherent spin and nuclear control are generated by an arbitrary waveform generator (AWG) and signal generator, respectively, then amplified, combined, and delivered to the device. All devices and the SNSPD are housed inside the ^3He cryostat for cryogenic operation.	21
2.6	Optical characterization of single ^{171}Yb ions in YVO_4 . (a) PL spectrum near the A transition of ^{171}Yb . Peaks corresponding to individual ions are observed; subsequent measurements refer to the highlighted ion. (b) Measured optical lifetime of the selected ion, showing Purcell-enhanced emission with a lifetime of $2.167 \pm 0.012 \mu\text{s}$. (c) Cyclicity of the same ion, measured to be 0.9969 ± 0.0003 . (d) Second-order correlation $g^{(2)}(\tau)$ for the indicated ion, with $g^{(2)}(0) = 0.010 \pm 0.002$, confirming single-photon emission. (e) Optical pump-probe measurement on the selected ion, revealing the F transition resonance.	24
2.7	Coherence and optical control of a single ^{171}Yb ion in YVO_4 . (a) Optical Rabi oscillations of a single ^{171}Yb ion; the solid line is a simulation based on the ion's optical Ramsey coherence time and measured lifetime. (b) Optical Ramsey interference measurement; the solid line is a fit assuming Gaussian decay, yielding a coherence time of $309 \pm 17 \text{ ns}$. (c) Optical spin echo measurement; the solid line is an exponential fit, yielding a coherence time of $3.66 \pm 0.12 \mu\text{s}$. Insets depict the pulse sequences used in each measurement.	25
2.8	Long-term spectral diffusion measurement of a single ^{171}Yb ion, tracked over a 14-hour period.	26
2.9	Pump-probe measurement of the F transition. The magnetic field is varied to examine the field dependence of the frequency splitting, revealing an avoided crossing.	27

2.10	Spin coherence and control of a single ^{171}Yb ion in YVO_4 . (a) Spin Rabi oscillations; the solid line is a cosine fit. (b) Spin Ramsey coherence measurement at a detuning of 0.1 MHz, with a fit including vanadium nuclear spins and echo decay. (c) Spin echo coherence, with a Gaussian fit yielding a time of $44.5 \pm 0.3 \mu\text{s}$. (d) XY8 dynamical decoupling measurement, optimized at a $5.8 \mu\text{s}$ wait time, and fit to an exponential decay with $16.8 \pm 0.8 \text{ ms}$ coherence. (e) T_1 relaxation measurement, fit to an exponential with a timescale of $53 \pm 3 \text{ ms}$	28
2.11	Randomized benchmarking of single-qubit control of ^{171}Yb . An average Clifford gate fidelity of 0.99975 ± 0.00002 is achieved.	29
2.12	Statistical properties of single ^{171}Yb ions measured across two devices. (a) F transition splitting as a function of A transition frequency. Ions with greater optical frequency offsets from the mean tend to exhibit larger F transition splittings. (b) Qubit transition frequency (f_g) as a function of A transition frequency, showing an inverse correlation.	30
2.13	Energy level structure of ^{51}V . ^{51}V has four doubly degenerate levels under zero magnetic field.	31
2.14	Details of the ZenPol pulse sequence. (a) The sequence consists of a series of π and $\pi/2$ pulses applied to the ^{171}Yb spin, combined with a square-wave RF field. This design dynamically decouples the ^{171}Yb central spin while generating a spin-preserving interaction with the ^{51}V nuclear spin ensemble. (b) Fourier spectra of the modulation functions $f_{x,y}^{\text{OH}}(t)$ (red) and $f_{x,y}^{\text{RF}}(t)$ (blue) highlighting the resonance conditions.	34
2.15	ZenPol spectroscopy. Data shown for 30 repetitions of the sequence with $B^{\text{RF}} = 0.6 \text{ G}$. Resonance condition is $1/2\tau = \omega/2\pi k$. Odd- k resonances (RF-induced) and even- k resonances (Overhauser-induced) are visible; red and blue denote measurements with and without RF field. Each resonance splits into two peaks, corresponding to register and bath ^{51}V spins. The inset compares spectra before (red) and after (purple) register polarization.	36

- 2.16 Magnon oscillations between ^{171}Yb and ^{51}V . (a) Coherent exchange (flip-flop) between ^{171}Yb and the ^{51}V register with all nuclear spins polarized. The oscillation frequency depends on the number of register spins and the RF drive strength (see inset). The red curve is measured with ^{171}Yb initialized into $|1\rangle_g$ and the blue curve is measured with ^{171}Yb initialized into $|0\rangle_g$. (b) ZenPol swap between ^{171}Yb and a single excited ^{51}V spin in the ω_c manifold. Reduced frequency confirms collective $\sqrt{N}$ scaling for four register spins. . . . 37
- 2.17 Coherence of ^{51}V storage. (a) Ramsey coherence time of ^{51}V storage, limited by Overhauser-field-induced ^{171}Yb dipole fluctuations ($T_2^* = 58 \pm 4 \mu\text{s}$). (b) Extended coherence using motional narrowing pulses on ^{171}Yb ($T_2 = 225 \pm 9 \mu\text{s}$). (c) Further extension with ^{51}V dynamical decoupling ($T_2 = 760 \pm 14 \mu\text{s}$). 38
- 2.18 Lifetime of ^{51}V states. (a) Measured lifetimes of $|W_v\rangle$ for three different decoupling protocols (as in Figure 2.17), indicating that $|W_v\rangle$ decay is coherence-limited. (b) Population lifetime (T_1) of the $|0_v\rangle$ state, measured to be $0.54 \pm 0.08 \text{ s}$ 39
- 3.1 Gate sequence for the entanglement distribution protocol. (Left) Energy level diagrams for two emitters with detuned optical transition frequencies ($\Delta\omega_0$). (Right) Schematic of the control sequence. The protocol begins with optical path phase stabilization ($Z_{\Delta\Phi}$), followed by the single-photon heralding process. Upon photon detection at time t_0 , dynamic rephasing and phase compensation sequences including spin and optical π pulses are applied to cancel phase errors from spectral diffusion and emitter frequency differences. 45
- 3.2 Experimental setup for remote entanglement distribution. (a) Optical setup: laser beams are routed to two independent $^{171}\text{Yb}:\text{YVO}_4$ devices, and emitted photons are collected and routed to various detection paths. (b) Detection configurations: Setup 1 is used for single-ion measurements, Setup 2 for bipartite entanglement heralding via interference, and Setup 3 for experiments involving more than two ions. (c) Three double-pass AOM setups used for controlling multiple ions within the inhomogeneous broadening range. (d) Bichromatic beam delivery setup for simultaneously addressing two ions. (e) Microwave control electronics for ground-state and excited-state spin manipulation. 46

3.3	Phase stabilization between two optical paths. (a) Schematic of the experimental setup for phase stabilization and probing. A stabilization pulse is sent, and the relative phase shift is detected by comparing the two paths. (b) Comparison of the Fourier-transformed phase noise of the probe pulse with (yellow) and without (blue) active stabilization. (c) Phase stability without feedback, characterized by the RMS phase difference over varying delays. The $1/e$ correlation time is approximately $282 \mu\text{s}$. (d) Phase histogram of the stabilized probe, showing a Gaussian distribution with a standard deviation of $2\pi \times 0.037$ radians. (e) Interference contrast between probe pulses detected via SNSPD, confirming high phase coherence (contrast = 0.944).	49
3.4	Photoluminescence spectra of the two devices. The ^{171}Yb ions used in the experiments are indicated by color peaks.	50
3.5	Detailed entanglement heralding sequence. (a) Flowchart of the entanglement protocol: ^{171}Yb ions are first initialized into the qubit manifold, before which the heralding sequence is executed. Detection of a herald photon triggers dynamic rephasing and state readout. (b) Initialization into the qubit manifold using optical pumping on the F_1 and F_2 transitions. (c) Entanglement heralding sequence, including qubit initialization, path-phase stabilization, and single-photon detection. (d) Upon photon detection, dynamic rephasing on the optical transition and feedforward phase compensation, dependent on the photon emission time and ion frequency difference, are applied in real time.	55
3.6	Readout sequence following entanglement heralding. Due to finite detection efficiency, the readout is probabilistic: outcome assignments are made according to the logic table provided, and cases not meeting the criteria are discarded.	56
3.7	X-basis parity oscillations measured at different stages of the entanglement protocol. Yellow: oscillations directly after heralding, limited by optical Ramsey coherence. Red: extended coherence following dynamic optical rephasing. Blue: phase-compensated parity showing suppressed oscillations, corresponding to a stationary Bell state. Dots represent experimental data; solid lines are simulations based on measured system parameters.	56

3.8	Quantum state tomography of the heralded entangled state. The state is reconstructed from measurements in all 9 two-qubit Pauli bases. Left: measured two-qubit correlations in the XX, YY, and ZZ bases. Right: absolute value of the reconstructed density matrix. The reconstructed state has a fidelity of 0.723 ± 0.007 and is obtained with a 500 ns heralding window, corresponding to an entanglement rate of 3.1 Hz.	57
3.9	Coherence of the heralded Bell state, measured via X-basis parity oscillations following application of an XY8 dynamical decoupling sequence on both ions. The exponential decay yields a Bell-state coherence time of 9.1 ± 0.4 ms, consistent with individual ion spin coherence limits.	60
3.10	Entanglement heralding rate and fidelity as a function of photon detection window size. Solid lines are exponential fits.	61
3.11	Two-photon entanglement heralding. (a) Experimental pulse sequence for the two-photon protocol. (b) Entanglement fidelity and heralding rate as functions of the photon detection window size; solid lines are exponential fits. (c) Absolute value of the reconstructed two-qubit density matrix measured with a 600 ns heralding window. $F = 0.81 \pm 0.02$, rate = 0.049 Hz	63
3.12	Individual ion control using AC Stark Shift. (a) Pulse sequence for individual ion control within a single device using differential ac Stark shifts. Optical pulses are applied during alternating periods of a qubit dynamical decoupling sequence to accumulate phase. (b) Measured differential phase shift between two ions as a function of optical pulse amplitude. The linear dependence demonstrates controllable individual Z rotations based on optical amplitudes.	66
3.13	Tripartite entanglement heralding sequence. (a) Experimental schematic of three-ion entanglement: two ions are housed within the same cavity and the third in a separate node. (b) Pulse sequence used to generate and herald W state via a single-photon detection, dynamic rephasing, and phase feedforward.	68

3.14	Parity measurements for the three-ion W state after heralding. (a) Experimental pulse sequence used to measure time-dependent X-basis parity following heralded three-ion entanglement. (b) Schematic illustration of coherence between different ion pairs within the W state. Each coherence term is extracted by tracing over the third ion. (c) Fourier-transformed spectra of the measured parity signals for all three ion pairs. The peaks indicate the coherent phase evolution frequencies corresponding to optical frequency differences between each ion pair.	69
3.15	Quantum state tomography of the heralded three-ion W state. The density matrix is reconstructed from 27 Pauli basis measurements using a 300 ns photon detection window. The resulting state has a fidelity of 0.592 ± 0.007 and an entanglement heralding rate of 2 Hz. .	70
3.16	Three-ion W state heralding rate and fidelity as a function of photon detection window size. Solid lines are exponential fits.	73
3.17	Details of the multiplexed entanglement heralding sequence. Top: flow chart of the experimental protocol. Sequential entanglement attempts are launched first on Ion 3/4, followed by Ion 1/2. Depending on which pair successfully emitted a heralded photon, the appropriate rephasing and phase correction sequences are conditionally applied. Bottom: subpanels highlight protocol elements distinct from standard bipartite heralding, including parallel ion initialization and sequential entanglement attempts.	75
3.18	Multiplexed remote entanglement heralding results. (a) Fidelity and heralding rate versus photon detection window size. Blue and purple curves correspond to standalone entanglement characterization of Pair 1 and Pair 2, respectively. Green data correspond to multiplexed entanglement results. At fixed detection window size, the entanglement rate is nearly doubled while fidelity remains close to the average of the two pairs. (b) Pauli X, Y, and Z measurements of the multiplexed entangled states, extracted with a 500 ns detection window.	77

- 3.19 Simulated population for all four ions under the multiplexed entanglement pulse sequence. Left: effect of the pulse sequence on Ion 3 and Ion 4 (Pair 2). Right: response of Ion 1 and Ion 2 (Pair 1), showing increased population in the excited manifold due to detuned qubit pulses, which contributes to the enhanced heralding rate. 78
- 4.1 Nuclear-spin control in central spin systems. (a) Schematic of a central spin (orange, ^{171}Yb) system with nuclear spins (blue, ^{51}V). The crystal structure is shown on the top left. Weak interactions between nuclear spins are depicted by blue dash lines. The central part of the lattice is enlarged and shown below the lattice schematic. A central spin interacts with and controls (orange arrow) the nuclear spins via hyperfine interactions, facilitating entanglement between nuclear spins (orange wavy lines). When nuclear spins are prepared in specific configurations, the nuclear spin states are protected from central spin noise (background waves). (b) The energy level structure shows the c transition of the ^{51}V nuclear spin, separated by 991 KHz. At zero magnetic field, ^{51}V nuclear spins are doubly degenerate, consisting of $|\pm m_I\rangle$ with opposite Zeeman sensitivities. 81
- 4.2 Table of highly entangled states and their sensitivity to common-mode magnetic field noise. The left column lists examples of the states. The middle column $S(|\psi\rangle)$ quantifies the common-noise sensitivity of the quantum state, normalized to that of a single-spin superposition. $S(|\psi\rangle) = 0$ indicates a protected state. The last column shows whether the superposition state is protected from the common noise field. States with zero noise sensitivity are robustly protected from common-mode noise. On the right, schematics of the dynamics of superposition states for non-decoherence-protected (top) and decoherence-protected (bottom) cases are shown on the Bloch sphere. 82

- 4.3 Controlled rotation gate sequence and experimental demonstration.
 (a) Gate sequence for implementing controlled rotation gates. ^{171}Yb π pulses are applied at the node of the RF oscillations, and the RF phase is flipped when a ^{171}Yb π pulse is applied. (b) Experimental results of demonstrating the controlled rotation gate. In the experiments, ^{171}Yb is initialized in different states, and ^{51}V is measured in the X basis after driving. The results show that the driving phase of ^{51}V depends on the initial state of ^{171}Yb . Experimental data, represented by blue and red dots, are fitted by oscillatory functions with gaussian decays (solid lines). 84
- 4.4 Yb-V GHZ state generation. The experimental results and sequence for Yb-V GHZ state generation utilizing $\pi/2$ controlled rotation gates. During the parity wait period, the state accumulates phase at $\Delta N = N_1 - N_0$ times ω_c . Here, N_0 and N_1 represent the number of Vanadium nuclear spins that are initially in $|\downarrow\rangle$ or $|\uparrow\rangle$ (bottom left), respectively. The sequence concludes with a reverse sequence to map the phase of the GHZ states to ^{171}Yb and optical readouts are performed on ^{171}Yb . The top left panel illustrates the measured GHZ state parity oscillations as the wait time is varied between the Yb-V GHZ preparation and readout sequence. 85
- 4.5 Fourier transform of the parity oscillations shown in Figure 4.4, revealing peaks at integer multiples of the Vanadium transition frequency, which arise from different initial states. 85
- 4.6 Vanadium spin number sensing. ^{51}V spin number sensing using the Yb-V GHZ state generation sequence. Phase accumulation depends on the initial ^{51}V state (left) and wait time (t_1), with the accumulated phase proportional to ΔN (middle). The blue curve shows the parity oscillations of the mean of the two initial states on the left. The purple curve represents the parity oscillation with spins initialized into $|\downarrow\downarrow\downarrow\downarrow\rangle$, while the red curve represents the parity oscillation with spins initialized into $|\uparrow\downarrow\downarrow\downarrow\rangle$. Specific harmonics can be selected by reading out ^{171}Yb at different wait times (right). 86

- 4.7 Experimental result of harmonic selection through post-selection. The left panels show the parity oscillations after post-election. The insets show the main contributing states, which is determined by different ^{51}V initialization maximizing the contribution of selected harmonics. The post-selection times (t_1) of different harmonics are shown at the middle top for each harmonics. The right panels show the corresponding Fourier spectrum. ^{51}V nuclear spins are initialized differently for different harmonic measurements. For example, a single spin is initialized into $|\uparrow\rangle$ for $1\omega_c$ post-selection (Section 4.2.1). 87
- 4.8 Two-stage ^{51}V spin number selection. (a) Experimental sequence for two-stage ^{51}V spin number selection. The time interval t_1 is set to $1/4/\omega_c$ to eliminate the $2\omega_c$ contribution, and t_2 is set to $1/2/\omega_c$ to suppress the $1\omega_c$ and $3\omega_c$ contributions. (b) Experimental results of demonstrating spin number selection at different stages. The contrast of the parity oscillations is enhanced with the implementation of two-stage spin number selection. 88
- 4.9 Experimental sequence for selecting different ^{51}V number states. The detailed sequence for GHZ block is shown in Figure 4.4. 88
- 4.10 Experimental sequence for sensing the decoherence-protected subspace. This sequence is used to generate Figure 4.11 and 4.12. The right-hand side illustrates the sequences used to generate DC fields and artificial noise on ^{51}V 90
- 4.11 Sensing of the decoherence protected subspace of ^{51}V . Experimental results of ^{51}V GHZ states under different DC magnetic fields, applied through Yb using square wave RF pulses interleaved with Yb π pulses. Each vertical axis data is obtained by Fourier transforming the time-domain measurement results. The $4\omega_c$ resonance exhibits field-dependent splitting. The orange lines are fitted lines for the split peaks. The magnetic field-dependent and independent peaks originate from the non-decoherence-protected and decoherence-protected subspaces, respectively. The states that contribute to each specific peak are shown on the right of the plot. 90

- 4.12 Decoherence-protected subspace of GHZ states. (a) Testing the decoherence-protected subspace by applying arbitrary common noise fields (σ) through ^{171}Yb . Here, we apply common noise fields through ^{171}Yb . The noise field is sampled using a Gaussian distribution with different standard deviations for different curves. Nuclear spins are fully initialized into $7/2$ state and the decay is fitted by Gaussian decays with different timescales and fixed amplitude ratios (solid lines), where the amplitude ratios are determined by the probability of being in specific states. The colored dots are the experimental data. The pink and blue regions indicate non-decoherence protected decay and decoherence-protected decay, respectively. With increasing noise field, the coherence decay of the decoherence-protected subspace is unaffected, but the coherence decay of the non-decoherence-protected subspace becomes faster. (b) The fitted short and long timescale decays for different noise fields. The fitted timescale is normalized to the long timescale ($164 \mu\text{s}$). The longer decay is fitted with a constant line. The short decay is fitted by considering a fixed gaussian decay from the long decay and a gaussian decay with a timescale proportional to $\frac{1}{\sigma}$ 91
- 4.13 Applications of decoherence-protected nuclear spin storage in a multiplexed quantum network node. (a) Experimental demonstration of swapping information from ^{171}Yb $|0\rangle$ and $|1\rangle$ to ^{51}V $|\downarrow\downarrow\downarrow\downarrow\rangle$ and $|\uparrow\uparrow\uparrow\uparrow\rangle$ with a schematic shown at the top. Experimental results of swapping the state into ^{51}V and swapping back to ^{171}Yb are shown at the bottom. ^{171}Yb is prepared in six cardinal Bloch states and the retrieved states are characterized through quantum state tomography. (b) Toolbox and applications of a decoherence-protected nuclear spin storage in a multiplexed quantum network node. The toolbox includes multiplexed remote entanglement distribution and decoherence-protected storage. With these capabilities, teleported gates and entanglement purification can be performed. 95

- 4.14 Storage time in ^{51}V $|\downarrow\downarrow\downarrow\downarrow\rangle$ and $|\uparrow\uparrow\uparrow\uparrow\rangle$ bases. (a) Experimental sequence for measuring storage time. The GHZ swap sequence, as described in Figure 4.15, is used to swap the ^{171}Yb state onto ^{51}V , which is then retrieved after certain waiting period. (b) Storage time for the $|+\rangle$ state. The Y-axis shows the $4\omega_c$ Fourier transformed amplitude of the parity oscillations. Experimental data, represented by purple dots, are fitted by Gaussian decay curves (solid lines). The short decay has a timescale of $27\pm 4 \mu\text{s}$, and the long decay is $200\pm 40 \mu\text{s}$. (c) Storage time for the $|0\rangle$ state. The decay timescale corresponds to the ^{51}V T_1 time 0.5 second, which is much longer than the probing period. 96
- 4.15 Experimental sequence for performing swap between $|0_g\rangle/|1_g\rangle$ and $|\downarrow\downarrow\downarrow\downarrow\rangle/|\uparrow\uparrow\uparrow\uparrow\rangle$ 96
- 4.16 Protocol for deterministic initialization into the decoherence-protected subspace. A ^{51}V sensing sequence is first applied with a DC magnetic field to sense the state of the nuclear spins. If ^{51}V is not in the decoherence-protected subspace, ZenPol is applied to flip either one or two ^{51}V spins depending on the sensed state. After flipping ^{51}V , controlled rotation gates on the a and b transitions of ^{51}V are applied to prepare the flipped ^{51}V to $|1/2\rangle$. With an applied DC field, controlled rotation gate can flip the sign of the $|1/2\rangle$ state. Then ZenPol or controlled rotation gates can be applied to polarize the spins to $|7/2\rangle$. Depending on the initial state, this leads to preparation of a decoherence-protected state, or another round of ^{51}V sensing can be applied. 97

4.17	Applications of decoherence-protected nuclear spin storage in a multiplexed quantum network node. (a) Protocol for entanglement purification in a multiplexed quantum network node. Orange spins represent optically addressable central spins, and the blue spins indicate decoherence-protected nuclear spin storage. First, multiplexed entanglement attempts are performed between nodes. Then, successfully entangled pairs are swapped to the decoherence-protected nuclear spin storage. Once two successful pairs of entanglement are established, entanglement purification is performed using teleported gates mediated by central spin qubits. Lastly, the purified entangled state can be used for further purification or other applications. Dashed lines between spins represent lower-fidelity entangled pairs. Solid lines between spins indicate purified or higher fidelity entangled pairs. (b) Simulations of how the multiplexing and nuclear spin storage improve entanglement purification. The figure illustrates the fidelity increase achieved through a single round of entanglement purification, plotted as a function of ion number. Simulations (colored dots) were conducted using different link efficiencies, defined as the ratio of entanglement rate to coherence decay rate. Solid lines are fitted as guided lines.	100
4.18	Simulation of Yb-V GHZ parity oscillations. (a) Comparison between experimental data (blue curve) and simulation results (red dots) for Yb-V GHZ state parity oscillations. (b) Comparison between experimental data (blue curve) and simulation results (red dots) for the Fourier spectrum of Yb-V GHZ state parity oscillations.	101
5.1	Device platform for four-level qudit operation. (a) Schematic showing crystal axes orientation relative to the device geometry. (b) SEM image of the fabricated photonic crystal cavity device.	105
5.2	Microwave driving setup for four-level qudit control. The heterodyne mixing system enables coherent driving of transitions between qubit and auxiliary manifolds.	106

5.3	Optical characterization of ^{171}Yb ions in 45° -cut devices. (a) Photoluminescence spectrum around the A transition showing individual ion peaks; the highlighted peak corresponds to the ion used for subsequent measurements. (b) Optical lifetime measurement revealing Purcell-enhanced emission with $\tau = 1.612 \pm 0.009 \mu\text{s}$. (c) Optical cyclicity measurement yielding $C = 0.99493 \pm 0.00007$	108
5.4	Microwave characterization of the four-level qudit system. (a) Energy level structure showing qubit and auxiliary manifolds. (b) ODMR spectrum identifying transitions between qubit and auxiliary states. (c) Coherent Rabi oscillations for all four qubit-auxiliary transitions. .	109
5.5	Coherence characterization of the auxiliary transition f_{aux} . (a) Ramsey dephasing measurements (blue points) with simulation including vanadium Overhauser noise (red line). (b) Spin echo decay (blue points) overlaid with an $13.2 \mu\text{s}$ exponential decay (red line). (c) XY8 dynamical decoupling extending coherence to $2.27 \pm 0.15 \text{ ms}$. (d) Spin lifetime measurement yielding $15.4 \pm 1.7 \text{ ms}$	110
5.6	^{51}V direct driving using auxiliary manifold. (a) Direct driving with varying drive time. (b) Rabi frequencies and decay constants of the Rabi oscillations.	111
A.1	The device schematic of hybrid device platform for coupling to $^{171}\text{Yb}:\text{YVO}_4$. The GaAs photonic crystal cavity and YVO_4 crystal are shown in gray and yellow, respectively. Yb ions are shown with orange arrows and are located inside YVO_4 . The crystal axes are shown next to the coordinate axes. The grating coupler (blue rectangle) is used for coupling light to a free-space setup. The photonic crystal cavity is indicated with a white rectangle.	132
A.2	Hybrid device simulations. (a), (b) Simulated E_y field of the fundamental TE mode in the XY and YZ planes, respectively. Note the evanescent decay of electric field inside YVO_4	134
A.3	Images of fabricated GaAs devices. (a) Scanning electron microscope image of the suspended GaAs photonic crystal cavity before transferring onto YVO_4 . (b) Optical image of the transferred device. The darker yellow substrate under the GaAs photonic crystal cavities is YVO_4	134

- A.4 Cavity reflection spectrum after transferring onto YVO_4 . Data are shown in green dots and overlaid with a fitted black line, which gives a quality factor of 5300. 135
- A.5 Experimental setup and detection of single Yb ions. (a) Experimental setup. Optical pulses from two frequency locked lasers, labeled A and C according to the driven optical transition (Figure A.5a), are sent to the device located in a ^3He fridge at 0.5 K. Emitted photons are routed to an SNSPD in the same cryostat. Laser pulses are shaped using two double-pass acousto-optic modulators, and optical pulses are removed from the detection through time filtering and an acousto-optic modulator shutter setup. (b) Resonant photoluminescence spectrum. The large emission peak at 304505 GHz corresponds to Yb isotopes with no nuclear spin. Isolated peaks inside the spectrum are mostly single Yb ions, the arrows indicate ions measured in subsequent experiments. Data was taken with 10 second integration time and 50 kHz repetition rate for each point. The fitted linewidth of red and blue ions are 27.3 MHz and 15.9 MHz, respectively. (c) Pulsed $g^{(2)}(t)$ measurement of the ion indicated with a blue arrow shows $g^{(2)}(0)=0.26 \pm 0.09$. (d) Lifetime measured through time-resolved photoluminescence of ions indicated by red ($4.2 \pm 0.1 \mu\text{s}$) and orange ($32 \pm 2 \mu\text{s}$) arrows are shown with corresponding colors. Experimental data are shown with dots and overlaid with fitted exponential decays. The fit to the orange curve also includes an additional contribution from background bulk ions due to its lower intensity. Bulk lifetime is shown with a black line. 137

A.6 Coherent optical control of a single ^{171}Yb ion. (a) Energy level structure of ^{171}Yb inside YVO_4 at zero magnetic field. Transition A (cavity enhanced) is shown in blue; transition C (non-enhanced) is shown in red. Cavity enhanced decay is shown with a dashed arrow. $|0\rangle_g, |1\rangle_g, |0\rangle_e, |1\rangle_e$ are magnetic field insensitive forming clock transitions. (b) Pump-probe measurement of the ^{171}Yb optical transitions. The pump frequency is varied around the C-transition with probe frequency fixed to the A-transition. In subsequent measurements, the ion is partially initialized by repeatedly pumping on this transition. The difference between the frequency noted in Figure A.6a and the measurement could be due to strain at the site of the ion. (c) Optical Rabi oscillation on the A-transition. (d) Optical Ramsey measurement on the A-transition gives $T_2^* = 69 \pm 15$ ns. (e) Optical echo measurement on the A-transition gives $T_2 = 330 \pm 50$ ns. 139

LIST OF TABLES

<i>Number</i>	<i>Page</i>
3.1 Optical cavity parameters for the two devices used in the experiments. Q is the quality factor, κ is the total energy decay rate of the cavity, and $\kappa_{\text{in}}/\kappa$ gives the ratio of the input coupling rate to the overall cavity linewidth.	50
3.2 Physical properties of four ions in the two devices.	52
3.3 Raw two-qubit counts in the 9 Pauli bases for Ion 1 and Ion 2 under a 500 ns heralding window. N_{ab} is the number of trials with Ion 1 in $ a\rangle$ and Ion 2 in $ b\rangle$	59

Chapter 1

INTRODUCTION

1.1 Structure of the Thesis

This thesis experimentally demonstrates the fundamental building blocks of quantum networks using ^{171}Yb doped in YVO_4 . Beyond establishing these foundational elements, we expand the platform's capabilities by exploiting the unique features inherent to our system.

Chapter 1 introduces the basic concepts of quantum networking and quantum repeaters, followed by an overview of rare-earth ion systems. The chapter concludes with a discussion of the advantages and current progress in utilizing rare-earth ion systems for quantum networking applications.

Chapter 2 provides a comprehensive introduction to the $^{171}\text{Yb}:\text{YVO}_4$ system, including the system Hamiltonian, experimental setup, optical and spin coherence properties of ^{171}Yb ions, and characterization of the nuclear spin environment.

Chapter 3 demonstrates remote entanglement distribution across two separate devices using $^{171}\text{Yb}:\text{YVO}_4$. Building upon bipartite entanglement distribution, we leverage multiple ions within the two nodes to achieve tripartite entanglement distribution and multiplexed remote entanglement distribution.

Chapter 4 exploits our ^{51}V nuclear spin ensemble to generate various Greenberger–Horne–Zeiling states. We further investigate the decoherence-protected subspace of these nuclear spin states, which constitute important resources for future quantum networking protocols.

Chapter 5 presents a newly designed photonic crystal cavity that enables coherent control of ^{171}Yb as a qudit system, establishing the foundation for future investigations of qudit quantum networking.

Chapter 6 summarizes the findings from the previous chapters and discusses future research directions.

Chapter A provides an additional discussion on hybrid photonic devices.

1.2 Quantum Networks and Quantum Repeaters

The theoretical foundations of quantum information and computing, first articulated by Paul Benioff, Richard Feynman, and Yuri Manin in the 1980s [1–3], revealed a new paradigm for information processing. This paradigm leverages uniquely quantum phenomena, such as superposition and entanglement, to unlock computational capabilities far beyond the reach of classical machines [4, 5]. In the decades since, remarkable experimental progress has been made across a diverse array of physical platforms, including superconducting qubits [6–9], trapped atoms and ions [10–13], silicon quantum dots [14–16], and photonic qubits [17, 18].

Within the expansive landscape of quantum information science, which encompasses quantum computation, sensing [19], and simulation [20], the field of quantum networking has emerged as a critical frontier [21, 22]. This thesis is dedicated to the central challenge of quantum networking: the distribution of quantum information and entanglement over long distances to connect disparate quantum devices.

The development of quantum networks is driven by a suite of transformative applications. These include secure communication protocols like Quantum Key Distribution (QKD) [23, 24], which offers security guaranteed by the laws of physics rather than computational complexity [25]; distributed quantum sensing, which can achieve measurement sensitivities beyond the standard quantum limit [26, 27]; distributed quantum computation [28, 29], which enables the connection of smaller quantum processors to form more powerful, modular computing systems; and blind quantum computation, where a client can securely delegate a computation to a remote quantum server without revealing their input, algorithm, or output [30–32]. By enabling quantum devices to operate together across arbitrary distances, these networks are essential for realizing the full potential of quantum technologies.

This introduction will first establish the fundamental principles and architectural requirements of long-distance quantum networking. It will then assess the leading experimental platforms in the context of these requirements, thereby motivating the specific focus of this thesis on rare-earth ions as a promising candidate for building the next generation of quantum networks.

1.2.1 Introduction to Quantum Repeaters

To distribute quantum information, one must select a transmission medium that minimizes interaction and loss. Optical photons are the ideal carriers due to their weak environmental coupling and low attenuation in optical fibers (approximately

0.2 dB/km at telecom wavelengths). However, even this minimal loss is exponential with distance, making the direct transmission of quantum states over continental scales infeasible.

In classical networks, this challenge is overcome by amplifying the signal at intermediate repeater stations. This approach is fundamentally forbidden in the quantum realm by the no-cloning theorem [33], which states that it is impossible to create an identical copy of an arbitrary, unknown quantum state. This prohibition necessitates a completely different strategy for long-distance quantum communication.

The solution is the quantum repeater [34–36]. The logic of the quantum repeater architecture unfolds as follows:

- **Divide and Conquer:** The long-distance channel is segmented into shorter, elementary links over which photon loss is manageable.
- **Elementary Entanglement:** Entanglement, typically a Bell state, is first generated between the adjacent nodes of each elementary link. With direct transmission, the success probability (P_{suc}) of establishing a Bell state between two nodes separated by distance L is fundamentally limited by photon loss (Figure 1.1):

$$P_{\text{suc}} \approx \frac{1}{2} e^{-L/L_{\text{att}}}, \quad (1.1)$$

where L_{att} is the fiber's attenuation length.

- **Entanglement Swapping:** Once entanglement is established on adjacent links, a protocol known as entanglement swapping is performed at the intermediate node [37]. This procedure effectively stitches the two shorter entangled links together, creating a single entangled pair that spans a longer distance without the constituent photons ever having to traverse that full distance.
- **Synchronization with Quantum Memory:** Since entanglement generation is probabilistic, waiting for all elementary links to succeed simultaneously would reintroduce an exponential scaling problem. This is solved by equipping each repeater node with a quantum memory. The memory can store a successfully generated entangled state on one link while the node continues to attempt entanglement generation on the adjacent link, thus serving as a critical synchronization resource.

- **Combating Noise with Purification:** Every operation, entanglement generation, storage, and swapping, is imperfect and introduces noise that degrades the fidelity of the entangled state. To counteract this, entanglement purification protocols can be employed [38], which distill a smaller number of high-fidelity pairs from a larger ensemble of noisy ones.

With a repeater chain of 2^{n-1} intermediate nodes (Figure 1.1), the success probability is no longer dominated by the exponential loss over the total distance L . Instead, it becomes:

$$P_{\text{suc}} \approx \frac{1}{2} (p_{\text{sw}})^{n-1} e^{-L/(2^n L_{\text{att}})}, \quad (1.2)$$

where p_{sw} is the success probability of the entanglement swapping operation. This architecture transforms the exponential dependence on total distance into a much more manageable polynomial scaling of resources, making long-distance quantum communication feasible.

The value of p_{sw} is highly dependent on the physical hardware used to implement the repeater node. For schemes that rely on interfering photons from separate sources using linear optics, the Bell-state measurement required for entanglement swapping is fundamentally probabilistic, limiting the success probability to $p_{\text{sw}} \leq \frac{1}{2}$. In contrast, matter-based platforms such as trapped ions or solid-state spins can perform entanglement swapping deterministically, allowing for a success probability that approaches unity $p_{\text{sw}} = 1$. This key difference in p_{sw} has profound implications for the overall efficiency and resource requirements of a quantum repeater chain. For interested readers, we refer to the following reviews [36, 39].

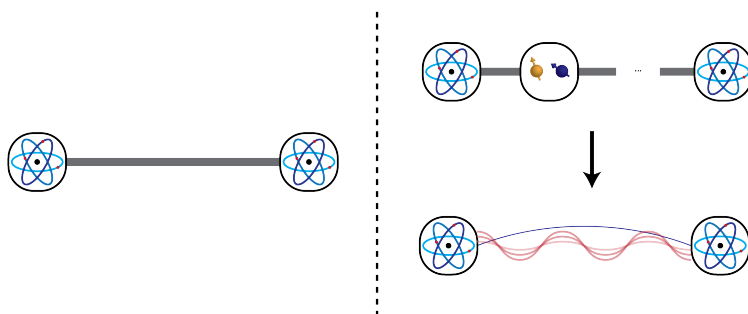


Figure 1.1: Schematic diagram of quantum repeaters. Left: direct entanglement generation between two end nodes is subject to exponential photon loss. Right: A quantum repeater chain divides the distance into shorter segments. Entanglement is first generated over these elementary links and then extended via entanglement swapping at the intermediate nodes.

1.2.2 Requirements of building a Quantum Network

The preceding discussion reveals a set of stringent requirements for the physical hardware at each network node. At its core, a node must function as a high-performance quantum interface, capable of faithfully mapping quantum information between a stationary matter qubit (for storage and processing) and a flying photonic qubit (for communication). This function imposes several key demands:

- **An Efficient Optical Interface:** The node must be able to generate high-fidelity spin-photon entanglement at a high rate. This is primarily determined by the photon collection efficiency, which is often enhanced using optical cavities.
- **High-Fidelity Qubit Control:** The matter qubits must support high-fidelity initialization, coherent manipulation (gates), and readout. These operations form the foundation for all network protocols.
- **Long-Lived Quantum Memory:** The node must possess a quantum memory with a coherence time long enough to store entangled states while entanglement is established across other segments of the network. This is essential for synchronization and scalability.

1.3 Different Experimental Platforms for Quantum Networking

With these requirements established, we can evaluate the leading experimental platforms for quantum networking. The field has matured beyond initial proof-of-principle experiments; the central challenge is now to engineer systems that simultaneously achieve high entanglement rates, high fidelities, and robust memory capabilities.

Trapped-ion platforms have long been the leader in entanglement fidelity and qubit coherence [40–42]. State-of-the-art demonstrations have achieved remote entanglement rates of 250 Hz and fidelities as high as 97%. The high-fidelity control, purity, and scalability of these systems have also enabled achievements such as spin-photon entanglement multiplexing with multiple ions [43–45]. However, the rate-limiting factor for high-rate entanglement remains the photon collection efficiency, which is constrained by the numerical aperture (NA) of the collection optics. Further integration with optical cavities is a promising route to improve this rate even further [43]

Trapped neutral atom platforms have also demonstrated remote entanglement distribution [46, 47]. Driven by recent progress in creating large, configurable neutral

atom arrays for quantum computation [11, 13, 48], there is growing interest in using these systems to build networks that connect quantum processors [49]. A key advantage is that neutral atoms are less sensitive to environmental electric field noise, which can facilitate better integration with cavities designed to enhance photon collection [50, 51].

Solid-state spins are leading contenders due to their potential for integration with nanodevices and the high degree of engineerability afforded by the variety of host crystals and qubit types [52]. Nitrogen-vacancy (NV) centers in diamond were one of the earliest platforms to demonstrate remote entanglement distribution [53, 54]. However, because NV centers lack inversion symmetry, their coherence properties can be degraded upon integration with nanophotonic devices [55]. Group-IV centers in diamond, such as the silicon-vacancy (SiV) and tin-vacancy (SnV), possess this inversion symmetry, making them much better candidates for cavity integration. SiV centers have been integrated with nanophotonic crystal cavities, demonstrating high cooperativities and, recently, remote entanglement distribution [56–58]. SnV centers have gained attention for their good optical and spin coherence and their ability to operate at higher temperatures [59–61]. Other promising solid-state systems include T and G centers in silicon, which benefit from mature fabrication technology [62–64]; defects in SiC, which have demonstrated long spin coherence times [65, 66]; quantum dots, which have shown remote entanglement despite more limited spin coherence [67]; and emitters in 2D materials, which have potential applications in quantum sensing [68].

Finally, microwave-to-optical transduction is an active area of research for connecting superconducting qubit devices, which operate in the microwave regime, to optical networks [69–71].

This summary reveals that while each platform has made remarkable progress, they all face fundamental trade-offs. This motivates the exploration of rare-earth ions, a platform whose unique properties are well-suited to address some of the challenges in quantum networking.

1.4 Rare-Earth Ions for Quantum Networking

1.4.1 Introduction to Rare-earth Ions Inside Crystals

Rare-earth ions (REIs) doped into crystals have a long history of diverse applications, including in laser technology [72], and are now central to emerging quantum technologies such as quantum memory [73, 74], microwave-to-optical transduction

[71, 75, 76], the study of many-body physics [77, 78], and quantum networking [79, 80].

The utility of REIs across these fields stems from their unique atomic structure. The 4f valence electrons are shielded by outer 5s and 5p orbitals, which effectively isolates them from environmental noise like lattice vibrations (phonons) and fluctuating electric fields, leading to high quantum efficiency. This natural protection gives rise to some of the longest optical and spin coherence times ever observed in a solid-state system. For example, $^{151}\text{Eu}^{3+}:\text{Y}_2\text{SiO}_5$ has demonstrated a spin coherence time of six hours [81], while $\text{Er}^{3+}:\text{Y}_2\text{SiO}_5$ has an optical coherence time exceeding 4 ms [82].

The characteristic optical transitions of REIs occur between electronic states within this 4f shell. For a free ion, these 4f-4f transitions are forbidden. However, when an REI is doped into a crystal, the surrounding crystal field breaks the symmetry, weakly allowing these transitions to occur. This crystal field also lifts the degeneracy of the free-ion energy levels, splitting them into a complex structure of crystal field levels, with selection rules that can be calculated using group theory [83, 84]. This interaction gives rise to the sharp, atom-like spectral lines characteristic of REIs. Notably, Erbium (Er^{3+}) possesses 4f-4f transitions at telecom wavelengths, making it ideal for integration with existing fiber-optic technology, while the ion central to this thesis, Ytterbium (Yb^{3+}), has its primary 4f-4f optical transition around 980 nm.

This exceptional coherence directly addresses the critical need for long-lived quantum memories in repeater architectures. Furthermore, REIs possess another key advantage: a narrow optical inhomogeneous broadening. While individual ions have extremely narrow (kHz-scale) optical transitions, the ensemble of ions in a crystal has a collective linewidth of hundreds of MHz to a few GHz [79, 85–87]. This means that hundreds of ions, each acting as an independent, spectrally-resolvable qubit, can be addressed within the bandwidth of a single photonic device. This enables frequency multiplexing, which can boost the entanglement rate from the fundamental limit of a single emitter (c/L) to $N \times c/L$, where N is the number of multiplexed ions. This provides a direct and scalable pathway to the high-throughput communication required for a functional quantum internet. Rare-earth ions can also be integrated in different crystals and have been synthesized in molecules for quantum applications [86, 88]. The homogeneous properties of rare-earth ions and the widely engineerable host environment make rare-earth ions among the most promising candidates for building quantum networks.

1.4.2 Detection of Single Rare-earth Ions

The same electronic shielding that grants REIs their superb coherence also makes their 4f-4f optical transitions very weak. This results in long radiative lifetimes (up to milliseconds), which severely limits the photon emission rate and makes the detection of single ions challenging.

The solution is to couple the ions to a high-quality optical cavity. Through the Purcell effect [89], a cavity with a high quality factor (Q) and a small mode volume (V) can dramatically enhance the spontaneous emission rate of an emitter by a factor $F_p \propto Q/V$. This enhancement shortens the optical lifetime, increases the photon emission rate, and channels the emitted photons efficiently into a single, collectible mode, thereby enabling an efficient spin-photon interface.

Significant experimental effort has been dedicated to engineering such cavities. Approaches include:

- **Directly Fabricated Cavities:** Photonic crystal cavities etched directly into the host material (e.g., YVO_4 , Si) offer the potential for the highest Q/V and strongest Purcell enhancement but can require challenging fabrication processes and thin film growth [90–94].
- **Hybrid Cavities:** These leverage mature silicon (or III-V) fabrication by placing a silicon photonic crystal cavity atop a rare-earth-doped crystal (e.g., CaWO_4 , YSO). This is a versatile approach but can only couple to ions near the crystal surface, where coherence may be degraded [79, 95–99].
- **Fabry-Perot Cavities:** These tunable cavities can be used with nanocrystals or bulk crystals, preserving the pristine coherence of deeply embedded ions, but typically have larger mode volumes and thus more modest Purcell enhancement [87, 100].

The diversity of these techniques reflects efforts to overcome the primary experimental hurdle of the platform, thereby unlocking the exceptional intrinsic quantum properties of rare-earth ions.

1.4.3 Advantages and Disadvantages of using Rare-earth Ions for Quantum Networking

We can now summarize the unique position of rare-earth ions for quantum networking applications:

- **Key Advantage (Memory):** The exceptionally long spin coherence times of REIs directly satisfy the requirement for robust, long-lived quantum memories, a cornerstone of any scalable repeater architecture.
- **Key Advantage (Bandwidth):** The narrow optical inhomogeneity provides a natural platform for high-bandwidth communication via frequency multiplexing, directly addressing the rate limitations that constrain other platforms.
- **Key Advantage (Resources):** The rich nuclear spin environment of many REI systems can be engineered to serve as additional multi-qubit registers for local processing or longer-term storage [101, 102].
- **Key Advantage (Multi-Qubit Processing):** Rare-earth ions can be doped into crystals at densities that allow multiple ions to be located in close proximity. This enables controlled dipole-dipole interactions, which are essential for performing multi-qubit gates and local quantum information processing within a network node [103, 104].
- **Key Advantage (Engineerability):** Due to their shielded transitions, REIs exhibit similar properties across a wide variety of host materials. This allows for extensive engineering of the host environment, from different crystals to synthesized molecules, to optimize for specific applications [86].
- **Central Challenge (Interface):** The intrinsically weak optical transitions necessitate the integration of sophisticated, high-Q/V optical cavities to achieve an efficient spin-photon interface, which represents the primary fabrication and engineering challenge.

1.4.4 Different Ways of Expanding the Capability of the Platform

This thesis confronts these challenges and exploits these advantages to advance the state-of-the-art in rare-earth-ion-based quantum networking. The following chapters present novel experimental contributions that expand the capabilities of this platform:

- **Chapter 3:** Having established the critical need to overcome rate limitations, this chapter presents a demonstration of multiplexed entanglement distribution using multiple, optically distinguishable ions in two remote devices, showcasing a viable path toward high-throughput quantum network.

- Chapter 4: To move beyond two-party entanglement, this chapter explores the rich nuclear spin environment of the host crystal, demonstrating a protocol to create decoherence-protected multipartite GHZ states, a key resource for quantum storage.
- Chapter 5: To further expand the information capacity of each network node, this chapter pushes beyond the qubit paradigm by demonstrating high-fidelity coherent control of the multi-level $^{171}\text{Yb}:\text{YVO}_4$ qudit system, opening a new frontier for high-dimensional quantum networking.

Chapter 2

$^{171}\text{Yb}:\text{YVO}_4$ AS A QUANTUM NETWORK NODE

The development of scalable quantum networks hinges on the creation of quantum nodes capable of distributing entanglement over long distances. A functional quantum network node must satisfy three fundamental requirements: (1) a high-fidelity quantum bit for local processing, (2) an efficient and coherent spin-photon interface for remote entanglement, and (3) access to a local, long-lived quantum memory for storing quantum states. Rare-earth ions (REIs) doped into solid-state hosts have emerged as a leading platform for this purpose, renowned for their exceptionally long optical and spin coherence times.

This chapter introduces the $^{171}\text{Yb}:\text{YVO}_4$ system as a promising candidate for realizing such a node. Its advantages stem from the simple electron and nuclear spin-1/2 structure of the ^{171}Yb isotope, which simplifies state manipulation, combined with the favorable properties of the yttrium orthovanadate (YVO_4) host crystal, such as high site symmetry and a rich nuclear spin environment that can be engineered into a quantum resource. We present a comprehensive experimental characterization of this platform, begin by detailing the energy level structure and spin Hamiltonian of a single $^{171}\text{Yb}^{3+}$ ion. We then describe the integration of these ions with nanophotonic crystal cavities, a crucial step to overcome weak optical transitions and enable efficient spin initialization, readout, and control. Following this, we present detailed measurements of the optical and spin coherence properties of a single ^{171}Yb ion, demonstrating state-of-the-art performance. Finally, we explore the surrounding nuclear spin environment and demonstrate how it can be transformed from a source of noise into a coherently controlled quantum memory. For broader context on the role of REIs in quantum networks, readers are referred to Chapter 1.

2.1 Energy Level Structure and System Hamiltonian

The ^{171}Yb isotope is doped as a trivalent ion (Yb^{3+}) into the YVO_4 host, ^{171}Yb ions substitute for yttrium at sites of D_{2d} symmetry (Figure 2.1(a)). This specific symmetry makes the ion's optical transitions insensitive to first-order DC Stark shifts [105], rendering it highly suitable for integration into nanophotonic devices where surface charges can create fluctuating electric fields. The YVO_4 host crystal has a zircon tetragonal symmetry (D_{4h}), with two equivalent a -axes and a unique c -axis,

which we defined as the z -axis throughout this work.

The crystal field splits the $4f^{13}$ electronic configuration into a ground ($^2F_{7/2}$) and an excited ($^2F_{5/2}$) multiplet, resulting in a lowest-energy Kramers doublet that can be described by an effective electron spin $S=1/2$ [84]. The interaction between nuclear and electron spin degrees of freedom defines the qubit's energy structure. For all experiments, we utilize the lowest crystal field levels of these multiplets, which are separated by an optical transition at approximately 984.5 nm [106]. The effective spin Hamiltonian for a single $^{171}\text{Yb}^{3+}$ ion is given by:

$$\hat{H}_{\text{Yb}} = \hat{\mathbf{S}} \cdot \mathbf{A} \cdot \hat{\mathbf{I}}_{\text{Yb}} + \mu_B \mathbf{B} \cdot \mathbf{g} \cdot \hat{\mathbf{S}} + \mu_N \mathbf{B} \cdot \mathbf{g}_I \cdot \hat{\mathbf{I}}_{\text{Yb}}, \quad (2.1)$$

where $\hat{\mathbf{S}}$ and $\hat{\mathbf{I}}_{\text{Yb}}$ are the electronic and nuclear spin-1/2 operators, respectively; $\mathbf{A}$ is the hyperfine interaction tensor; μ_B and μ_N are the Bohr and nuclear magnetons; $\mathbf{B}$ is the local magnetic field; and $\mathbf{g}$, $\mathbf{g}_I$ are the electronic and nuclear g -tensors. The nuclear Zeeman interaction is small and is therefore neglected. The values for the hyperfine and g -tensors in the ground and excited states, determined from previous experiments [101, 106], are:

$$\mathbf{A}_g = 2\pi \times \begin{bmatrix} 0.675 & 0 & 0 \\ 0 & 0.675 & 0 \\ 0 & 0 & -4.82 \end{bmatrix} \text{ GHz} \quad (2.2)$$

$$\mathbf{A}_e = 2\pi \times \begin{bmatrix} 3.37 & 0 & 0 \\ 0 & 3.37 & 0 \\ 0 & 0 & 4.86 \end{bmatrix} \text{ GHz} \quad (2.3)$$

$$\mathbf{g}_g = 2\pi \times \begin{bmatrix} 0.85 & 0 & 0 \\ 0 & 0.85 & 0 \\ 0 & 0 & -6.08 \end{bmatrix} \quad (2.4)$$

$$\mathbf{g}_e = 2\pi \times \begin{bmatrix} 1.7 & 0 & 0 \\ 0 & 1.7 & 0 \\ 0 & 0 & 2.51 \end{bmatrix}. \quad (2.5)$$

At zero magnetic field, the hyperfine interaction splits both the ground and excited state manifolds into three levels (Figure 2.1(b)). Each manifold contains two non-degenerate levels ($|0\rangle_g$, $|1\rangle_g$ for the ground state; $|0\rangle_e$, $|1\rangle_e$ for the excited state) and one degenerate level ($|\text{aux}\rangle_g$, $|\text{aux}\rangle_e$). The non-degenerate levels are first-order insensitive to magnetic field fluctuations, which gives them enhanced coherence

properties, making them ideal qubit states. We define our ground-state qubit levels as:

$$|0\rangle_g = (|\downarrow_{Yb}\uparrow_{Yb}\rangle - |\uparrow_{Yb}\downarrow_{Yb}\rangle)/\sqrt{2},$$

$$|1\rangle_g = (|\downarrow_{Yb}\uparrow_{Yb}\rangle + |\uparrow_{Yb}\downarrow_{Yb}\rangle)/\sqrt{2}$$

where $\uparrow_{Yb} / \downarrow_{Yb}$ denote the electronic spin states and $\uparrow_{Yb} / \downarrow_{Yb}$ denote the nuclear spin states of ^{171}Yb .

The optical transitions between these levels are governed by polarization selection rules, which can be derived from group theory [107, 108] and are illustrated in Figure 2.1(b). Transitions polarized along the crystal c-axis (red arrows) are distinct from those polarized along the a-axis (blue arrows). Forbidden transitions are shown in grey. The figure also indicates the microwave transitions within the ground (f_g) and excited (f_e) state manifolds that are used for coherent spin control. For further details on measurement techniques and parameter extraction, refer to [107].

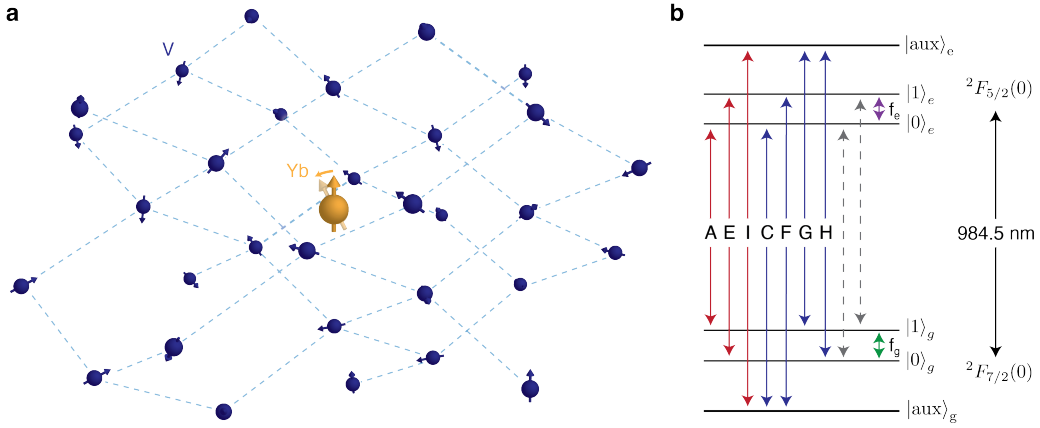


Figure 2.1: Yb environment and energy level structure. (a) Schematic illustration of ^{171}Yb embedded in YVO_4 . The ^{171}Yb ion (yellow, center) is depicted as a central spin, surrounded by the dominant nuclear spin contributors (^{51}V nuclei, blue). (b) Energy level diagram of ^{171}Yb in YVO_4 . Red arrows indicate optical transitions along the crystal c axis, while blue arrows represent transitions along the a axis. Dotted gray lines mark forbidden transitions. The figure also highlights microwave transitions along the crystal c axis: f_g (ground state) at approximately 675 MHz and f_e (excited state) at approximately 3.37 GHz.

2.2 Device Platform

The 4f–4f optical transitions of rare-earth ions are intrinsically weak, leading to slow spontaneous emission rates that hinder the efficient generation of single pho-

tons required for a spin-photon interface. To enable efficient optical initialization, readout, and photon interfacing, we employ nanophotonic crystal cavities. These structures offer high quality factors and small mode volumes, yielding strong Purcell enhancement (i.e., increased spontaneous emission rate into the cavity mode) and thus improved photon collection. The Purcell factor quantifies this enhancement and is proportional to Q/V_{mode} , where Q is the cavity quality factor and V_{mode} is the electromagnetic mode volume (see Section 2.2.2).

Below, we describe device architecture and fabrication steps enabling single-ion control and readout.

2.2.1 Device Design and Fabrication

YVO₄ chips used in this work are fabricated from undoped boules grown by Gamdan Optics with a residual concentration of 20 ppb ¹⁷¹Yb. The undoped boules are subsequently cut and polished by external suppliers (Figure 2.2a). To enable both optical and microwave control, we fabricate gold coplanar waveguides (CPWs) and nanophotonic crystal cavities directly on the YVO₄ chips. Since coherent qubit control requires a magnetic driving field aligned with the crystal’s *c*-axis, we use *c*-cut YVO₄ samples to ensure alignment between the CPW field and the polarization of the qubit transitions. The boules are diced into chips with dimensions of 3 mm × 3 mm × 0.5 mm. Several methods exist for fabricating nanophotonic crystal cavities on YVO₄ substrates. The most straightforward method would be to directly pattern a cavity on a thin-film YVO₄ substrate; however, due to the lack of commercially available high-quality YVO₄ thin films and the challenges associated with fabricating one, we adopt two alternative approaches in this thesis. The first is direct patterning of cavities into bulk YVO₄ using focused ion beam (FIB) milling, which is the technique used for the devices studied throughout this work. The second is a hybrid integration approach described in Appendix A.

2.2.1.1 Coplanar Waveguide Fabrication

Gold CPWs are fabricated using a standard electron beam lithography (EBL) process. Their geometry is optimized using COMSOL simulations to achieve a characteristic impedance of 50Ω, ensuring efficient delivery of microwave power to the ion and minimizing signal reflections that could otherwise interfere with coherent control. The central signal line is tapered from 300 μm (at the edge) down to 50 μm (in the center), while the gap between the signal line and ground plane tapers from

130 μm to 20 μm . An optical microscope image of a fabricated gold coplanar waveguide is shown in Figure 2.2b. The fabrication process is summarized below:

1. Clean the YVO_4 chip using acetone and isopropanol (IPA), followed by nitrogen blow dry.
2. Deposit 50 nm of chromium (Cr) and 115 nm of gold (Au) using an electron-beam evaporator. Cr serves both as an adhesion layer for Au and as a conductive layer for focused ion beam milling.
3. Clean the YVO_4 chip and spin-coat the chip with ZEP 520A resist. Due to the small chip size (3mm), it is temporarily affixed to a larger silicon wafer using Kapton tape. The chip is pre-heated on a 180°C hotplate for 3 minutes, then spin-coated at 3000 rpm (twice), followed by a post-bake at 180°C for another 3 minutes.
4. Load the chip into the EBL system, and write the pattern using a beam current of 5 nA and an exposure dose of 200 $\mu\text{C}/\text{cm}^2$.
5. Develop the pattern in ZED-N50 developer for 90 seconds, gently agitating the beaker to ensure uniform development. Rinse with IPA and blow dry.
6. Etch the exposed gold areas using gold etchant for 30 seconds, followed by a DI water rinse and nitrogen drying.
7. Remove the remaining ZEP 520A resist by soaking the chip in Remover PG overnight on a hotplate at 170°C.

2.2.1.2 Photonic Crystal Cavity Design and Fabrication

In this section, we briefly describe the design principles and fabrication procedures for the nanophotonic crystal cavities used in our experiments [109]. Photonic crystal (PhC) cavities are optical resonators formed by introducing a defect between two highly reflective mirror segments in a periodic dielectric structure, thereby confining light at a target wavelength [108]. The mirror segments are engineered so that transmission of the target wavelength is strongly suppressed, while a localized cavity mode forms at the central defect. Using such a cavity enhances the interaction between photons and rare-earth ions via the Purcell effect, increasing spontaneous emission rates and photon collection efficiency.

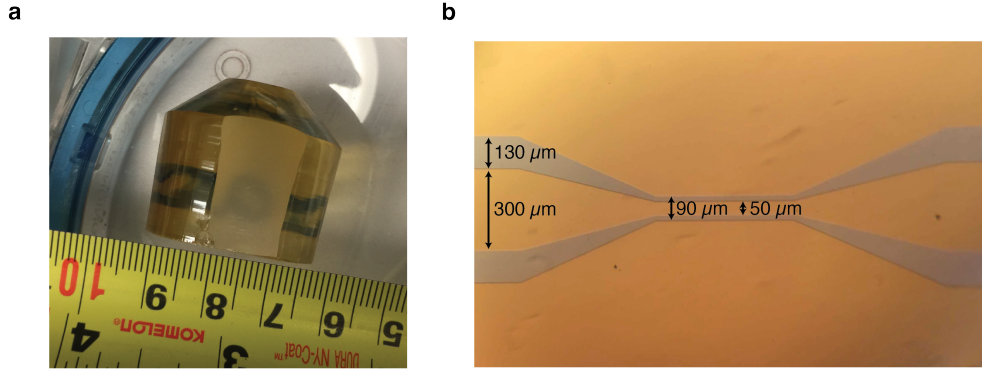


Figure 2.2: YVO_4 boule and coplanar waveguides. (a) YVO_4 boule grown by Gamdan Optics. (b) Gold coplanar waveguide fabricated on a YVO_4 chip, imaged under an optical microscope.

In this work, we focus on one-dimensional (1D) photonic crystal nanobeam cavities, which minimize the optical mode volume and thus maximize the Purcell enhancement for embedded emitters. To design the mirror segments, frequency-domain band structure calculations (e.g., using the MPB package [110]) are employed; a larger photonic band gap yields stronger mirror reflectivity and better mode confinement. The structure is first optimized to maximize the photonic band gap around the $^{171}\text{Yb}^{3+}$ transition wavelength (984.5 nm), ensuring high-quality optical confinement for the relevant transition.

A quadratic period tapering is implemented to smoothly transition from the highly periodic mirror regions to the central defect, producing a nearly Gaussian cavity mode profile and minimizing radiative losses [111]. The entire cavity structure is then simulated using finite-difference time-domain (FDTD) methods to optimize the quality factor (Q) at the target wavelength. Commercial and open-source software packages, such as COMSOL, Lumerical, or MEEP, are used for these simulations (see Figure 2.3(a)). Our final design utilizes a nanobeam with 34 periods, where 14 central periods are quadratically tapered to form the localized cavity mode.

In our experiments, we specifically target the “A” optical transition of $^{171}\text{Yb}^{3+}$, which is spectrally isolated from other isotopes. To maximize coupling to this transition on c -cut YVO_4 samples, we design TM-polarized PhC cavities that support dominant electric fields aligned along the crystal c -axis. The nanobeam geometry consists of a triangular cross-section with periodic air holes, and the lattice parameters are scaled from reference designs at 880nm to 984.5nm [109].

Fabrication of the PhC cavities is performed using a focused gallium ion beam (FIB, Nova 600 NanoLab) to directly mill the structures into the YVO_4 substrate. This technique enables maskless prototyping directly in hard crystalline materials like YVO_4 , a significant advantage given the lack of high-quality, commercially available thin films, but is relatively slow. The additional drawback is the potential for crystal damage and gallium ion implantation from the Ga^+ beam, which can introduce optical loss and create charge traps that exacerbate spectral diffusion [112]. We mitigate these deleterious effects by using a multi-step milling process: a higher ion current is used for bulk material removal to define the triangular nanobeam, followed by a lower current to thin the beam and reduce sidewall roughness. After nanobeam formation, the sample stage is rotated so that the beam is perpendicular to the ion column for precise patterning of the mirror and defect grooves at low current (23pA), with in-situ scanning electron microscope (SEM) imaging used to minimize specimen current. Finally, optical couplers are added by milling angled facets (45° to the sample surface) at each end of the cavity (Figure 2.3(b)).

Post-fabrication, devices are immersed in chromium etchant for 30 seconds to remove the conductive Cr layer deposited before FIB processing. The completed YVO_4 chips are then mounted on copper stubs using silver paint and wire-bonded to a printed circuit board (PCB) for measurement (Figure 2.3(c)). For the devices that we used in the experiments, we fabricated one-sided cavities, where we reduce the mirror periods on one side to increase the coupling to the waveguide. Optical characterization reveals typical cavity quality factors (Q) of approximately 10^4 (Figure 2.3(d)), and measured output coupling ratios ($\kappa_{\text{in}}/\kappa$) around 0.12. The simulated mode volume of the device is about $0.095\mu\text{m}^3$ [91], calculated as:

$$V_{\text{mode}} = \frac{\int dV \epsilon(\mathbf{r}) |E(\mathbf{r})|^2}{\max(\epsilon(\mathbf{r}) |E(\mathbf{r})|^2)}, \quad (2.6)$$

where $E(\mathbf{r})$ is the local electric field and $\epsilon(\mathbf{r})$ is the permittivity distribution in the cavity. This integrated nanophotonic platform achieves strong light-matter coupling with single rare-earth ions, forming the basis for high-efficiency spin-photon interfaces and quantum network protocols.

2.2.2 Cavity QED Regime

The interaction between a quantum emitter and an optical cavity is described by the framework of cavity quantum electrodynamics (cQED). The cavity modifies the local density of optical states, leading to changes in the physical properties of

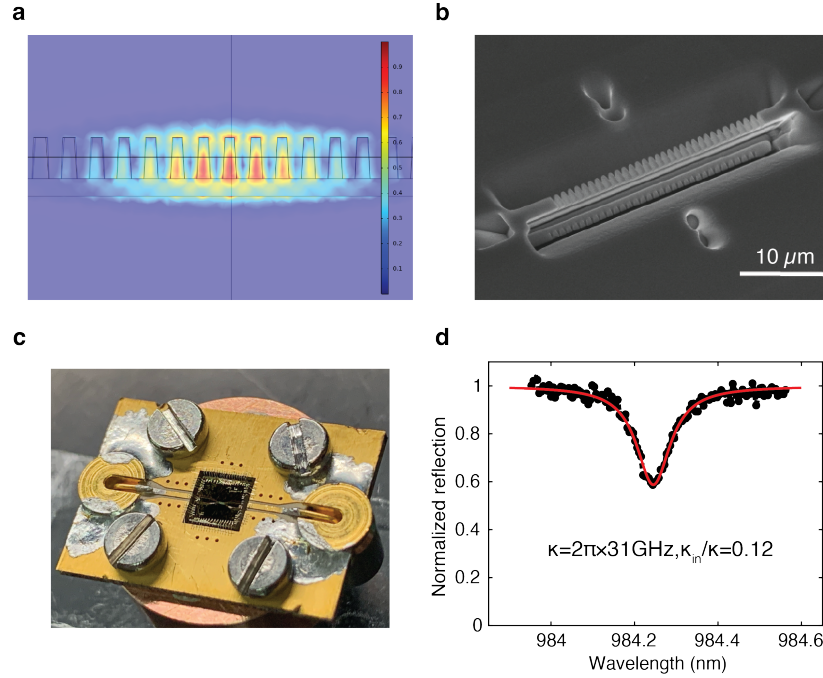


Figure 2.3: Design and fabrication of photonic crystal cavities. (a) Simulated electric field profile of the localized cavity mode in a one-dimensional photonic crystal nanobeam; the color scale indicates normalized electric field strength. (b) SEM image of a fabricated nanophotonic crystal cavity milled in YVO₄. (c) Photo of the assembled device, showing the chip mounted on a copper stub and wire-bonded to a PCB for measurement. (d) Experimental reflection spectrum of the fabricated photonic crystal cavity, exhibiting a quality factor (Q) of approximately 10,000 and an input coupling ratio $\kappa_{in}/\kappa = 0.12$.

atoms, such as their spontaneous emission rates. The atom-cavity system can be characterized by three key parameters: g , the atom cavity coupling strength; κ , the cavity decay rate; and γ , the atom decay rate [113].

For the cavity presented in Section 2.2.1.2, $\kappa = 2\pi \times 31.4 \text{ GHz}$. The atom-cavity coupling rate can be estimated from the atomic transition dipole moment ($1.06 \times 10^{-31} \text{ C} \cdot \text{m}$) using the following equation [91, 106]:

$$g = \frac{\mu}{\hbar} \sqrt{\frac{\hbar \omega}{2\epsilon_0 n^2 V}}, \quad (2.7)$$

where μ is the transition dipole moment, ω is the transition frequency, n is the refractive index of the medium, and V is the cavity mode volume. The maximum coupling strength is expected to be $g_{\max} = 2\pi \times 25.5 \text{ MHz}$.

The bulk excited state lifetime of ^{171}Yb in YVO_4 is measured to be $267 \mu\text{s}$ (corresponding to $\gamma_0 = 1/(267 \mu\text{s})$), with a branching ratio of approximately 0.35 for this transition. These parameters place our system in the bad cavity regime, where $\kappa \gg g, \gamma$. In this regime, the emitter lifetime is reduced through the Purcell effect [114], and can be calculated by using:

$$\gamma_{\text{cav}}/\gamma_0 = 1 + 4g^2/\kappa\gamma = 1 + \beta F_p, \quad (2.8)$$

where β is the branching ratio of the transition and F_p is the Purcell enhancement factor. This yields a minimum lifetime of approximately $1.9 \mu\text{s}$. The Purcell enhancement also leads to preferential emission of photon into the cavity mode, with the fraction given by

$$\frac{\beta F_p}{1 + \beta F_p}, \quad (2.9)$$

which is over 99%. While this on-chip collection efficiency is near-unity, the final end-to-end efficiency is limited by a cascade of subsequent loss mechanisms. As detailed in the next section, factors such as waveguide-to-fiber coupling reduce the overall measured efficiency to approximately 1.5%, highlighting key areas for future engineering improvements.

2.2.3 Device and Experimental Setup

The measurements are performed in a ^3He cryostat (Bluefors LD-He250) at a base temperature of 0.5 K. This cryogenic operation is essential to suppress phonon-mediated spin-lattice relaxation [84], which is critical for achieving the long optical and spin coherence times required for quantum networking. Devices are mounted onto a copper stub, which is then attached to a custom L-shaped mount fabricated from oxygen-free copper to ensure optimal thermal anchoring (Figure 2.4). Optical access is provided via a lens doublet mounted on an x-y-z piezoelectric nanopositioner (Attocube), allowing for precise alignment of the optical mode with the nanophotonic devices.

The resonance frequency of the photonic crystal cavity is tuned to match the ^{171}Yb optical transitions by controlled nitrogen gas condensation, delivered through dedicated copper tuning lines. Ambient magnetic fields are actively cancelled using a custom-wound superconducting magnetic coil, minimizing unwanted Zeeman shifts and enabling reproducible spin control.

The optical and microwave control setup is illustrated in Figure 2.5. Initialization and readout optical pulses on the A and F transitions of ^{171}Yb are generated by a

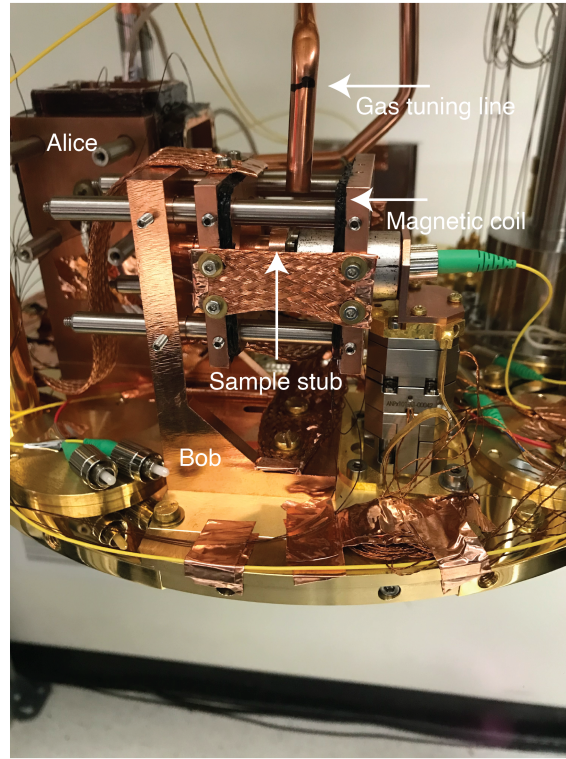


Figure 2.4: Device setups inside a ^3He cryostat. Two device setups (“Alice” and “Bob”) are shown. Different components are labelled in the figure.

titanium:sapphire (Ti:Sapph) laser (M2 Solstis) and continuous-wave (CW) external cavity diode lasers (Toptica DL-Pro), respectively. The Ti:Sapph laser is stabilized to a high-finesse reference cavity using Pound-Drever-Hall (PDH) locking, while the diode lasers are offset-locked to the Ti:Sapph using beat-note feedback, ensuring long-term frequency stability and accuracy. Acousto-optic modulators (AOMs), arranged in two- and three-double-pass configurations [115], produce optical pulses with precise timing and frequency control for selective addressing of ^{171}Yb transitions.

Ground-state microwave control (centered around 675MHz) and RF signals for qubit and nuclear spin manipulation are synthesized directly by a programmable arbitrary waveform generator (Tektronix 5204AWG), then amplified (Amplifier Research 10U1000). Excited-state microwave control at 3.37 GHz is generated by a signal generator (SRS SG384), gated with a fast microwave switch, and amplified (Minicircuits ZHL-16W-43-S+). Both microwave paths are combined using a diplexer (Marki DPXN2) and routed to the device.

Integration and timing of experimental sequences are managed by a Quantum Machines OPX control platform, which synchronizes laser and microwave pulses, manages readout, and triggers both optical and spin-state measurements. Detection is achieved using a superconducting nanowire single-photon detector (SNSPD, Photon Spot), with the output signal amplified and registered by the OPX for time-correlated photon counting.

The optical collection path is optimized to maximize the overall detection efficiency. Photons emitted from the device are efficiently coupled into an optical fiber and directed to the superconducting nanowire single-photon detector (SNSPD). Of the collected light, 99% is transmitted to the detection channel via a fiber coupler, and a free-space acousto-optic modulator (AOM) functions as a fast optical shutter with an efficiency of 66%. The SNSPD itself has a detection efficiency of 85%, resulting in a total optical detection efficiency of approximately 55% for light delivered to the detector. Considering additional system-level losses—including the 45° input/output couplers (25% efficiency) and the coupling from the cavity to the waveguide (12%)—the overall end-to-end efficiency from cavity emission to photon detection is about 1.5%.

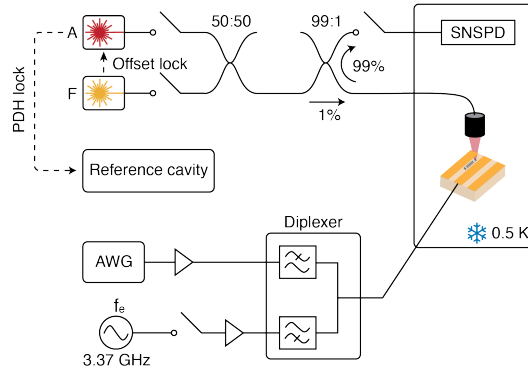


Figure 2.5: Optical and microwave experimental setup. Laser beams are combined and directed to the photonic crystal cavity. Light emitted from the device is collected through a fiber coupler and detected with a superconducting nanowire single-photon detector (SNSPD). Microwave and RF signals for coherent spin and nuclear control are generated by an arbitrary waveform generator (AWG) and signal generator, respectively, then amplified, combined, and delivered to the device. All devices and the SNSPD are housed inside the ^3He cryostat for cryogenic operation.

2.3 Single Ion Properties

After introducing the experimental setup and physical system, we now characterize the essential optical and spin properties of single ^{171}Yb ions, which underpin their suitability as quantum network nodes.

2.3.1 Photoluminescence Spectrum and $g^{(2)}$ measurements

We begin by acquiring the photoluminescence (PL) spectrum around the A transition of ^{171}Yb ions (Figure 2.6(a)). Due to variations in local crystal strain and environment, each ^{171}Yb ion can exhibit a slightly different optical transition frequency, resulting in an inhomogeneous broadening of approximately 200 MHz in YVO_4 [106]. Near the center of this distribution, multiple ions may overlap in frequency; by probing the spectral wings, we identify isolated PL peaks that correspond to individual ions. The following measurements focus on a specific, well-isolated ^{171}Yb ion in the device, though many ions with similar properties are present.

To determine the optical lifetime, we resonantly excite the selected ion and perform time-resolved PL measurements. The measured lifetime is $2.167 \pm 0.012 \mu\text{s}$ (Figure 2.6(b)), more than two orders of magnitude shorter than the bulk value of $267 \mu\text{s}$ [106]. This substantial reduction is attributed to Purcell enhancement from coupling the ion to a nanophotonic crystal cavity, which has a quality factor of 10^4 and a mode volume of approximately $0.095 \mu\text{m}^3$. The increased emission rate enhances the ion's cyclicity, which is the probability of emission into the desired transition per optical cycle, making it more efficient for quantum information applications.

In bulk YVO_4 , the A transition exhibits a branching ratio of approximately 0.35 [106]. Cavity coupling modifies this ratio, which can be estimated using

$$\beta^{\text{cav}} = 1 - (1 - \beta) \frac{T_1^{\text{cav}}}{T_1^{\text{bulk}}}, \quad (2.10)$$

where β^{cav} is the branching ratio after Purcell enhancement, β is the branching ratio in bulk YVO_4 , T_1^{cav} and T_1^{bulk} are the optical lifetime after and before coupling to the optical cavity, respectively. This predicts a lower bound on the branching ratio of about 0.995, which assumes that the other crystal levels do not decay to $|1\rangle_g$. This Purcell-enhanced emission rate is directly responsible for the high cyclicity of the optical transition. By shortening the excited state lifetime, the probability of off-resonant decay to other ground-state levels during a single excitation cycle is suppressed. The cyclicity is experimentally confirmed by initializing the ion to $|1\rangle_g$ and applying repeated optical π pulses; the count rate decay yields a cyclicity

of 0.9969 ± 0.0003 (Figure 2.6(c)), consistent with this prediction. This near-unity cyclicity is a key enabler for the high-fidelity readout.

To verify single-photon emission, we measure the second-order autocorrelation function $g^{(2)}(\tau)$ using pulsed optical excitation [116]. Between each pulse, initialization ensures the ion is reset to $|1\rangle_g$. The measured value, $g^{(2)}(0) = 0.010 \pm 0.002$ (Figure 2.6(d)), is well below the single-photon threshold of 0.5, providing unambiguous proof of a single quantum emitter.

Finally, to verify the ion's identity and probe additional ground state structure, we perform pump-probe spectroscopy on the F transition. Here, the frequency of the pump pulse is scanned while PL is detected on the A transition (Figure 2.6(e)). The resulting spectrum reveals two peaks, indicative of non-degeneracy in the $|aux\rangle_g$ state; further discussion of this structure is presented in a later section.

2.3.2 Optical Driving and Coherence

A key requirement for distributing entanglement between quantum nodes is the ability to generate indistinguishable photons and to coherently control the ion's states. To demonstrate coherent optical manipulation and to characterize the optical coherence properties of ^{171}Yb ions, careful state initialization is essential. We begin by optically pumping the ions multiple times on both F transitions and allowing for spontaneous relaxation, followed by additional pumping on the A transition and the application of an excited-state microwave pulse (f_e). This sequence deterministically prepares the ion in the $|0\rangle_g$ ground state. A subsequent microwave π pulse on the f_g transition (detailed later) then initializes the ion into the $|1\rangle_g$ state.

Optical Rabi oscillations are measured by driving the $|1\rangle_g \leftrightarrow |0\rangle_e$ transition with optical pulses of varying durations (Figure 2.7(a)), demonstrating coherent control with an optical π -pulse time of approximately 100 ns. Ramsey coherence experiments, performed by applying two optical $\frac{\pi}{2}$ pulses separated by a variable delay, yield an optical Ramsey coherence time of 309 ± 17 ns (Figure 2.7(b)).

By inserting an optical π pulse between the Ramsey $\frac{\pi}{2}$ pulses (optical echo sequence), we probe the frequency stability on shorter timescales. The measured optical echo coherence time is $3.66 \pm 0.12 \mu\text{s}$ (Figure 2.7(c)). The substantial recovery of coherence from the Ramsey to the echo measurement, with the echo coherence time approaching the lifetime-limited value of $2T_{1,\text{opt}} \approx 4.3 \mu\text{s}$, indicates that the dominant source of optical dephasing is low-frequency noise. This is consistent with spectral diffusion arising from charge fluctuations in the vicinity of the nanostructure [91].

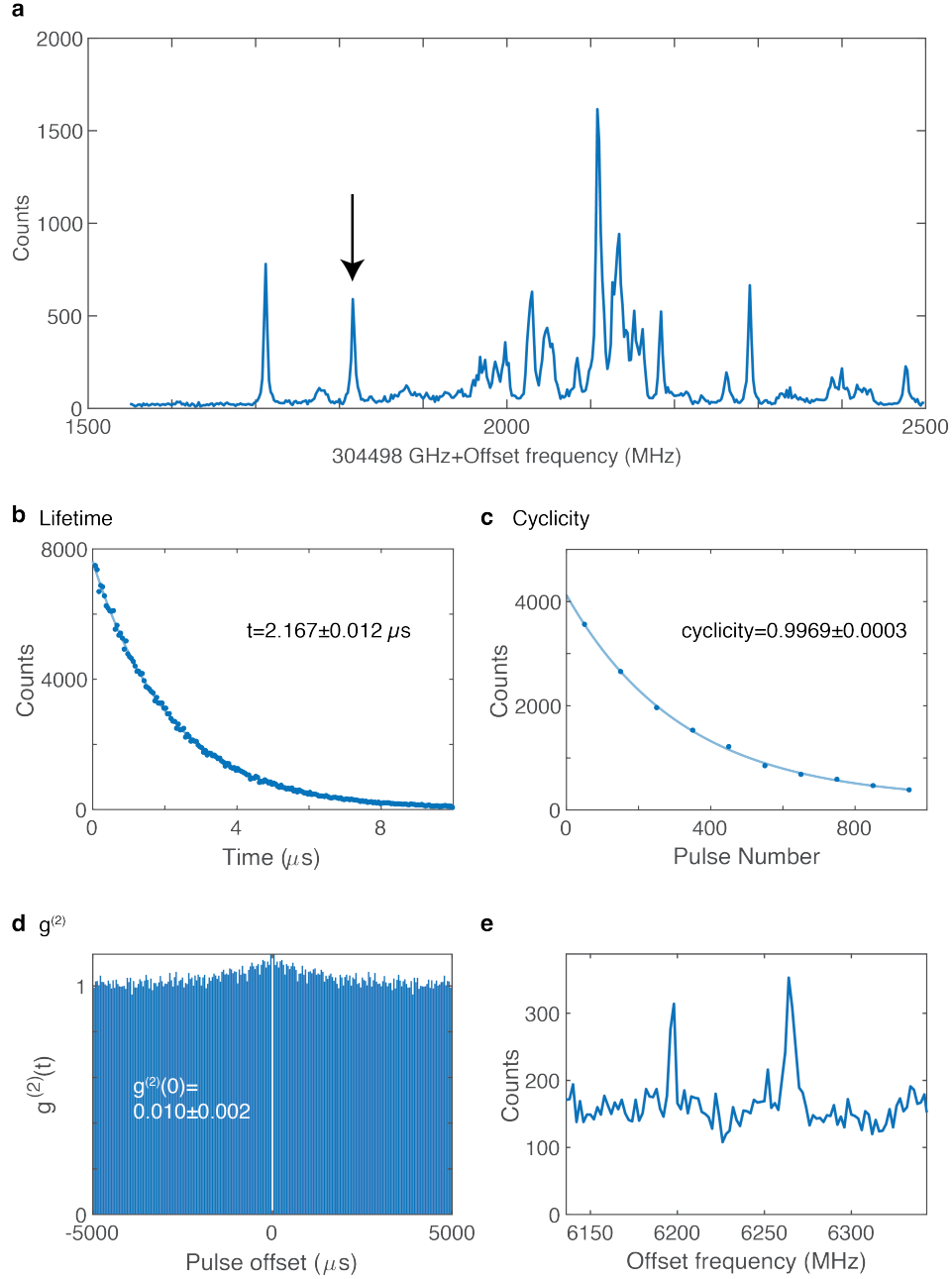


Figure 2.6: Optical characterization of single ^{171}Yb ions in YVO_4 . (a) PL spectrum near the A transition of ^{171}Yb . Peaks corresponding to individual ions are observed; subsequent measurements refer to the highlighted ion. (b) Measured optical lifetime of the selected ion, showing Purcell-enhanced emission with a lifetime of $2.167 \pm 0.012 \mu\text{s}$. (c) Cyclicity of the same ion, measured to be 0.9969 ± 0.0003 . (d) Second-order correlation $g^{(2)}(\tau)$ for the indicated ion, with $g^{(2)}(0) = 0.010 \pm 0.002$, confirming single-photon emission. (e) Optical pump-probe measurement on the selected ion, revealing the F transition resonance.

A long time-scale spectral diffusion measurement on the A transition was also performed to measure its stability (Figure 2.8).

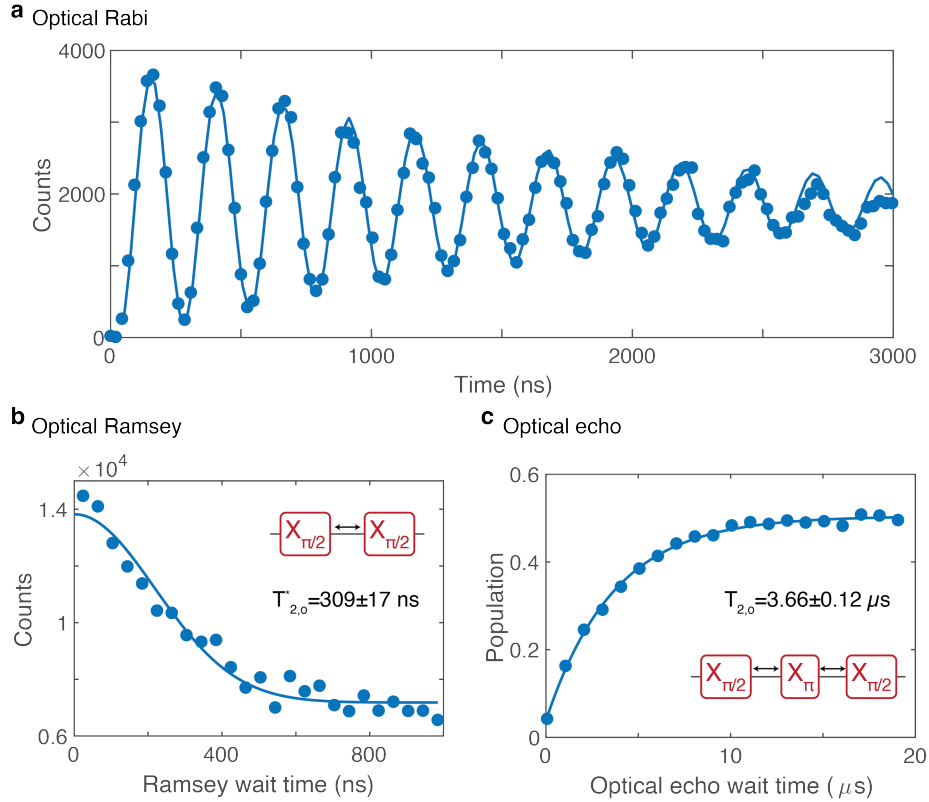


Figure 2.7: Coherence and optical control of a single ^{171}Yb ion in YVO_4 . (a) Optical Rabi oscillations of a single ^{171}Yb ion; the solid line is a simulation based on the ion's optical Ramsey coherence time and measured lifetime. (b) Optical Ramsey interference measurement; the solid line is a fit assuming Gaussian decay, yielding a coherence time of 309 ± 17 ns. (c) Optical spin echo measurement; the solid line is an exponential fit, yielding a coherence time of 3.66 ± 0.12 μ s. Insets depict the pulse sequences used in each measurement.

2.3.3 Aux State Splitting

In the pump-probe measurement of the F transition (Figure 2.6(e)), we observe two distinct spectral peaks—an unexpected outcome given the nominal site symmetry of ^{171}Yb in YVO_4 . To investigate the origin of this splitting, we perform additional measurements as a function of applied magnetic field. An avoided crossing is evident in Figure 2.9, indicating that the observed splitting arises from a physical mechanism beyond simple hyperfine structure.

We attribute this splitting to a slight asymmetry in the local crystal environment,

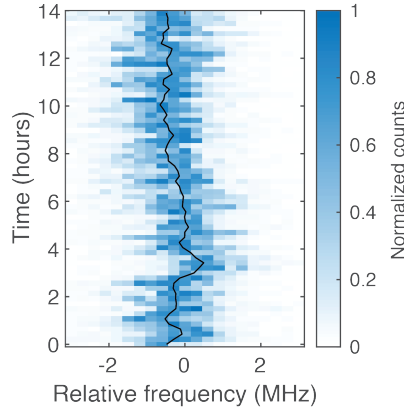


Figure 2.8: Long-term spectral diffusion measurement of a single ^{171}Yb ion, tracked over a 14-hour period.

which lifts the degeneracy of the two a -axes and results in an anisotropic hyperfine tensor. When the hyperfine couplings differ along all three crystal axes, the $|\text{aux}\rangle_g$ state can split into two first-order magnetic-field-insensitive states. These states can be represented as:

$$|\text{aux}\rangle_{g0} = (|\uparrow_{\text{Yb}}\uparrow_{\text{Yb}}\rangle - |\downarrow_{\text{Yb}}\downarrow_{\text{Yb}}\rangle)/\sqrt{2},$$

$$|\text{aux}\rangle_{g1} = (|\uparrow_{\text{Yb}}\uparrow_{\text{Yb}}\rangle + |\downarrow_{\text{Yb}}\downarrow_{\text{Yb}}\rangle)/\sqrt{2}.$$

The splitting between these two levels varies between ions, typically ranging from 10 to 100 MHz. Additional measurements and discussion of the underlying physics can be found in Chapter 5.

2.3.4 Spin Driving and Coherence

Distributing quantum entanglement over long distances requires local qubits to maintain coherence while photons are transmitted between nodes, making long qubit coherence times essential for scalable quantum networks. Here, we demonstrate high-fidelity coherent spin control and measurements of spin coherence for single ^{171}Yb ions in YVO_4 .

In these experiments, the ^{171}Yb ion is first initialized to the $|0\rangle_g$ state using repeated optical pumping on both F transitions, followed by waiting for optical decay. Subsequent optical π on the A transition together with a subsequent excited-state microwave (f_e) π pulse deterministically prepares the ion in $|0\rangle_g$. An additional ground-state microwave π pulse on f_g is then applied to initialize the ion into $|1\rangle_g$. For all measurements, population readout is achieved via the contrast in photon

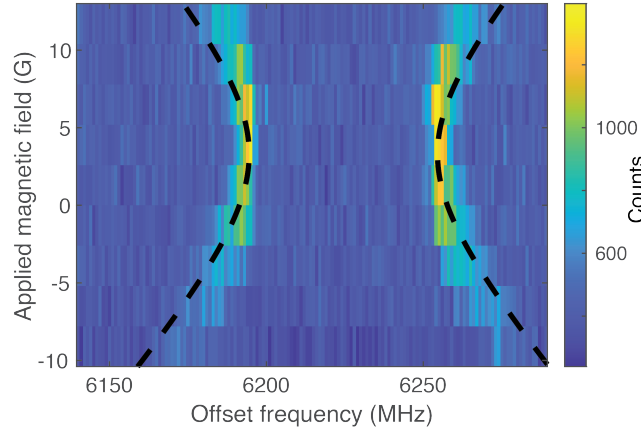


Figure 2.9: Pump-probe measurement of the F transition. The magnetic field is varied to examine the field dependence of the frequency splitting, revealing an avoided crossing.

counts between the $|0\rangle_g$ and $|1\rangle_g$ states, with the $|0\rangle_g$ population measured by inserting a qubit π pulse prior to the readout sequence.

Figure 2.10(a) shows high-contrast Rabi oscillations between the $|0\rangle_g$ and $|1\rangle_g$ states, demonstrating high-fidelity coherent control of the single spin qubit. We further perform spin Ramsey and spin echo measurements to assess coherence properties, as shown in Figure 2.10(b) and (c). The spin Ramsey signal exhibits a coherence time limited primarily by interactions with the surrounding ^{51}V nuclear spin bath. The data are fit using a model that incorporates both nuclear bath and echo decay, enabling extraction of the ^{51}V nuclear g -factor (approximately 1.3). The spin echo experiment reveals a significantly increased coherence time of $44.5 \pm 0.3 \mu\text{s}$, well above the Ramsey time, indicating that dephasing in Ramsey measurements is mainly due to slow, quasi-static noise.

Further coherence extension is achieved using XY8 dynamical decoupling sequences. By carefully selecting the interpulse delay (optimized at $5.8 \mu\text{s}$ to avoid ^{51}V resonance frequencies), the coherence time is prolonged to $16.8 \pm 0.8 \text{ ms}$ Figure 2.10(d). Current results suggest that both spin echo and XY8 coherence times are limited by stochastic nuclear spin flip-flops in the environment, though this remains an area of active investigation using theoretical models such as correlated cluster expansions [117]. The measured T_1 relaxation time of the ^{171}Yb qubit is $53 \pm 3 \text{ ms}$ (Figure 2.10(e)); ongoing studies with lower doping concentrations in our group have observed T_1 values between 100 and 200 ms.

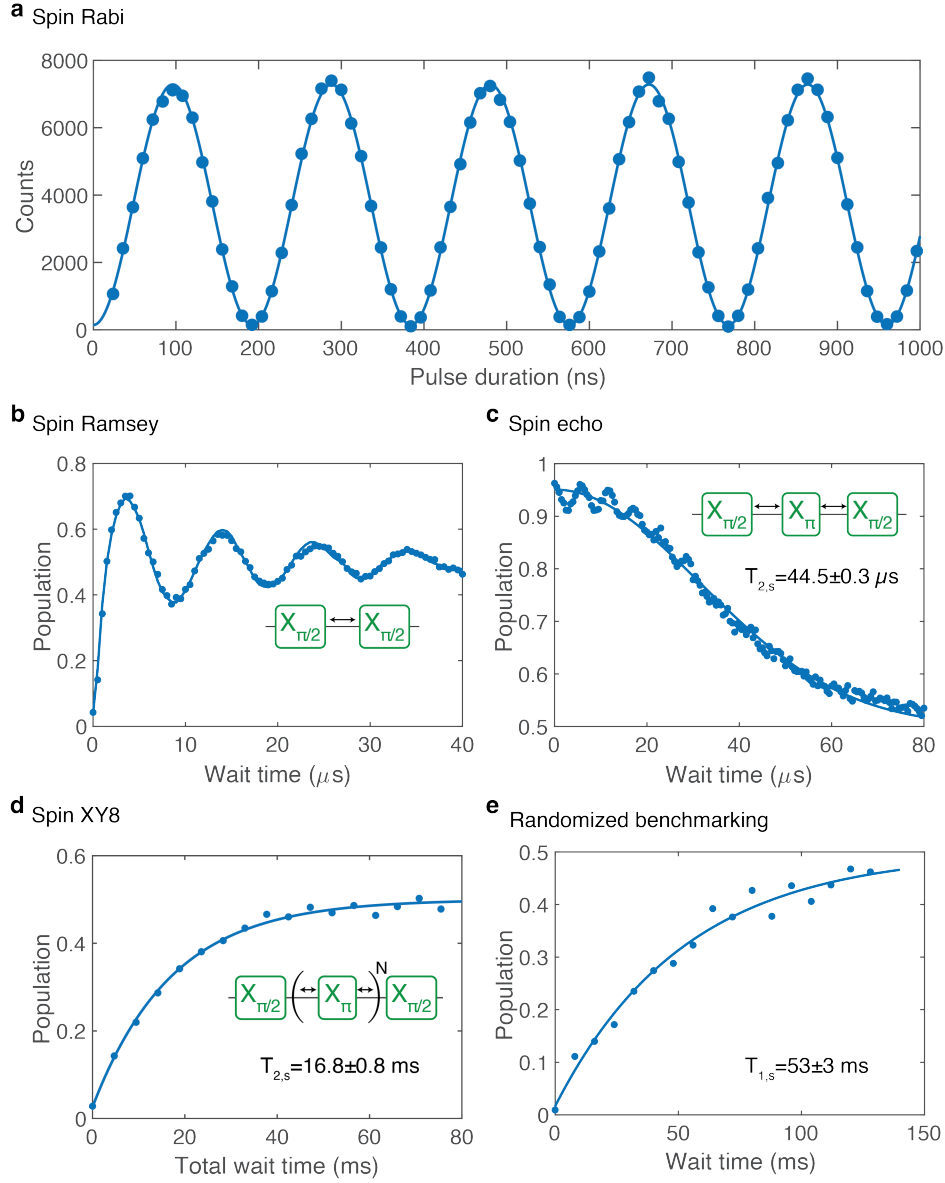


Figure 2.10: Spin coherence and control of a single ^{171}Yb ion in YVO_4 . (a) Spin Rabi oscillations; the solid line is a cosine fit. (b) Spin Ramsey coherence measurement at a detuning of 0.1 MHz, with a fit including vanadium nuclear spins and echo decay. (c) Spin echo coherence, with a Gaussian fit yielding a time of $44.5 \pm 0.3 \mu\text{s}$. (d) XY8 dynamical decoupling measurement, optimized at a $5.8 \mu\text{s}$ wait time, and fit to an exponential decay with $16.8 \pm 0.8 \text{ ms}$ coherence. (e) T_1 relaxation measurement, fit to an exponential with a timescale of $53 \pm 3 \text{ ms}$.

To quantify gate control fidelity, we perform single-qubit randomized benchmarking on individual ^{171}Yb ions following the protocol in [118]. Random sequences of Clifford gates are applied such that the qubit ideally returns to its initial state. The

decay of the qubit population as a function of the Clifford sequence length M is fitted to:

$$P_{|1\rangle_g}(M) = P_{|1\rangle_g}(0)p_c^M + P_\infty,$$

$$F_c = (1 + p_c)/2,$$

where $P_{|1\rangle_g}(M)$ is the measured population in $|1\rangle_g$ after M Clifford gates, p_c relates to the average gate fidelity, P_∞ is the population for a randomized state, and F_c is the extracted Clifford gate fidelity. This method is robust to SPAM errors. We obtain an average gate fidelity of 0.99975 ± 0.00002 (Figure 2.11), confirming the high quality and robustness of spin control in this platform.

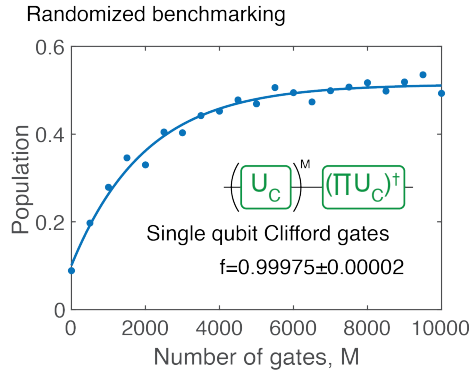


Figure 2.11: Randomized benchmarking of single-qubit control of ^{171}Yb . An average Clifford gate fidelity of 0.99975 ± 0.00002 is achieved.

2.3.5 Statistics of Single ^{171}Yb Ions

To assess device performance and variability beyond a single qubit, we collected statistical data on several individual ^{171}Yb ions measured across two different devices, labeled “Alice” and “Bob.” For each ion, key spectroscopic properties were extracted, including transition frequencies and F transition splittings. Notably, we observe that qubit coherence times (T_2) and energy relaxation lifetimes (T_1) do not exhibit any discernible correlation with either the optical transition frequency or the qubit transition (microwave) frequency; as such, these quantities are not included in the summary figures. The XY8 spin coherence times are usually in the range of 10 to 30 ms, and most of the ions have very similar spin echo coherence times.

Figure 2.12 presents two representative correlations. In Figure 2.12(a), we plot the splitting of the F transition as a function of the A transition optical frequency. The data reveal that ions whose A transition frequencies lie further from the spectral

center—typically corresponding to ions subject to stronger local strain or environmental shifts—exhibit larger F transition splittings. This trend suggests a link between local crystal fields and the anisotropy affecting the auxiliary states, connecting microscopic material properties to macroscopic spectroscopic observables.

Figure 2.12(b) shows the relationship between the qubit transition frequency (f_g) and the A transition optical frequency. Here, a negative correlation is observed: ions with higher optical transition frequencies tend to display lower qubit transition frequencies. Such correlations provide insight into how microscopic environment and host crystal parameters influence the energy structure of individual ^{171}Yb ions.

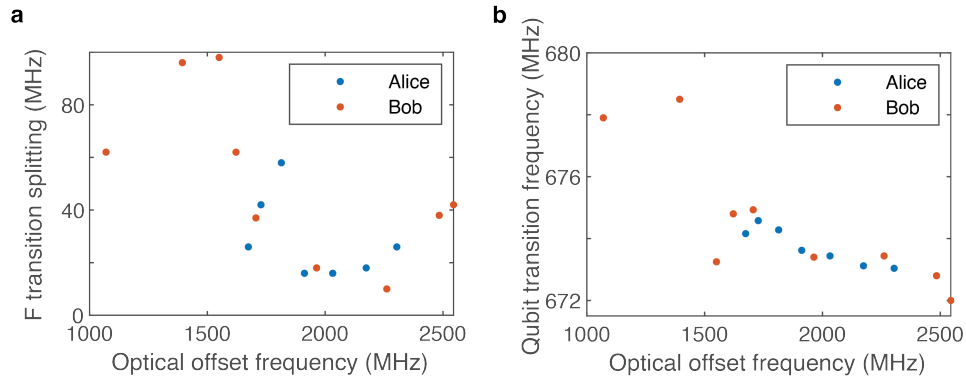


Figure 2.12: Statistical properties of single ^{171}Yb ions measured across two devices. (a) F transition splitting as a function of A transition frequency. Ions with greater optical frequency offsets from the mean tend to exhibit larger F transition splittings. (b) Qubit transition frequency (f_g) as a function of A transition frequency, showing a inverse correlation.

2.4 Nuclear Spin Coupling

Nuclear spins in the solid-state environment provide a powerful resource for quantum information processing and quantum networking [54, 119, 120]. In addition to serving as auxiliary long-lived quantum memories for single rare-earth qubits, collective nuclear spin ensembles can be leveraged for quantum simulation and many-body physics studies [121, 122]. In the yttrium orthovanadate (YVO_4) host lattice, the principal constituents are yttrium (spin-1/2), vanadium (^{51}V , spin-7/2), and oxygen (spin-zero). At zero magnetic field, only the ^{51}V nuclei exhibit non-trivial energy structure due to the quadrupolar interaction [123], and of all isotopes present, ^{51}V dominates the local nuclear spin environment as it possesses both a high nuclear g -factor and high natural abundance.

This section provides an overview of how nuclear spin ensembles, particularly those comprised of ^{51}V , can be coherently controlled and utilized for quantum memory, focusing on the main physical couplings, Hamiltonians, and pulse sequences. We also discuss the distinction between nearby (“register”) and distant (“bath”) ^{51}V nuclei and describe how quantum state storage and retrieval is realized via engineered interactions.

2.4.1 System Hamiltonian

^{51}V nuclear spins have spin-7/2 and are governed by the Hamiltonian (Figure 2.13):

$$\hat{H}_V = Q\hat{I}_z^2 + \mu_N \mathbf{B} \cdot \mathbf{g}_v \cdot \hat{\mathbf{I}}, \quad (2.11)$$

where Q is the nuclear electric quadrupole parameter, $\hat{I}_z$ and $\hat{\mathbf{I}}$ are the z-component and vector of the ^{51}V nuclear spin operators, respectively, μ_N is the nuclear magneton, and $\mathbf{g}_v$ is the nuclear g-tensor of ^{51}V , which is given by:

$$\mathbf{g}_v = \begin{bmatrix} 0.6 & 0 & 0 \\ 0 & 0.6 & 0 \\ 0 & 0 & 1.3 \end{bmatrix} \quad (2.12)$$

g_{vz} is estimated by fitting ^{171}Yb Ramsey coherence times and g_{vx} is estimated by calibrating the RF field amplitude the experiment result of direct driving (Chapter 4).

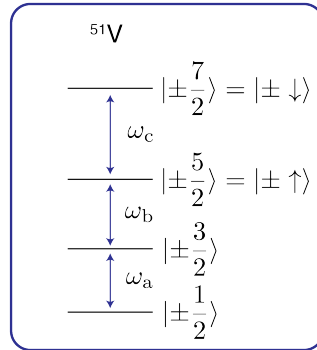


Figure 2.13: Energy level structure of ^{51}V . ^{51}V has four doubly degenerate levels under zero magnetic field.

The interaction between ^{171}Yb and ^{51}V is described by the dipolar Hamiltonian:

$$\hat{H}_{\text{int}} = \frac{\mu_0 \mu_B \mu_N}{4\pi |\mathbf{r}|^3} [3(\mathbf{g} \cdot \hat{\mathbf{S}} \cdot \hat{\mathbf{r}})(\mathbf{g}_v \cdot \hat{\mathbf{I}} \cdot \hat{\mathbf{r}}) - (\mathbf{g} \cdot \hat{\mathbf{S}}) \cdot (\mathbf{g}_v \cdot \hat{\mathbf{I}})], \quad (2.13)$$

where $\mathbf{r}$ is the displacement vector between ^{171}Yb - ^{51}V . By transforming the ^{171}Yb basis to $|0\rangle_g$ and $|1\rangle_g$ and applying the Schrieffer-Wolff transformation [124] up to second order, treating the electronic Zeeman and ^{171}Yb - ^{51}V interaction as perturbative terms, the effective Hamiltonian becomes:

$$\hat{H}_{\text{eff}} = \omega_0 \tilde{S}_z + Q \hat{I}_z^2 + \mu_N \mathbf{B} \cdot \mathbf{g}_v \cdot \hat{\mathbf{I}} + \hat{H}_{\text{pert}}, \quad (2.14)$$

where $\hat{H}_{\text{pert}}$ contains the perturbative contributions. Neglecting the effects of $|\text{aux}\rangle_g$, due to small g_x^2 , g_y^2 , and larger energy splitting between $|\text{aux}\rangle_g$ and ^{171}Yb qubit states, the perturbative term simplifies to:

$$\begin{aligned} \hat{H}_{\text{pert}} = \frac{\tilde{S}_z}{2\omega_0} [\mu_B g_z B_{\text{drive}} + \sum_i \frac{\mu_0 \mu_B \mu_N g_z}{4\pi |\mathbf{r}_i|^3} (3\hat{r}_{x,i} \hat{r}_{z,i} g_{vx} \hat{I}_{x,i} + 3\hat{r}_{y,i} \hat{r}_{z,i} g_{vy} \hat{I}_{y,i} \\ + (3\hat{r}_{z,i}^2 - 1) g_{vz} \hat{I}_{z,i})] \end{aligned} \quad (2.15)$$

where $\tilde{S}_z = (|1\rangle\langle 1| - |0\rangle\langle 0|)/2$, the sum runs over all ^{51}V , and $\hat{r}_{x,i}$, $\hat{r}_{y,i}$, $\hat{r}_{z,i}$ are the direction cosines between ^{171}Yb and each ^{51}V . Since the bath spins have different resonance frequencies from the register spins (will be introduced in the next section), we treat the bath nuclear spins as mixed states and trace out their contributions. The Overhauser field (field noise on the central spin) from the bath spins is given by:

$$B_{\text{OH}} = \sum_{i \in \text{bath}} \frac{\mu_0 \mu_N g_{vz}}{4\pi |\mathbf{r}_i|^3} (3\hat{r}_{z,i}^2 - 1) \hat{I}_{z,i}. \quad (2.16)$$

Substituting this into the expression above, the perturbed Hamiltonian becomes:

$$\begin{aligned} \hat{H}_{\text{pert}} = \tilde{S}_z \left[\frac{\mu_B^2 g_z^2 (B_{\text{drive}} + B_{\text{OH}})^2}{2\omega_0} + \frac{\mu_B g_z (B_{\text{drive}} + B_{\text{OH}})}{\omega_0} \left(\sum_{i \in \text{reg}} a_{x,i} \hat{I}_{x,i} + a_{y,i} \hat{I}_{y,i} + a_{z,i} \hat{I}_{z,i} \right) \right. \\ \left. + \frac{1}{2\omega_0} \left(\sum_{i \in \text{reg}} a_{x,i} \hat{I}_{x,i} + a_{y,i} \hat{I}_{y,i} + a_{z,i} \hat{I}_{z,i} \right)^2 \right], \end{aligned} \quad (2.17)$$

with

$$\begin{aligned} a_{x,i} &= \frac{\mu_0 \mu_B \mu_N g_z}{4\pi |\mathbf{r}_i|^3} 3\hat{r}_{x,i} \hat{r}_{z,i} g_{vx}, \\ a_{y,i} &= \frac{\mu_0 \mu_B \mu_N g_z}{4\pi |\mathbf{r}_i|^3} 3\hat{r}_{y,i} \hat{r}_{z,i} g_{vy}, \\ a_{z,i} &= \frac{\mu_0 \mu_B \mu_N g_z}{4\pi |\mathbf{r}_i|^3} (3\hat{r}_{z,i}^2 - 1) g_{vz}. \end{aligned} \quad (2.18)$$

The first term represents the detuning of the ^{171}Yb due to magnetic fields. The second term describes the induced interaction between ^{171}Yb and ^{51}V . The third

term corresponds to the ^{171}Yb mediated interaction between ^{51}V . By performing a unitary transformation on the second term, we can obtain:

$$\hat{H}_{\text{DD}} = B_{\text{drive}} \tilde{S}_z (a_x \hat{I}_x^{(i)} + a_z \hat{I}_z^{(i)}), \quad (2.19)$$

with

$$\begin{aligned} a_x^2 &= a_{x,i}^2 + a_{y,i}^2 \\ a_z &= a_{z,i}. \end{aligned} \quad (2.20)$$

2.4.2 Nuclear Spin Polarization and Control Sequence (ZenPol)

We employ Hamiltonian engineering to design pulse sequences that enable coherent control of the ^{51}V nuclear spins in the spin bath. Several challenges must be addressed in this process. First, the nuclear spin bath contains multiple ^{51}V nuclei. Second, it is necessary to cancel out the detuning of ^{171}Yb caused by the Overhauser field (the first term in Equation 2.17). Third, we must suppress the contribution from the Overhauser-induced coupling between ^{171}Yb and ^{51}V spins (part of the second term in Equation 2.17) while preserving the desired driving term B_{drive} . The effects of the engineered pulse sequence can be analyzed using average Hamiltonian theory [125]. The figures in this section is reproduced from [101].

Figure 2.14 illustrates the engineered pulse sequence for coherent control of ^{51}V nuclear spins. The ^{171}Yb spin is toggled via the applied pulses to achieve dynamic decoupling from unwanted noise while enabling selective interactions. The square-wave RF field induces controlled dipolar interactions on ^{171}Yb that are distinct from the stochastic Overhauser fields.

In the toggling frame picture [126], the transformation of the operator S_z under the pulse control unitary U_c is given by $S_z \rightarrow \tilde{S}_z = U_c^\dagger S_z U_c$. Over one sequence period, the toggling-frame Hamiltonian can be expressed as:

$$\begin{aligned} \hat{H}_{\text{tog}} &= \Delta(t) [f_x^{\text{OH}}(t) \hat{S}_x + f_y^{\text{OH}}(t) \hat{S}_y] + \Sigma_i Q(\hat{I}_z^{(i)})^2 \\ &\quad + B_z^{\text{OH}} [f_x^{\text{OH}}(t) \hat{S}_x + f_y^{\text{OH}}(t) \hat{S}_y] [a_x^{(i)} \hat{I}_x + a_z^{(i)} \hat{I}_z] \\ &\quad + B_z^{\text{RF}} [f_x^{\text{RF}}(t) \hat{S}_x + f_y^{\text{RF}}(t) \hat{S}_y] [a_x^{(i)} \hat{I}_x + a_z^{(i)} \hat{I}_z], \end{aligned} \quad (2.21)$$

where $\Delta(t) = \frac{\mu_B^2 g_z^2 (B^{\text{RF}}(t) + B_{\text{OH}})^2}{2\omega_0}$, $f_{x,y}^{\text{OH}}(t)$ represent the time-dependent modulations of the ^{171}Yb spin components induced by the pulse sequence, and $f_{x,y}^{\text{RF}}(t) = B_{\text{RF}}(t) \times f_{x,y}^{\text{OH}}(t)$ (Figure 2.14(a)).

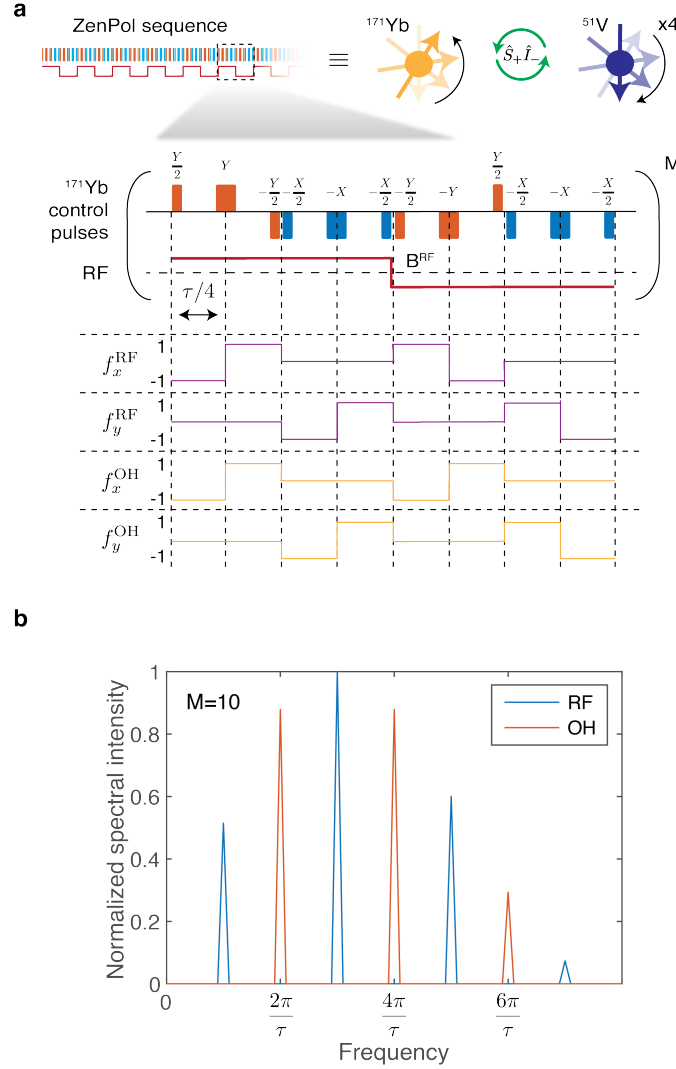


Figure 2.14: Details of the ZenPol pulse sequence. (a) The sequence consists of a series of π and $\pi/2$ pulses applied to the ^{171}Yb spin, combined with a square-wave RF field. This design dynamically decouples the ^{171}Yb central spin while generating a spin-preserving interaction with the ^{51}V nuclear spin ensemble. (b) Fourier spectra of the modulation functions $f_{x,y}^{\text{OH}}(t)$ (red) and $f_{x,y}^{\text{RF}}(t)$ (blue) highlighting the resonance conditions.

By focusing on a single ^{51}V nuclear transition and moving to its rotating frame, the Overhauser-related contributions average out to zero provided the modulation functions $f_{x,y}^{\text{OH}}(t)$ avoid resonance overlaps with nuclear transitions, as illustrated in Figure 2.14(b). Under the resonance condition, the effective Hamiltonian reduces to:

$$\hat{H}_{\text{avg}} \propto B^{\text{RF}} \sum_i (\hat{S}_+ \hat{I}_-^{(i)} + \hat{S}_- \hat{I}_+^{(i)}). \quad (2.22)$$

which describes a coherent, spin-preserving flip-flop interaction between the ^{171}Yb electron spin and the ^{51}V nuclear spin ensemble. Detailed derivations and a comprehensive discussion of the sequence can be found in [101]. ^{171}Yb -mediated indirect interactions between distinct ^{51}V nuclei are neglected here, as they are effectively cancelled by toggling the ^{171}Yb .

2.4.3 Nuclear Spin Polarization and Control

We performed high-resolution nuclear spin spectroscopy using the ZenPol sequence (Figure 2.15). When the sequence is resonant with specific ^{51}V nuclear spin transitions, it induces interaction between ^{171}Yb and ^{51}V nuclei. Under these conditions, ^{171}Yb becomes entangled with the nuclear spin ensemble, resulting in a measured population of 50% for the ^{171}Yb qubit. The resonances appear as dips in Figure 2.15, directly revealing the ^{51}V energy structure.

Because ^{51}V is a spin-7/2 nucleus, its quadrupolar interaction in YVO_4 produces four non-degenerate transitions at zero magnetic field, as determined by Equation 2.11 and illustrated in Figure 2.13. These transitions, labeled by their respective frequency spacings, obey $\omega_c = 3\omega_a$ and $\omega_b = 2\omega_a$. In each resonance condition in Figure 2.15, we observe two distinct peaks: one associated with “register” spins (i.e., second-shell ^{51}V nuclei directly coupled to ^{171}Yb , with $\omega_c = 991$ kHz), and another corresponding to more distant “bath” spins (with $\omega_c = 1028$ kHz). The register spins are shifted due to the frozen core effect, and they constitute a deterministic and reproducible resource, which is a crucial advantage for scalability. Further analysis confirms that the register comprises four equidistant second-shell ^{51}V spins per ^{171}Yb ion.

To polarize and control these nuclear spins, we tune the ZenPol sequence to the ω_b and ω_c transitions (using $k = 3$ and $k = 5$ resonance conditions, respectively), as highlighted in Figure 2.15. Note that the ω_a transitions are spectrally overlapped with other transitions when applying ZenPol, so polarization of the manifolds relies on nuclear spin T_1 relaxation processes.

With register spins polarized into the $|7/2\rangle$ state, we demonstrate coherent spin transfer between ^{171}Yb and the ^{51}V ensemble using the ZenPol sequence on the ω_c transition. By initializing ^{171}Yb in $|1\rangle_g$, we observe coherent exchange of spin excitation, so-called magnon oscillations, between the electron and the nuclear spin register (Figure 2.16a). The excitation delocalizes across the four register spins, creating an entangled W state,

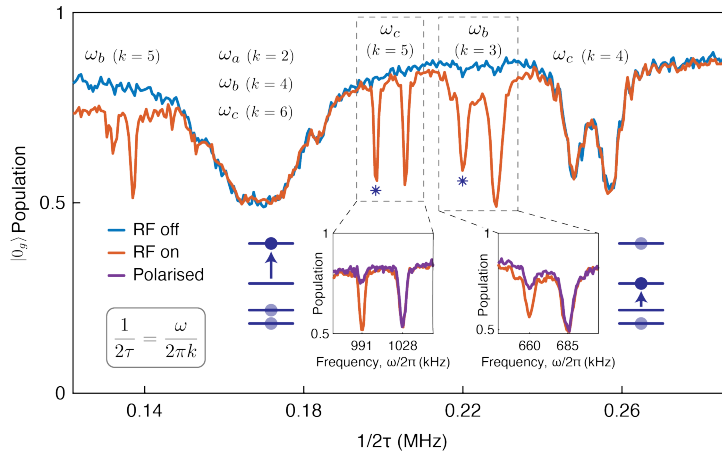


Figure 2.15: ZenPol spectroscopy. Data shown for 30 repetitions of the sequence with $B^{\text{RF}} = 0.6$ G. Resonance condition is $1/2\tau = \omega/2\pi k$. Odd- k resonances (RF-induced) and even- k resonances (Overhauser-induced) are visible; red and blue denote measurements with and without RF field. Each resonance splits into two peaks, corresponding to register and bath ^{51}V spins. The inset compares spectra before (red) and after (purple) register polarization.

$$|W_v\rangle = (|\uparrow\downarrow\downarrow\downarrow\rangle + |\downarrow\uparrow\downarrow\downarrow\rangle + |\downarrow\downarrow\uparrow\downarrow\rangle + |\downarrow\downarrow\downarrow\uparrow\rangle)/2. \quad (2.23)$$

We further confirm the number of register spins by selectively preparing a single ^{51}V spin in the $|5/2\rangle$ state and performing a ZenPol swap, which is done by first polarizing into $|3/2\rangle$ and injecting a spin into $|5/2\rangle$ using ZenPol. The oscillation frequency is reduced by a factor of two compared to the fully polarized case (Figure 2.16b), consistent with the expected $\sqrt{N}$ enhancement for $N = 4$ identical register spins coupled collectively to the central electron.

This magnon-like mode enables quantum state storage: by applying ZenPol, we can swap an arbitrary qubit superposition $(\alpha |0\rangle_g + \beta |1\rangle_g)$ from ^{171}Yb into the collective state of the nuclear register,

$$(\alpha |0\rangle_g + \beta |1\rangle_g) |0_v\rangle \rightarrow |0\rangle_g (\alpha |0_v\rangle + \beta |W_v\rangle) \quad (2.24)$$

where $|0_v\rangle = |\downarrow\downarrow\downarrow\downarrow\rangle$. This protocol provides a robust quantum memory due to the long coherence times of the nuclear spins and illustrates the power of engineered electron-nuclear spin coupling within YVO_4 .

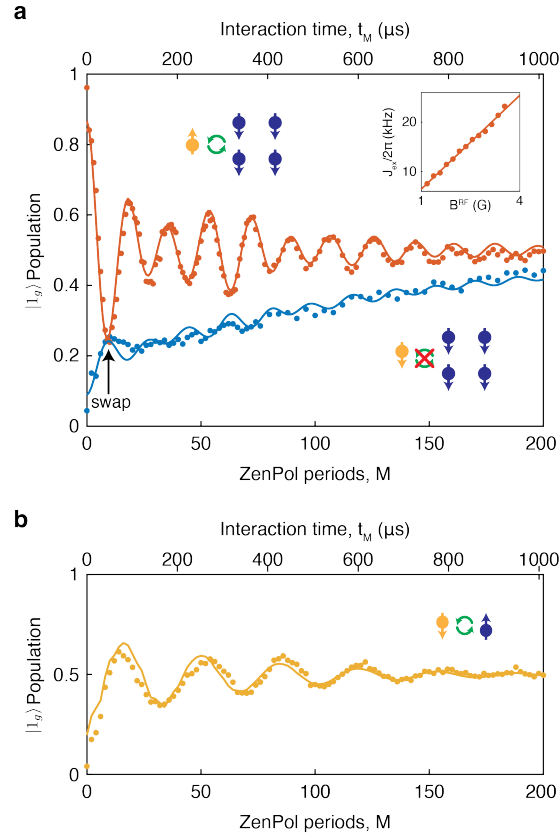


Figure 2.16: Magnon oscillations between ^{171}Yb and ^{51}V . (a) Coherent exchange (flip-flop) between ^{171}Yb and the ^{51}V register with all nuclear spins polarized. The oscillation frequency depends on the number of register spins and the RF drive strength (see inset). The red curve is measured with ^{171}Yb initialized into $|1\rangle_g$ and the blue curve is measured with ^{171}Yb initialized into $|0\rangle_g$. (b) ZenPol swap between ^{171}Yb and a single excited ^{51}V spin in the ω_c manifold. Reduced frequency confirms collective $\sqrt{N}$ scaling for four register spins.

2.4.4 Coherence and Lifetime of the Nuclear Spins

Two key properties of the nuclear spin storage are its coherence time (T_2) and lifetime (T_1). The coherence time is measured by initializing the ^{51}V nuclear ensemble into a superposition state and, after a variable delay, swapping the quantum information back onto ^{171}Yb using ZenPol for readout. Figure 2.17 summarizes these coherence measurements. The Ramsey coherence time is measured to be $58 \pm 4 \mu\text{s}$, limited by decoherence arising from the Overhauser-field-induced ^{171}Yb dipole. By applying motional narrowing (i.e., inserting periodic π pulses on ^{171}Yb during the wait time), the Overhauser-induced dephasing is suppressed and the storage coherence extends to $225 \pm 9 \mu\text{s}$. Further, by using dynamical decoupling with two π pulses applied

directly to ^{51}V , the coherence time increases to $760 \pm 14 \mu\text{s}$. Details of the ^{51}V π pulse generation are described in Chapter 4.

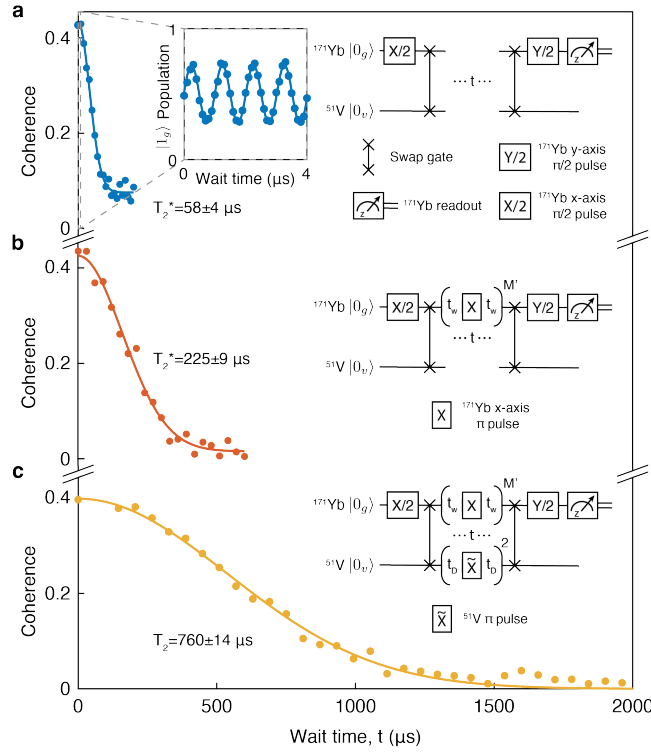


Figure 2.17: Coherence of ^{51}V storage. (a) Ramsey coherence time of ^{51}V storage, limited by Overhauser-field-induced ^{171}Yb dipole fluctuations ($T_2^* = 58 \pm 4 \mu\text{s}$). (b) Extended coherence using motional narrowing pulses on ^{171}Yb ($T_2 = 225 \pm 9 \mu\text{s}$). (c) Further extension with ^{51}V dynamical decoupling ($T_2 = 760 \pm 14 \mu\text{s}$).

We also measure population lifetimes for both the collective $|W_v\rangle$ and fully polarized $|0_v\rangle$ nuclear states (Figure 2.18). The $|W_v\rangle$ state has a lifetime limited by its coherence, as decoherence can lead to population leakage into other W states; thus, the T_1 for $|W_v\rangle$ is roughly half the corresponding coherence time in Figure 2.17. In contrast, the $|0_v\rangle$ state exhibits a much longer T_1 , measured to be $0.54 \pm 0.08 \text{ s}$.

2.5 Summary

In this chapter, we have presented a comprehensive experimental characterization of single $^{171}\text{Yb}^{3+}$ ions in YVO_4 , establishing this platform as a leading candidate for the development of quantum network nodes. We demonstrated how the strategic integration of these ions with nanophotonic crystal cavities overcomes their intrinsically weak optical transitions, enabling an efficient spin-photon interface.

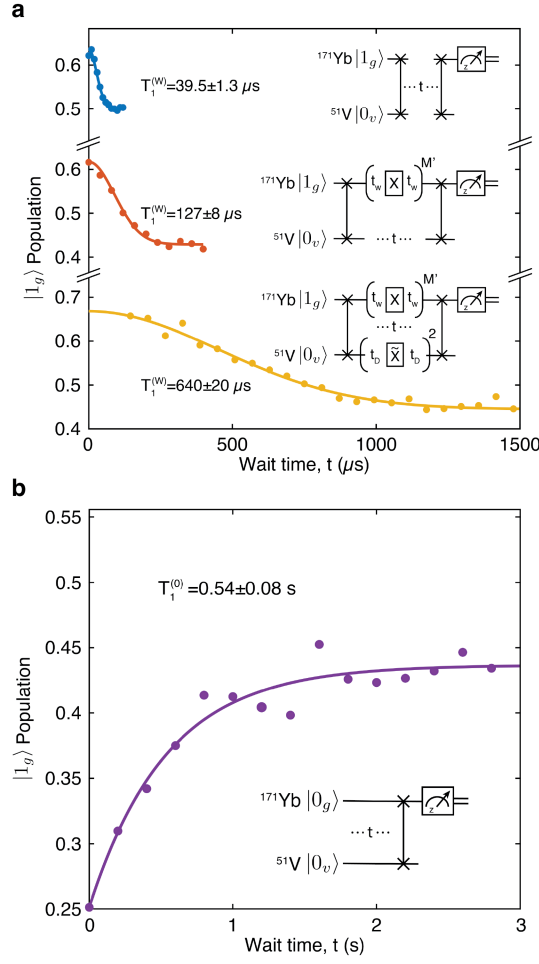


Figure 2.18: Lifetime of ^{51}V states. (a) Measured lifetimes of $|W_v\rangle$ for three different decoupling protocols (as in Figure 2.17), indicating that $|W_v\rangle$ decay is coherence-limited. (b) Population lifetime (T_1) of the $|0_v\rangle$ state, measured to be $0.54 \pm 0.08 \text{ s}$.

Our results include the demonstration of high-purity single-photon emission, millisecond-scale spin coherence times achieved through dynamical decoupling, and a single-qubit gate fidelity of 0.99975%. These metrics collectively satisfy the stringent requirements for a high-performance quantum bit. A key contribution of this work is the transformation of the surrounding ^{51}V nuclear spin bath from a decoherence source into a functional quantum resource. Through advanced Hamiltonian engineering, we achieved coherent control over a register of proximal nuclear spins,

demonstrating their polarization and the storage and retrieval of quantum states via collective magnon excitations.

Taken together, the results presented in this chapter validate the $^{171}\text{Yb}:\text{YVO}_4$ system as a robust and versatile platform, integrating a high-fidelity qubit, a coherent optical interface, and an on-chip quantum memory—the three essential components of a scalable quantum network node.

Chapter 3

MULTIPLEXED QUANTUM NETWORKING USING $^{171}\text{Yb}:\text{YVO}_4$

One of the essential building blocks of quantum networking is the ability to distribute entanglement remotely between separate devices. In this chapter, we present experiments demonstrating remote entanglement distribution across two independent $^{171}\text{Yb}:\text{YVO}_4$ devices. By leveraging the presence of multiple spectrally resolvable ^{171}Yb ions, separated by several hundred megahertz within each device, we implement multiplexed entanglement protocols involving two pairs of ions, resulting in an increased rate of entanglement generation. Additionally, we utilize three ions to prepare highly entangled W states that involve more than two nodes.

The chapter begins with a description of the entanglement protocol. We then introduce the experimental setups used in these demonstrations. Finally, we present results for both heralded bipartite entanglement and multi-ion (beyond bipartite) entanglement generation and verification. Figures in the Chapter is reproduced from [80]. Building upon the foundational system characterization presented in previous chapters, this chapter details the first experimental demonstrations of remote entanglement distribution, multiplexing, and multipartite entanglement using single rare-earth ions.

3.1 Remote Entanglement Protocol

Remote entanglement distribution requires that the spin of each quantum node is entangled with an emitted photon, after which interfering the photons from separate devices erases which-path information and enables heralded entanglement generation. There are several approaches for encoding quantum information onto photons and generating spin-photon entanglement, including the use of number (Fock) states, time-bin encoding, polarization, dual-rail (path) encoding, or frequency channels [127]. Entanglement protocols can broadly be classified as either emission-based or reflection-based: emission-based protocols rely on photons generated via spontaneous emission from the qubit, while reflection-based protocols exploit the state-dependent reflection spectrum of a cavity-qubit system, which requires high optical cooperativity. Crucially, achieving high-fidelity remote entanglement over long distances demands both long-lived spin coherence and highly

coherent optical transitions in the underlying quantum emitter.

3.1.1 Single and Two Photon Protocols

Based on the cavity QED parameters in our system (see Section 3.2), the optical cooperativities of our devices are not high enough to support reflection-based entanglement protocols, which typically require strong cavity-emitter coupling. Therefore, in this work, we focus on emission-based protocols. Specifically, we use the photon number basis, where $|0\rangle_p$ denotes the vacuum state and $|1\rangle_p$ denotes a single-photon Fock state. We discuss two such protocols: one that heralds entanglement via single-photon detection, and another that relies on the detection of two photons [128, 129].

In the following, we consider a pair of identical emitters, each assumed to be perfectly coherent and independently controllable. Each emitter has a spin qubit transition between $|0\rangle_g$ and $|1\rangle_g$, and an optically allowed transition connecting $|1\rangle_g$ to the excited state $|e\rangle$.

In the single-photon heralding scheme, both emitters are initially prepared in the $|0\rangle_g$ ground state. A weak pulse is then applied to induce a small spin rotation, preparing each emitter into a superposition:

$$(\sqrt{1-\alpha}|0\rangle_g + \sqrt{\alpha}|1\rangle_g) \otimes (\sqrt{1-\alpha}|0\rangle_g + \sqrt{\alpha}|1\rangle_g), \quad (3.1)$$

with a small excitation amplitude $\alpha \ll 1$. Optical π pulses are applied next to selectively excite the $|1\rangle_g \rightarrow |e\rangle$ transition:

$$(\sqrt{1-\alpha}|0\rangle_g + \sqrt{\alpha}|e\rangle) \otimes (\sqrt{1-\alpha}|0\rangle_g + \sqrt{\alpha}|e\rangle). \quad (3.2)$$

After spontaneous emission from $|e\rangle \rightarrow |1\rangle_g$, and assuming indistinguishability, the emitted photons are coherently combined into a single spatial mode. The resulting joint spin-photon state is:

$$(1-\alpha)|0\rangle_g|0\rangle_g|0\rangle_p + \sqrt{1-\alpha}\sqrt{\alpha}(|0\rangle_g|1\rangle_g + |1\rangle_g|0\rangle_g)|1\rangle_p + \alpha|1\rangle_g|1\rangle_g|2\rangle_p, \quad (3.3)$$

where $|n\rangle_p$ denotes an n -photon state. Upon a single-photon detection, assuming finite detection efficiency η , the vacuum term $|0\rangle_g|0\rangle_g$ is carved out, and the $|1\rangle_g|1\rangle_g$ component can be neglected due to the $\alpha^2 \ll 1$ suppression. The resulting spin state is:

$$\frac{1}{\sqrt{2}}(|0\rangle_g|1\rangle_g + |1\rangle_g|0\rangle_g). \quad (3.4)$$

The detection of a single photon thus heralds the successful creation of the target entangled Bell state. The heralding probability is approximately $2\alpha^2\eta$. The main drawback of the single-photon scheme is the requirement for phase stabilization between the two optical paths, and a fidelity that is limited by finite α [130].

The two-photon protocol, originally proposed by Barrett and Kok [128], offers higher state purity and does not require path phase stabilization. It begins with each emitter prepared in an equal spin superposition:

$$\frac{1}{2}(|0\rangle_g + |1\rangle_g) \otimes (|0\rangle_g + |1\rangle_g). \quad (3.5)$$

Optical π pulses are applied to excite $|1\rangle_g \rightarrow |e\rangle$:

$$\frac{1}{2}(|0\rangle_g + |e\rangle) \otimes (|0\rangle_g + |e\rangle). \quad (3.6)$$

Following spontaneous emission, the joint spin-photon state becomes:

$$\frac{1}{2}(|0\rangle_g |0\rangle_g |0\rangle_p + |0\rangle_g |1\rangle_g + |1\rangle_g |0\rangle_g) |1\rangle_p + |1\rangle_g |1\rangle_g |2\rangle_p). \quad (3.7)$$

If only one photon is detected, the $|0\rangle_g |0\rangle_g$ term is eliminated. For low detection efficiency η , we cannot project out $|1\rangle_g |1\rangle_g$ with high fidelity in a single detection event:

$$\frac{1}{4}((|0\rangle_g |1\rangle_g + |1\rangle_g |0\rangle_g)(\langle 0|_g \langle 1|_g + \langle 1|_g \langle 0|_g) + 2|1\rangle_g |1\rangle_g \langle 1|_g \langle 1|_g). \quad (3.8)$$

To mitigate this, a spin π pulse is applied to invert $|0\rangle_g \leftrightarrow |1\rangle_g$, followed by a second optical π pulse. This drives the joint system into:

$$\frac{1}{4}((|0\rangle_g |e\rangle + |e\rangle |0\rangle_g)(\langle 0|_g \langle e| + \langle e| \langle 0|_g) + 2|0\rangle_g |0\rangle_g \langle 0|_g \langle 0|_g). \quad (3.9)$$

Upon detecting a second photon, the $|0\rangle_g |0\rangle_g$ component is excluded, and the final projected spin state is:

$$\frac{1}{2}(|0\rangle_g |1\rangle_g + |1\rangle_g |0\rangle_g)(\langle 0|_g \langle 1|_g + \langle 1|_g \langle 0|_g). \quad (3.10)$$

The detection of a second photon projects the system into the desired entangled state, completing the protocol. The success probability of the two-photon protocol is approximately $\eta^2/2$, which becomes much lower than that of the single-photon scheme when the detection efficiency η is small. Nonetheless, the two-photon protocol features inherently higher fidelity and robustness to optical path-phase fluctuations [129].

3.1.2 Dynamic Rephasing and Feedforward Phase Compensation

To enable remote entanglement distribution in our system, several key challenges must be addressed. First, ^{171}Yb in YVO_4 lacks a first-order DC Stark shift, meaning the optical transition frequencies of individual ions cannot be externally tuned. As a result, frequency mismatches caused by static inhomogeneous broadening, on the order of 200 MHz, must be accounted for. Second, the optical coherence time (≈ 300 ns) is significantly shorter than the limit set by spontaneous emission. For this system, the fundamental coherence limit is twice the excited-state lifetime (≈ 4 μs), implying that the measured optical Ramsey time is dephasing-limited due to spectral diffusion and other noise sources.

To address both issues, we developed a dynamic rephasing protocol specifically designed to enable high-fidelity entanglement distribution in the presence of spectral inhomogeneity and optical dephasing [80, 131, 132].

The pulse sequence is illustrated in Figure 3.1. Because the protocol is sensitive to optical path phase differences between the two devices, it begins with a phase stabilization pulse ($Z_{\Delta\Phi}$). Next, a single-photon interference protocol is performed: each emitter undergoes a weak spin rotation (via an α -pulse on the qubit transition) followed by an optical π pulse. If a single photon is detected at time t_0 , the entanglement is heralded. However, because the two emitters have distinct optical transition frequencies (differing by $\Delta\omega_0$), the resulting spin state acquires a phase depending on t_0 :

$$\frac{1}{\sqrt{2}}(e^{-i\Delta\omega_0 t_0} |0\rangle_g |1\rangle_g + |1\rangle_g |0\rangle_g). \quad (3.11)$$

Here, $\Delta\omega_0$ fluctuates in time due to spectral diffusion, leading to decoherence. To mitigate this, a dynamic rephasing sequence is applied: spin π pulses invert the qubit states, and optical π pulses, separated by the original emission time interval t_0 , re-excite the ions and drive them back to the ground state via stimulated emission. This acts as an optical echo that cancels the spectral diffusion and corrects for spin dephasing. Importantly, the delay time is computed in real time and updated for each heralding.

After rephasing, the resulting two-qubit state becomes:

$$\frac{1}{\sqrt{2}}(e^{-i\tilde{\Delta\omega}_0 t_0} |1\rangle_g |0\rangle_g + |0\rangle_g |1\rangle_g), \quad (3.12)$$

where $\tilde{\Delta\omega}_0$ is the frequency difference between the optical drive lasers. Unlike the original emitter frequency difference $\Delta\omega_0$, this quantity is defined and stabilized in

the laser systems, and the associated phase $\Delta\tilde{\omega}_0 t_0$ is known. As a result, we can apply a virtual Z gate to each emitter, by adjusting the phase of subsequent spin control pulses, to remove this deterministic phase and recover a coherent Bell state. However, the main remaining limitation of the protocol is spontaneous emission that may occur during the rephasing process. This introduces incoherent errors that cannot be corrected by phase compensation and reduce the final fidelity of the heralded entangled state. Ultimately, by combining photon detection timing and knowledge of the optical drive frequencies, we can deterministically correct the relative phase and herald the target Bell state:

$$\frac{1}{\sqrt{2}}(|1\rangle_g |0\rangle_g + |0\rangle_g |1\rangle_g). \quad (3.13)$$

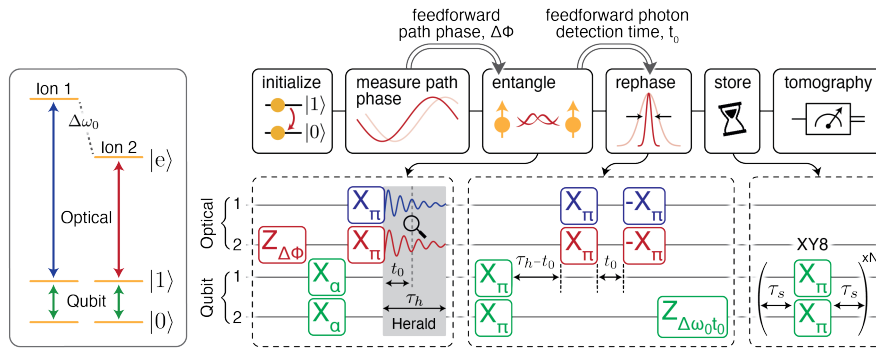


Figure 3.1: Gate sequence for the entanglement distribution protocol. (Left) Energy level diagrams for two emitters with detuned optical transition frequencies ($\Delta\omega_0$). (Right) Schematic of the control sequence. The protocol begins with optical path phase stabilization ($Z_{\Delta\Phi}$), followed by the single-photon heralding process. Upon photon detection at time t_0 , dynamic rephasing and phase compensation sequences including spin and optical π pulses are applied to cancel phase errors from spectral diffusion and emitter frequency differences.

3.2 Experimental Setup

In this section, we describe the experimental setup used to generate entanglement between ions located in separate devices (Figure 3.2). The setup builds upon the single-ion configuration described in Section 2.2.3, with several added components to support remote entanglement and phase stabilization.

An additional laser at 987.9 nm (L2, Moglabs Cateye), locked to a high-finesse reference cavity Pound-Drever-Hall (PDH) stabilization, is used for optical phase stabilization between devices. The A transition of the ions is addressed by a continuous-

wave titanium-sapphire laser (L1, M2 Solstis). The outputs of L1 and L2 are selected via AOM1 and AOM2, then split by beamsplitter BS1 to direct light to both devices.

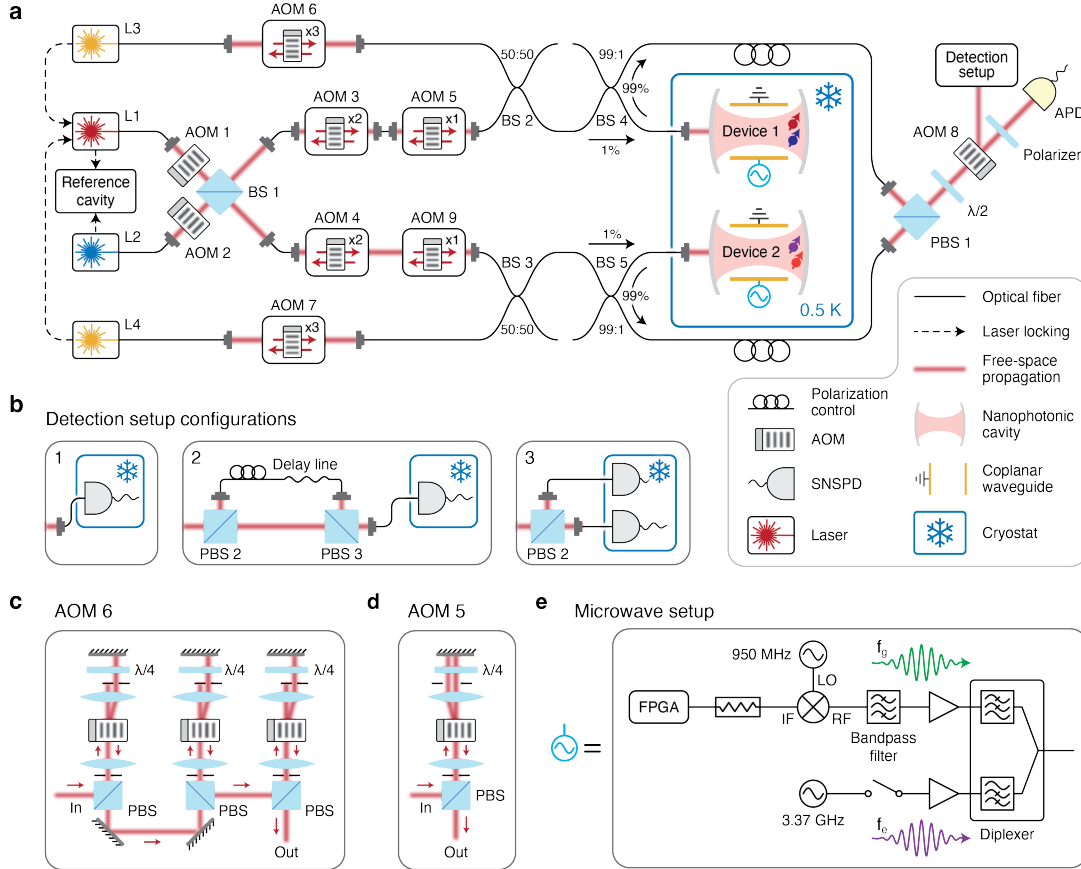


Figure 3.2: Experimental setup for remote entanglement distribution. (a) Optical setup: laser beams are routed to two independent $^{171}\text{Yb}:\text{YVO}_4$ devices, and emitted photons are collected and routed to various detection paths. (b) Detection configurations: Setup 1 is used for single-ion measurements, Setup 2 for bipartite entanglement heralding via interference, and Setup 3 for experiments involving more than two ions. (c) Three double-pass AOM setups used for controlling multiple ions within the inhomogeneous broadening range. (d) Bichromatic beam delivery setup for simultaneously addressing two ions. (e) Microwave control electronics for ground-state and excited-state spin manipulation.

AOM3 and AOM4 are each configured in a two-double-pass arrangement to serve as optical shutters for Device 1 and Device 2, respectively. AOM5 and AOM9 form bichromatic setups used for simultaneously addressing two ions within the same device. Additional frequency-offset-locked lasers (L3 and L4, Toptica DL Pro) are used to address the F transitions. To enable frequency-resolved control

over multiple ions within each device's 600 MHz ion frequency separations, these control beams are shaped with three double-pass AOM setups enabling flexible, frequency-selective addressing.

Both devices are housed inside the same cryostat but are optically and electronically isolated. Optical polarization within the collection paths is adjusted using electronic polarization controllers (OZ Optics EPC) and the outputs are combined via a polarizing beamsplitter (PBS1). AOM8 is used to toggle between the entanglement heralding detection setup and a secondary photodetector path (Avalanche Photodiode, Laser Components A-DUBE-S500-240) for measuring the interference-induced phase difference between optical paths.

Three detection setups are configured for different experimental needs (Figure 3.2(b)). Setup 1 contains a superconducting nanowire single-photon detector (SNSPD) and is used primarily for single-ion characterization. Setup 2, used for heralded bipartite entanglement distribution, includes a fiber delay line to compensate for the π phase shift introduced by path reflection. Photonic modes from the two devices, prepared in orthogonal polarizations, are mixed via a 45° polarization rotation and a polarizing beamsplitter (PBS2). The resulting spin-photon entangled state is:

$$\frac{1}{2}[(|10\rangle + |01\rangle e^{-i\Delta\omega_0 t}) |H\rangle + (|10\rangle - |01\rangle e^{-i\Delta\omega_0 t}) |V\rangle], \quad (3.14)$$

where $|H\rangle$ and $|V\rangle$ correspond to the photonic modes in the transmitted and reflected paths, respectively. By applying a delay of duration $\pi/\Delta\omega_0$, a relative phase shift transforms the state into:

$$\frac{1}{2}[(|10\rangle + |01\rangle e^{-i\Delta\omega_0 t})(|H\rangle + |V\rangle)], \quad (3.15)$$

allowing for probabilistic Bell state heralding. Setup 3 is designed for entanglement involving more than two ions, utilizing two SNSPDs after PBS2. Phase differences introduced by the optical paths are corrected via feedforward from the detection outcome: conditional spin phase adjustments (virtual Z gates) are applied based on which SNSPD registers a click.

The microwave control hardware is shown in Figure 3.2(e). Excited-state microwave pulses (f_e) are implemented as described in Section 2.2.3. However, ground-state microwave control is modified in this experiment due to the frequency limitations of the Quantum Machines OPX+ platform, whose maximum output frequency is below the spin transition frequency. To address this, a heterodyne setup is used to generate the control pulses. A waveform at approximately 275 MHz from the OPX+

is mixed with a 950 MHz local oscillator (Holzworth HS 9002A). The resulting signal is then bandpass filtered, and amplified (Amplifier Research 10U1000 for Device 1 and Minicircuits ZHL-20W-13SW+ for Device 2).

Detection signals from the SNSPDs and avalanche photodiode are amplified, and routed to analog input channels of the OPX system. One of the OPX ports is shared between the SNSPD and APD using a diplexer. The Quantum Machines OPX+ platform provides real-time conditional logic (feedforward) at microsecond timescales, which is essential for dynamic feedback during entanglement protocols.

3.2.1 Phase Stabilization

Single-photon heralding protocols require optical path-phase stability between emitters to maintain coherent interference [133]. This requirement becomes especially important in scenarios with low photon detection efficiency, where single-photon protocols offer higher entanglement heralding rates than two-photon schemes.

A 10 μs -long off-resonant pulse is used for stabilization. This minimizes heating of cryogenic components while allowing sufficient signal strength to detect the relative optical phase. The interference between the two beams is measured using an avalanche photodiode (APD), providing the optical phase difference:

$$\Delta\Phi_{\lambda_2} = 2\pi\Delta L/\lambda_2 \quad (3.16)$$

where ΔL is the optical path length difference and λ_2 is the wavelength of the phase-stabilization laser. If $\Delta\Phi_{\lambda_2}$ remains stable, the relative phase of the A transition drive at wavelength λ_1 is also stabilized, provided $\Delta\Phi_{\lambda_1} - \Delta\Phi_{\lambda_2}$ remains constant. For a typical path length difference of ~ 3 meters, this condition holds as long as the laser drift remains below 70 MHz between the two wavelengths.

To validate the stabilization protocol, we measure the temporal evolution of $\Delta\Phi_{\lambda_1}$ with and without feedback (Figure 3.3(b)). Without stabilization, the optical phase drifts significantly, with a single-exponential correlation decay time of approximately 282 μs (Figure 3.3(c)). During stabilization, the measured phase from the APD is used to feedforward a correction to the microwave phase driving the AOMs. This feedback ensures the optical pulses maintain a stable relative phase between devices.

To quantify residual error, we apply a probe pulse 30 μs after the stabilization pulse and monitor the phase stability over repeated trials (Figure 3.3(d)). The resulting

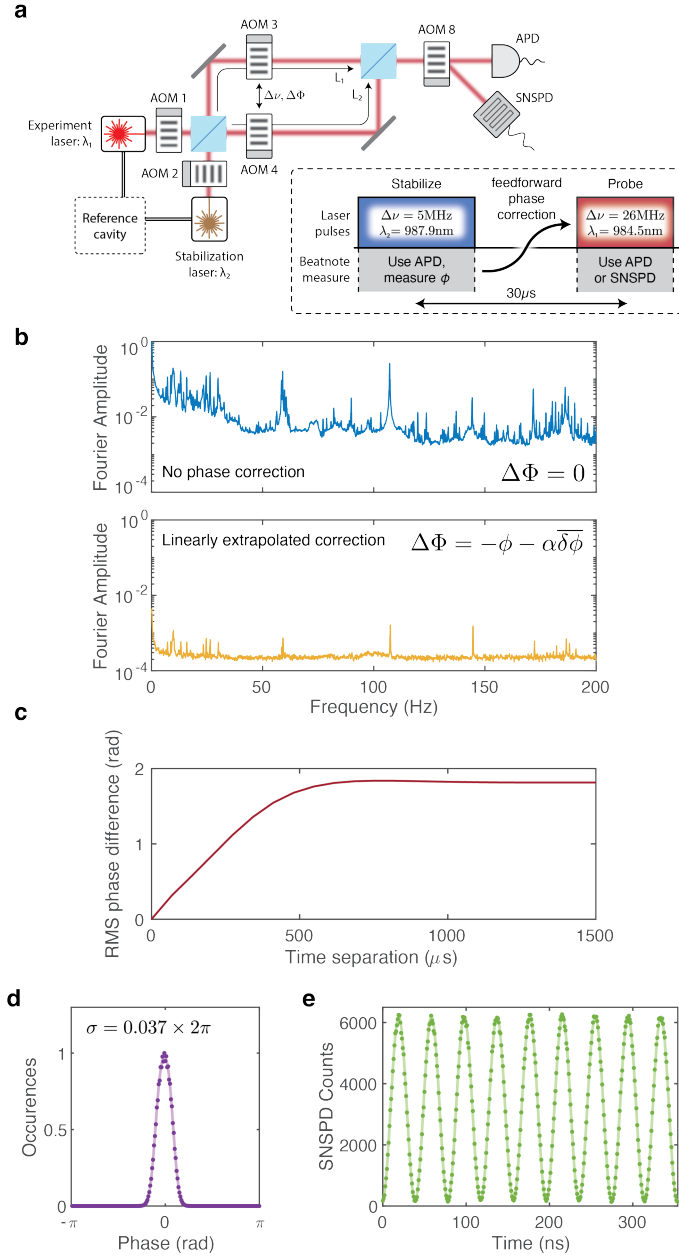


Figure 3.3: Phase stabilization between two optical paths. (a) Schematic of the experimental setup for phase stabilization and probing. A stabilization pulse is sent, and the relative phase shift is detected by comparing the two paths. (b) Comparison of the Fourier-transformed phase noise of the probe pulse with (yellow) and without (blue) active stabilization. (c) Phase stability without feedback, characterized by the RMS phase difference over varying delays. The $1/e$ correlation time is approximately $282\ \mu\text{s}$. (d) Phase histogram of the stabilized probe, showing a Gaussian distribution with a standard deviation of $2\pi \times 0.037$ radians. (e) Interference contrast between probe pulses detected via SNSPD, confirming high phase coherence (contrast = 0.944).

phase distribution has a standard deviation of $2\pi \times 0.037$ radians, demonstrating high stability suitable for entanglement generation experiments.

To verify that detection hardware does not limit visibility, we attenuate the probe pulses and record interference fringes directly using an SNSPD. The observed interference contrast is 0.944 (Figure 3.3(e)), indicating that SNSPD timing resolution is sufficient to resolve interference fringes.

3.2.2 Device Parameters

Devices used in this work are fabricated using the focused ion beam milling process described in Section 2.2.1. Key optical cavity parameters for the two devices used in the entanglement experiments are summarized in Table 3.1. Both cavities exhibit optical quality factors on the order of 10,000, with cavity-to-waveguide coupling ratio of approximately 0.1.

	Device 1	Device 2
Q	$(9.7 \pm 0.2) \times 10^3$	$(1.27 \pm 0.07) \times 10^4$
κ	$2\pi \times (31.4 \pm 0.8) \text{ GHz}$	$2\pi \times (24.4 \pm 1.3) \text{ GHz}$
$\kappa_{\text{in}}/\kappa$	0.117 ± 0.004	0.065 ± 0.004

Table 3.1: Optical cavity parameters for the two devices used in the experiments. Q is the quality factor, κ is the total energy decay rate of the cavity, and $\kappa_{\text{in}}/\kappa$ gives the ratio of the input coupling rate to the overall cavity linewidth.

Photoluminescence (PL) spectra for both devices are shown in Figure 3.4. Each spectrum highlights the ^{171}Yb ions used in the experiments, identified by different colors. These ions were selected based on spectral isolation from neighboring ions.

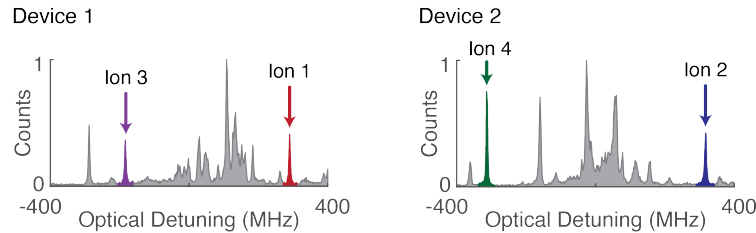


Figure 3.4: Photoluminescence spectra of the two devices. The ^{171}Yb ions used in the experiments are indicated by color peaks.

3.3 Ion Properties

In this section, we summarize the key physical properties of four individually addressed ^{171}Yb ions used across the two nanophotonic devices. These properties are presented in Table 3.2. The table includes both optical and spin coherence metrics, as well as photon emission characteristics that are relevant for remote entanglement and quantum networking protocols.

One particularly important observation is that all ions exhibit optical echo coherence times approaching the theoretical limit, set by twice the optical lifetime. This confirms that the optical decoherence is dominated by slowly varying spectral diffusion rather than irreversible dephasing mechanisms.

	Ion 1	Ion 2	Ion 3	Ion 4
Optical lifetime, $T_{1,o}$	$2.17 \pm 0.017 \mu s$	$0.985 \pm 0.003 \mu s$	$2.33 \pm 0.019 \mu s$	$1.115 \pm 0.010 \mu s$
Single photon efficiency, p_{det}	$(1.100 \pm 0.002) \times 10^{-2}$	$(4.60 \pm 0.05) \times 10^{-3}$	$(1.396 \pm 0.002) \times 10^{-2}$	$(5.210 \pm 0.013) \times 10^{-3}$
Qubit initialization fidelity	0.9976 ± 0.0003	0.9954 ± 0.0008	0.9979 ± 0.0002	0.9962 ± 0.0004
Resonant noise count rate	$8.3 \pm 0.5 \text{ Hz}$	$7.5 \pm 0.5 \text{ Hz}$	$1.8 \pm 0.2 \text{ Hz}$	$1.2 \pm 0.2 \text{ Hz}$
Optical Ramsey coherence, $T_{2,o}^*$	$310 \pm 20 \text{ ns}$	$220 \pm 10 \text{ ns}$	$328 \pm 16 \text{ ns}$	$221 \pm 7 \text{ ns}$
Optical linewidth (standard deviation), γ^*	$2\pi \times (0.73 \pm 0.05) \text{ MHz}$	$2\pi \times (1.02 \pm 0.05) \text{ MHz}$	$2\pi \times (0.69 \pm 0.03) \text{ MHz}$	$2\pi \times (1.02 \pm 0.03) \text{ MHz}$
Optical echo coherence, $T_{2,o}$	$3.66 \pm 0.12 \mu s$	$1.75 \pm 0.04 \mu s$	$3.89 \pm 0.19 \mu s$	$1.89 \pm 0.11 \mu s$
Qubit lifetime, $T_{1,s}$	$53 \pm 3 \text{ ms}$	$23 \pm 1 \text{ ms}$	$55 \pm 6 \text{ ms}$	$17.0 \pm 1.1 \text{ ms}$
Qubit XY8 coherence, $T_{2,s}$	$21.2 \pm 0.7 \text{ ms}$	$16.9 \pm 0.7 \text{ ms}$	$27 \pm 2 \text{ ms}$	$12.0 \pm 0.7 \text{ ms}$

Table 3.2: Physical properties of four ions in the two devices.

3.4 Bipartite Entanglement Heralding

3.4.1 Experiment Details

With the device architectures and entanglement protocol described in earlier sections, we implement heralded Bell state generation between Ion 1 and Ion 2 (identified in Figure 3.4). The full sequence is illustrated in Figure 3.5, and consists of several stages including initialization, path-phase stabilization, spin-photon entanglement, heralded photon detection, and real-time rephasing and measurement.

The sequence begins by initializing each ^{171}Yb ion into the qubit manifold ($|0\rangle_g$, $|1\rangle_g$) using optical pumping on the F_1 and F_2 transitions (Figure 3.5(b)). Subsequent optical and excited state π pulses prepare the qubits deterministically in the $|0\rangle_g$ state. A path-phase stabilization pulse is then applied, followed by the single-photon entanglement sequence. If a photon is detected—heralding successful entanglement between the two emitters—qubit dynamical decoupling pulses are applied during the processing delay to preserve coherence before real-time feedforward operations are performed.

These feedforward operations include dynamic rephasing on the optical transition, as well as phase compensation based on the known photon emission time and the frequency difference between the two ions (Figure 3.5(d)). To mitigate errors from remaining populations of the excited state after dynamic rephasing pulses, dynamical decoupling sequences incorporating both qubit and excited-state π pulses are applied immediately after the rephasing stage (see also Figure 3.5(d)).

In the final stage of measurement, the two ions are readout using conditional optical detection, as shown in Figure 3.6. Due to finite detection efficiency, the readout process is probabilistic: outcomes are accepted only if they meet specific photon detection criteria; all others are discarded. Pulse condition assignments and the detection logic are described in the accompanying table (Figure 3.6). The average overall readout efficiency is approximately 0.18, with an average inferred fidelity of 0.93.

Because the readout post-selects out events in which either ion occupies the metastable auxiliary state $|\text{aux}\rangle_g$, it is important to quantify residual population in that level following qubit initialization. This residual can be determined by comparing photon count rates during thermal equilibrium and after repumping via the F transition. The remaining population in $|\text{aux}\rangle_g$ is measured to be 0.023 ± 0.015 .

Because each ion has a small but non-zero probability ($\approx 1.2 \times 10^{-3}$) of relaxing to the $|\text{aux}\rangle_g$ state during each entanglement attempt, we repeat the heralding protocol up to 50 times before re-pumping the ion using the F transition.

Device asymmetries, such as differing photon collection efficiencies and optical lifetimes, require careful adjustment of qubit α pulse amplitudes to balance the spin-photon entanglement contributions. In this experiment, we set the spin rotation amplitude to $\alpha_1 = 0.062$ for Device 1 and $\alpha_2 = 0.078$ for Device 2 to equalize photon detection probabilities during the heralded interference process.

3.4.2 Parity Oscillations of the Heralded Bell States

Based on the protocols described in earlier sections, we measure the X-basis parity of the two-ion system at various points during the entanglement sequence. Specifically, parity measurements are performed immediately after heralded photon detection (yellow trace), after dynamic optical rephasing (red trace), and after phase compensation via virtual Z rotation (blue trace), as shown in Figure 3.7.

To measure X-basis parity, both ions are rotated into the X basis prior to readout. The parity is then computed as: $P_{00} + P_{11} - P_{10} - P_{01}$, where P_{ab} is the probability of measuring the two ions in the state $|ab\rangle$, with $a, b \in 0, 1$. Assuming negligible coherence between $|00\rangle$ and $|11\rangle$, the X-basis parity serves as a direct probe of the coherence between $|01\rangle$ and $|10\rangle$, which is the relevant off-diagonal term in the target Bell state.

In Figure 3.7, we observe that immediately after heralding (yellow trace), the parity exhibits clear oscillations at the frequency offset between the two ions' optical transitions, approximately 31 MHz. The oscillation contrast decays with a time constant of 200 ns, limited by the optical Ramsey coherence times of the emitters. After applying dynamic rephasing (red trace), the coherence is preserved for significantly longer durations, well beyond the optical Ramsey coherence times. This improvement results from the active cancellation of slow frequency drifts due to spectral diffusion via real-time rephasing on the optical transitions.

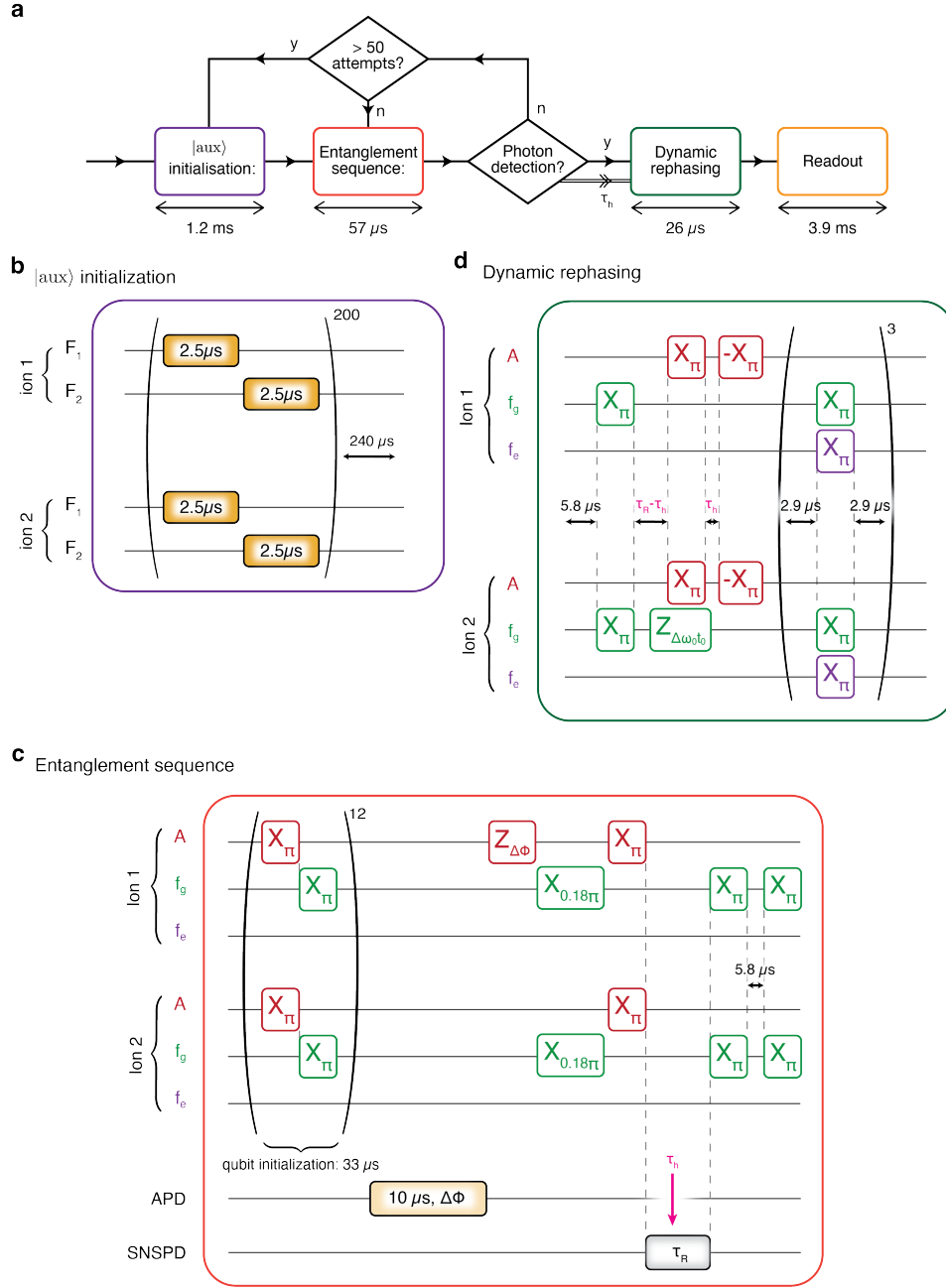


Figure 3.5: Detailed entanglement heralding sequence. (a) Flowchart of the entanglement protocol: ^{171}Yb ions are first initialized into the qubit manifold, before which the heralding sequence is executed. Detection of a herald photon triggers dynamic rephasing and state readout. (b) Initialization into the qubit manifold using optical pumping on the F_1 and F_2 transitions. (c) Entanglement heralding sequence, including qubit initialization, path-phase stabilization, and single-photon detection. (d) Upon photon detection, dynamic rephasing on the optical transition and feedforward phase compensation, dependent on the photon emission time and ion frequency difference, are applied in real time.

Readout

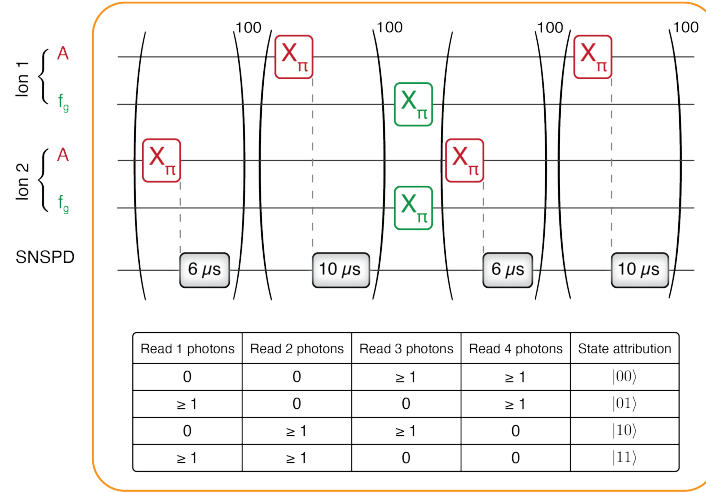


Figure 3.6: Readout sequence following entanglement heralding. Due to finite detection efficiency, the readout is probabilistic: outcome assignments are made according to the logic table provided, and cases not meeting the criteria are discarded.

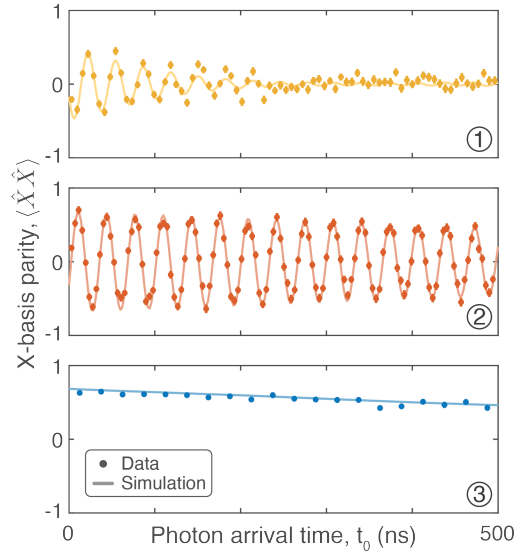


Figure 3.7: X-basis parity oscillations measured at different stages of the entanglement protocol. Yellow: oscillations directly after heralding, limited by optical Ramsey coherence. Red: extended coherence following dynamic optical rephasing. Blue: phase-compensated parity showing suppressed oscillations, corresponding to a stationary Bell state. Dots represent experimental data; solid lines are simulations based on measured system parameters.

Finally, after applying phase compensation, implemented by shifting the spin control frame in real-time to correct the phase caused by frequency difference between two ions, the parity oscillations are fully removed (blue trace), yielding a static parity signal consistent with a deterministic Bell state. A small residual decay persists, which we attribute to spontaneous emission events occurring during the optical rephasing interval; this decay is fundamentally limited by the optical lifetime of the ions.

3.4.3 Quantum State Tomography of the Heralded Bell States

We characterize the heralded Bell state generated between Ion 1 and Ion 2 using maximum likelihood quantum state tomography (QST) [134]. The quantum state is reconstructed from measurements performed in a complete set of nine two-qubit Pauli bases. Each measurement consists of photon counts in the four computational basis states, from which we estimate the density matrix. Figure 3.8 shows the reconstructed state obtained using a 500 ns photon detection window, yielding a fidelity of 0.723 ± 0.007 with respect to the ideal Bell state. The corresponding rate for heralded entanglement is 3.1 Hz.

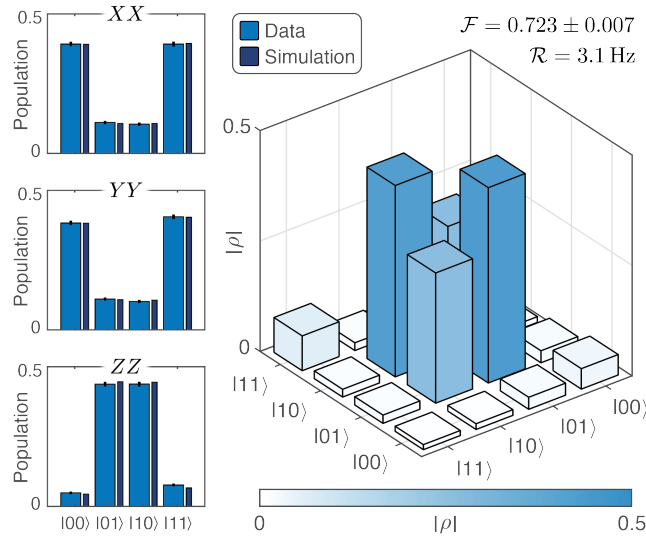


Figure 3.8: Quantum state tomography of the heralded entangled state. The state is reconstructed from measurements in all 9 two-qubit Pauli bases. Left: measured two-qubit correlations in the XX, YY, and ZZ bases. Right: absolute value of the reconstructed density matrix. The reconstructed state has a fidelity of 0.723 ± 0.007 and is obtained with a 500 ns heralding window, corresponding to an entanglement rate of 3.1 Hz.

To reconstruct the two-qubit density matrix, we measure the system in each of the nine two-qubit Pauli bases. Each measurement produces four outcome photon numbers: $N_{ab}^{(ij)}$ for $a, b \in 00, 01, 10, 11$, where $i, j \in X, Y, Z$ label the basis for Ion 1 and Ion 2, respectively. These counts correspond to the number of events where Ion 1 is found in state $|a\rangle$ and Ion 2 in $|b\rangle$. The full set of measured values is presented in Table 3.3 for a 500 ns photon acceptance window.

We model the probability of observing a given set of counts as a multinomial distribution derived from a trial density matrix $\hat{\rho}$. For a given basis (i, j) , the likelihood function is [53]:

$$\mathcal{L}^{(ij)}(\hat{\rho}) = \frac{N^{(ij)}!}{N_{00}^{(ij)}!N_{01}^{(ij)}!N_{10}^{(ij)}!N_{11}^{(ij)}!} \times \left[n_{00}^{(ij)} \right]^{N_{00}^{(ij)}} \left[n_{01}^{(ij)} \right]^{N_{01}^{(ij)}} \left[n_{10}^{(ij)} \right]^{N_{10}^{(ij)}} \left[n_{11}^{(ij)} \right]^{N_{11}^{(ij)}}, \quad (3.17)$$

where $N^{(ij)} = \sum_{a \in \{00, 01, 10, 11\}} N_a^{(ij)}$ is the total number of counts for a given basis and $\{n_a^{(ij)}\}$ are the predicted outcome probabilities derived from the model density matrix $\hat{\rho}$ projected into the (i, j) basis.

To incorporate readout imperfections, we apply a correction using a measured readout transformation matrix $R_{a,b}$, so that the predicted probabilities $\{p_a^{(ij)}\}$ are given by: $p_a^{(ij)} = \sum_{b \in \{00, 01, 10, 11\}} R_{a,b} n_b^{(ij)}$. This transformation accounts for readout infidelities and is obtained from independent calibration using well-prepared computational basis states. For example, the predicted population in the $|00\rangle$ outcome when measuring in the XX basis is:

$$p_{00}^{(XX)} = \text{Tr}\{|00\rangle\langle 00| \hat{U}_{XX}^\dagger \hat{\rho} \hat{U}_{XX}\} R_{00,00} + \text{Tr}\{|01\rangle\langle 01| \hat{U}_{XX}^\dagger \hat{\rho} \hat{U}_{XX}\} R_{00,01} + \text{Tr}\{|10\rangle\langle 10| \hat{U}_{XX}^\dagger \hat{\rho} \hat{U}_{XX}\} R_{00,10} + \text{Tr}\{|11\rangle\langle 11| \hat{U}_{XX}^\dagger \hat{\rho} \hat{U}_{XX}\} R_{00,11}.$$

Similar expressions are used for all measurement bases and outcomes. For computational efficiency, the likelihood function are redefined with $\{p_a^{(ij)}\}$ and we approximate the multinomial distribution as a product of normal distributions when $N^{(ij)}$ is large:

$$\mathcal{L}^{(ij)}(\hat{\rho}) \propto \prod_{a \in \{00, 01, 10, 11\}} e^{-\left[\frac{(\bar{n}_a^{(ij)} - p_a^{(ij)})^2}{2\bar{n}_a^{(ij)}(1 - \bar{n}_a^{(ij)})} \right] N^{(ij)}},$$

where $\tilde{n}_a^{(ij)} = N_a^{(ij)} / N^{(ij)}$ is the normalized measured population. To include all the measurement results in different bases, the likelihood function is the product of all the individual likelihood function for all the bases:

$$\mathcal{L}(\hat{\rho}) = \prod_{i,j \in \{X,Y,Z\}} \mathcal{L}^{(ij)}(\hat{\rho}).$$

The total log-likelihood function across all bases is then given by:

$$\mathcal{L}(\hat{\rho}) = \sum_{i,j \in \{X,Y,Z\}} \sum_{a \in \{00,01,10,11\}} \left[\frac{- \left[\tilde{n}_a^{(ij)} - p_a^{(ij)} \right]^2 N^{(ij)}}{2 \tilde{n}_a^{(ij)} (1 - \tilde{n}_a^{(ij)})} \right].$$

To reconstruct the physical density matrix $\hat{\rho}$, we maximize this log-likelihood under the constraints that $\hat{\rho}$ is Hermitian, positive semi-definite, and has unit trace. The optimization is performed using the CVX convex optimization package in MATLAB [135, 136]. Error bar of the measurement can be derived using bootstrapping method.

The raw uncorrected two-qubit measurement outcomes used for tomography are provided in Table 3.3: The readout correction matrix used to relate measured and

Ion 1 Basis	Ion 2 Basis	N_{00}	N_{01}	N_{10}	N_{11}
X	X	1493	489	492	1385
X	Y	558	739	715	534
X	Z	660	718	648	576
Y	X	693	579	541	683
Y	Y	1472	492	486	1442
Y	Z	593	633	614	677
Z	X	564	672	643	599
Z	Y	633	679	624	692
Z	Z	356	1646	1673	349

Table 3.3: Raw two-qubit counts in the 9 Pauli bases for Ion 1 and Ion 2 under a 500 ns heralding window. N_{ab} is the number of trials with Ion 1 in $|a\rangle$ and Ion 2 in $|b\rangle$.

actual populations is:

$$\begin{pmatrix} p_{00} \\ p_{01} \\ p_{10} \\ p_{11} \end{pmatrix} = \begin{pmatrix} 0.959 & 0.052 & 0.044 & 0.002 \\ 0.018 & 0.925 & 0.001 & 0.042 \\ 0.023 & 0.001 & 0.937 & 0.051 \\ 0.000 & 0.022 & 0.018 & 0.905 \end{pmatrix} \begin{pmatrix} n_{00} \\ n_{01} \\ n_{10} \\ n_{11} \end{pmatrix},$$

where n_{ab} are the true populations and p_{ab} are the measured results. After maximum likelihood estimation with readout correction, the reconstructed two-qubit density matrix is:

$$\hat{\rho} = \begin{pmatrix} 0.046 + 0.000i & 0.026 + 0.009i & 0.013 + 0.003i & -0.002 - 0.011i \\ 0.026 - 0.009i & 0.442 + 0.000i & 0.286 + 0.071i & -0.017 + 0.008i \\ 0.013 - 0.003i & 0.286 - 0.071i & 0.433 + 0.000i & -0.010 + 0.015i \\ -0.002 + 0.011i & -0.017 - 0.008i & -0.010 - 0.015i & 0.079 + 0.000i \end{pmatrix}.$$

where the state ordering follows the basis $|00\rangle, |01\rangle, |10\rangle, |11\rangle$, with $|\text{Ion 1}, \text{Ion 2}\rangle$ labeling convention. The absolute values of the density matrix elements are visualized in Figure 3.8.

3.4.4 Coherence of the Heralded Bell States

To characterize the coherence time of the heralded Bell state, we measure the two-qubit X-basis parity as a function of delay after entanglement generation (Figure 3.9). Following the heralding event, both qubits are subjected to an XY8 dynamical decoupling sequence to mitigate decoherence and prolong spin coherence.

The observed Bell-state coherence decays with a characteristic time constant of 9.1 ± 0.4 ms, as shown in Figure 3.9. This timescale is consistent with being limited by the individual spin coherence times of Ion 1 and Ion 2 under XY8 decoupling, measured to be 21.2 ms and 16.9 ms, respectively. The observed decay therefore reflects the expected composite decoherence of the two-qubit system under local noise, confirming long-lived entanglement fidelity preservation over millisecond timescales.

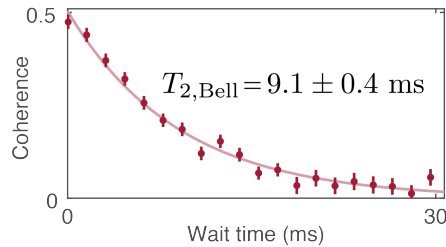


Figure 3.9: Coherence of the heralded Bell state, measured via X-basis parity oscillations following application of an XY8 dynamical decoupling sequence on both ions. The exponential decay yields a Bell-state coherence time of 9.1 ± 0.4 ms, consistent with individual ion spin coherence limits.

3.4.5 Rate and Fidelity

We evaluate the trade-off between entanglement fidelity and heralding rate by varying the detection acceptance window size and analyzing the resulting statistics, as summarized in Figure 3.10. The entanglement fidelity decreases from 0.758 ± 0.016 to 0.588 ± 0.004 as the heralding window size increases from 100 ns to $2.9 \mu\text{s}$, while the corresponding entanglement rate increases from 0.73 Hz to 9.4 Hz.

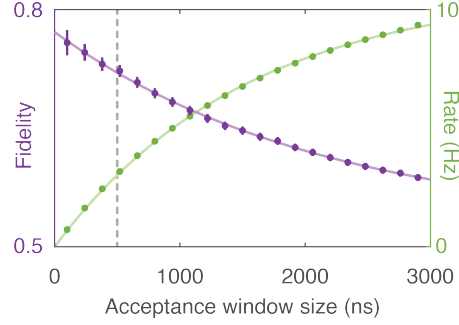


Figure 3.10: Entanglement heralding rate and fidelity as a function of photon detection window size. Solid lines are exponential fits.

In the low photon detection efficiency limit, the entanglement heralding rate for the single-photon protocol can be approximated by:

$$R = R_{\text{exp}} \times [\alpha_1 p_{\text{det}}^{(1)} (1 - e^{-\tau_A/T_{1,o}^{(1)}}) + \alpha_2 p_{\text{det}}^{(2)} (1 - e^{-\tau_A/T_{2,o}^{(1)}})], \quad (3.18)$$

where $R_{\text{exp}} = 12.3 \text{ kHz}$ is the experiment repetition rate, τ_A is the heralding window size, other parameters can be found in Table 3.2. Using measured parameters and $\tau_A = 500 \text{ ns}$, this model predicts a heralding rate of $3.48 \pm 0.03 \text{ Hz}$, closely matching the experimentally observed rate of 3.1 Hz . The photon collection efficiencies for the two devices are broken down in the table below.

Experimental contribution	Device 1 efficiency	Device 2 efficiency
Cavity–waveguide coupling	0.117 ± 0.004	0.065 ± 0.004
Chip–fibre coupling	0.27	0.12
Optical setup	0.66	0.65
SNSPD	0.85	0.85

Enhancing both the cavity-to-waveguide and chip-to-fiber coupling efficiencies is critical for improving heralding rates. This could be achieved through improved

fabrication methods and coupling strategies, such as tapered fiber coupling techniques [137]. Additionally, the repetition rate of the protocol is currently limited by the time required for qubit initialization. Since qubit initialization involves multiple optical decay cycles, reducing the optical lifetime of the qubit via enhanced Purcell factors (e.g., through higher-Q cavities) would enable faster repetition rates.

In terms of fidelity, the dominant sources of error are spontaneous emission during the rephasing window, optical pulse infidelities, and dark counts, each contributing approximately 5% to the total error budget, as supported by simulations reported in Ref. [80]. To improve performance, future implementations may benefit from using lower-concentration samples, which reduce background photon events and enable stronger optical pulses with reduced emission from neighboring emitters. Furthermore, if higher quality factor devices can be made, rephasing may be performed on alternative excited-state transitions not Purcell-enhanced, provided dominant optical dephasing is common-mode between levels. In combination, these improvements would allow for both higher heralded entanglement rates and fidelities in next-generation devices.

3.4.6 Two-photon Protocol

In addition to the single-photon protocol, we also implement a two-photon entanglement protocol in our system. Unlike the single-photon scheme, the fidelity of the two-photon protocol is not limited by the small excitation amplitude (α), and it naturally supports higher entanglement fidelities. Additional advantages include the absence of a requirement for optical path phase stabilization and built-in verification that both ^{171}Yb ions remain in the qubit manifold, since only valid photon detections from both ions contribute to the heralded events.

Figure 3.11(a) shows the experimental sequence for two-photon entanglement heralding. The protocol uses two consecutive heralding windows, separated by simultaneous application of spin and excited-state π pulses. This sequence is designed to reduce infidelity arising from residual optical excited-state population and to enhance readout contrast in the computational basis. When a single photon is detected at time t_0 during the first heralding window, and another photon is detected at time t_1 in the second window, the two-qubit state is projected into:

$$|\psi\rangle = |10\rangle + e^{-i\Delta\omega_{12}(t_1-t_0)} |01\rangle, \quad (3.19)$$

where $\Delta\omega_{12}$ is the optical frequency difference between the ions.

After heralding, the protocol accounts for the timing order of photon detections: if $t_1 > t_0$, an odd number of dynamical decoupling sequences, comprising simultaneous spin and excited-state π pulses, are applied before the dynamic rephasing step, effectively canceling spectral diffusion. If $t_0 > t_1$, an even number of sequences are applied. Additional decoupling pulses are added after dynamic rephasing so that the total number of pulses is even in both cases, ensuring consistency for all heralded events.

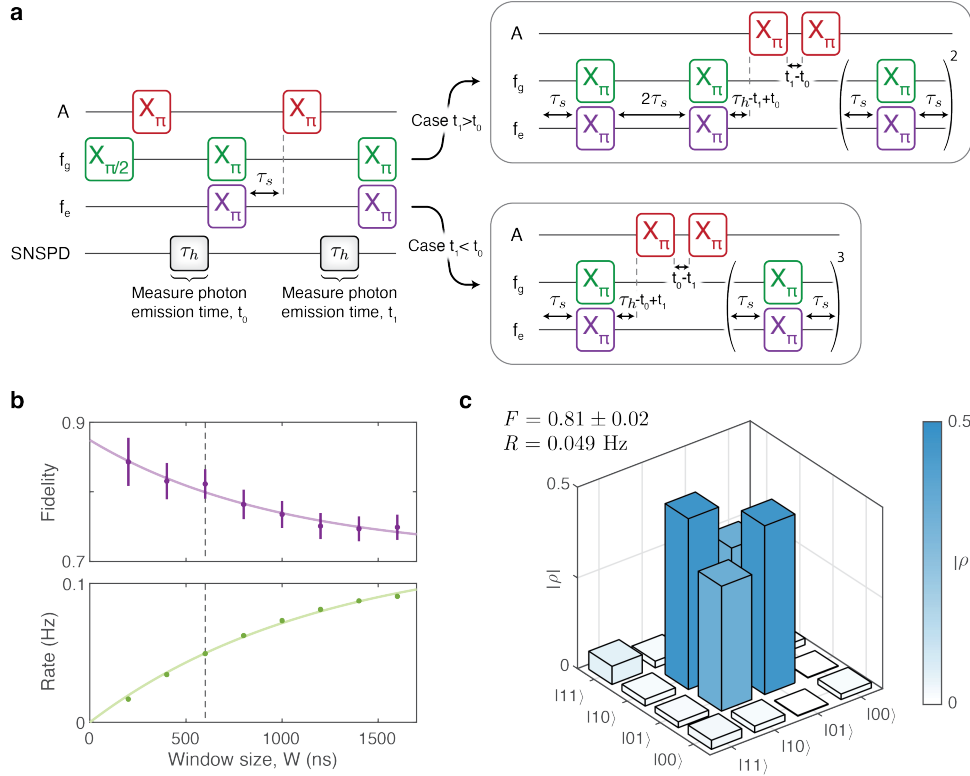


Figure 3.11: Two-photon entanglement heralding. (a) Experimental pulse sequence for the two-photon protocol. (b) Entanglement fidelity and heralding rate as functions of the photon detection window size; solid lines are exponential fits. (c) Absolute value of the reconstructed two-qubit density matrix measured with a 600 ns heralding window. $F = 0.81 \pm 0.02$, rate = 0.049 Hz

Figure 3.11(b) presents the measured tradeoff between entanglement fidelity and heralding rate for varying detection window sizes. The absolute value of the reconstructed density matrix, obtained with a 600 ns two-sided window, is shown in Figure 3.11(c). The resulting Bell state fidelity is 0.81 ± 0.02 at a heralding rate of 0.049 Hz.

The experimental data underlying the density matrix in Figure 3.11(c) are presented below. In the table, N_{ab} refers to the number of counts where Ion 1 is measured in state $|a\rangle$ and Ion 2 in state $|b\rangle$; each pair of basis settings corresponds to tomography in one of the nine Pauli bases.

Ion 1 Basis	Ion 2 Basis	N_{00}	N_{01}	N_{10}	N_{11}
X	X	134	29	35	129
X	Y	69	67	72	66
X	Z	79	77	82	76
Y	X	65	89	70	70
Y	Y	119	36	35	117
Y	Z	67	69	77	64
Z	X	69	63	82	85
Z	Y	78	70	75	67
Z	Z	15	122	102	19

The readout transformation matrix is measured to be:

$$\begin{pmatrix} p_{00} \\ p_{01} \\ p_{10} \\ p_{11} \end{pmatrix} = \begin{pmatrix} 0.936 & 0.052 & 0.049 & 0.003 \\ 0.027 & 0.911 & 0.001 & 0.048 \\ 0.035 & 0.002 & 0.923 & 0.051 \\ 0.001 & 0.034 & 0.027 & 0.898 \end{pmatrix} \begin{pmatrix} n_{00} \\ n_{01} \\ n_{10} \\ n_{11} \end{pmatrix},$$

where p_{ab} and n_{ab} denote measured and actual populations, respectively, with labeling according to $|\text{Ion 1, Ion 2}\rangle$. Applying maximum likelihood quantum state tomography, the reconstructed density matrix is:

$$\hat{\rho} = \begin{pmatrix} 0.015 + 0.000i & 0.011 + 0.014i & -0.001 + 0.001i & 0.019 - 0.015i \\ 0.011 - 0.014i & 0.470 + 0.000i & 0.343 + 0.028i & 0.018 - 0.001i \\ -0.001 - 0.001i & 0.343 - 0.028i & 0.465 + 0.000i & 0.004 - 0.016i \\ 0.019 + 0.015i & 0.018 + 0.001i & 0.004 + 0.016i & 0.050 + 0.000i \end{pmatrix}.$$

This matrix is ordered in the computational basis, with the convention $|\text{Ion 1, Ion 2}\rangle$.

3.5 Tripartite Entanglement Heralding

To extend the capabilities of our quantum node beyond bipartite entanglement, we leverage the presence of multiple spectrally distinguishable ions within a single device to demonstrate multipartite remote entanglement distribution. In particular, we implement a protocol involving two ions in one node and a third ion in a separate

node, enabling the heralding of tripartite entanglement across the full system. This marks a key advancement toward scalable multi-node quantum networks.

In the following subsections, we first introduce the methods used for independent control of spectrally distinct ions within a single device. We then describe the experiment in detail, which enable successful tripartite entanglement generation and verification.

3.5.1 Single Device Individual Ion Control

In the tripartite entanglement heralding experiment, Ion 1 and Ion 3 in Device 1 and Ion 2 in Device 2 are utilized. Because microwave control is implemented via a coplanar waveguide, the same microwave pulse is simultaneously applied to all ions within the same device. Since the frequency separation between the qubit transitions of ions within a device is smaller than the overall microwave pulse bandwidth, direct frequency-selective control is not possible. As a result, distinguishing qubit control between the two ions on the same chip requires additional techniques.

To address this, we leverage the fact that the ions within a device have distinct optical transition frequencies. By applying a far-detuned optical pulse to both ions, we induce a differential ac Stark shift that acts as a local Z rotation. Because the detuning from resonance is different for each ion, the ac Stark shift accumulated during the same pulse is different, enabling independent, single-qubit rotations within the otherwise globally controlled microwave system [138].

For sufficiently far detuned optical pulses (i.e., where the detuning is much greater than the spontaneous emission rate), the induced phase difference between two ions is given by:

$$\phi = T\Omega^2\left(\frac{1}{\Delta_2} - \frac{1}{\Delta_1}\right), \quad (3.20)$$

where T is the optical pulse duration, Ω is the Rabi frequency, Δ_1 and Δ_2 are the detunings from the optical resonance for Ion 1 and Ion 3, respectively.

The pulse sequence used for this control is shown in Figure 3.12(a). A qubit dynamical decoupling sequence mitigates spin dephasing, and ac Stark-inducing optical pulses are applied during the odd-numbered intervals so that the induced phase accumulates constructively over time. This differential phase shift can be observed experimentally by preparing the two ions in superposition states and measured the accumulated relative phase.

In Figure 3.12(b), we plot the experimentally measured differential phase shift

between Ion 1 and Ion 3 as a function of optical pulse amplitude. The dynamical decoupling sequence used consists of 32 π pulses with $5.8 \mu\text{s}$ spacing. The optical pulse is detuned by $-2\pi \times 50 \text{ MHz}$ for Ion 1 and $+2\pi \times 425 \text{ MHz}$ for Ion 3. The observed linear relationship validates the differential phase accumulation and enables an ion-specific Z rotation gate. When combined with global single-qubit gates, full single-qubit control over individual ions is achieved. This scheme can be extended to larger numbers of ions: control over N ions requires N–1 different detunings to enable universal local Z control.

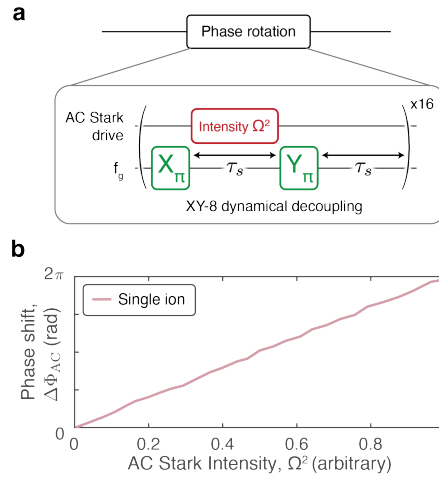


Figure 3.12: Individual ion control using AC Stark Shift. (a) Pulse sequence for individual ion control within a single device using differential ac Stark shifts. Optical pulses are applied during alternating periods of a qubit dynamical decoupling sequence to accumulate phase. (b) Measured differential phase shift between two ions as a function of optical pulse amplitude. The linear dependence demonstrates controllable individual Z rotations based on optical amplitudes.

In addition to ac Stark shift control, we exploit the intrinsic frequency difference between the two qubit transitions to implement ion-specific Z rotations by introducing controlled wait intervals. Specifically, Ion 1 and Ion 3 have a qubit frequency separation of $2\pi \times 1.13 \text{ MHz}$, which enables differential Z rotations via a controlled wait time of approximately 440 ns. For example, measurement in the ZX Pauli basis is implemented by the sequence:

$$Y_{\pi/4} Z_{+\pi/2}^{(1)} Z_{-\pi/2}^{(3)} Z_{\pi/4}, \quad (3.21)$$

where superscripts denote the specific ion addressed. Similar logic applies for enabling other Pauli basis measurements. The application of such wait-time-based

Z gates is practical only when the qubit frequency difference is sufficiently large; otherwise, extended wait times may degrade fidelity due to dephasing.

3.5.2 Tripartite entanglement Heralding Sequence

To extend the bipartite single-photon entanglement heralding protocol to a three-qubit system, we implement a generalized scheme involving three spectrally resolvable ions located across two devices (Figure 3.13). Prior to the entanglement sequence, all three ions are initialized into the ground qubit state $|0\rangle_g$. Spin excitations are then applied using small-angle α pulses, analogous to the bipartite single-photon protocol. The resulting joint quantum state is:

$$|\psi\rangle = (\sqrt{1-\alpha_1}|0\rangle_g + \sqrt{\alpha_1}|1\rangle_g) \otimes (\sqrt{1-\alpha_2}|0\rangle_g + \sqrt{\alpha_2}|1\rangle_g) \otimes (\sqrt{1-\alpha_3}|0\rangle_g + \sqrt{\alpha_3}|1\rangle_g). \quad (3.22)$$

Optical π pulses are then applied to drive the $|1\rangle_g \rightarrow |e\rangle$ transition, followed by a single-photon detection event at time t_0 . Assuming small α_i and only one photon detected, the leading-order spin state is projected into a W-like entangled state:

$$|\psi\rangle = \frac{1}{3}(|100\rangle + e^{-i\Delta\omega_{12}t_0}|010\rangle + e^{-i\Delta\omega_{13}t_0}|001\rangle), \quad (3.23)$$

where $\Delta\omega_{1i}$ is the optical frequency difference between Ion 1 and Ion i . $|000\rangle_g$ is excluded, as it emits no photon, and higher-order multi-photon states are neglected due to the small contributions. The spin rotation amplitudes α_i are carefully chosen to equalize the photon emission probabilities from each ion, ensuring balanced W-state components.

After heralding, dynamic optical rephasing is performed using the measured photon emission time t_0 to cancel phase errors caused by spectral diffusion. Following this, a feedforward correction is applied to compensate for the deterministic relative phases arising from the optical frequency differences between the ions. This sequence coherently transforms the initial spin-photon entanglement into a tripartite entangled W-state across spatially separated emitters.

3.5.3 Parity Oscillations between Pairs of Ions

To characterize the time-dependent coherence of the heralded three-ion W state, we measure all three qubits in the X basis as a function of heralded time. Specifically, we define the pairwise coherence between ions i and j , denoted as C_{ij} , by the expectation value

$$\text{Tr}(\hat{\rho} |01\rangle_{ij} \langle 10|_{ij} \otimes |0\rangle_k \langle 0|_k), \quad (3.24)$$

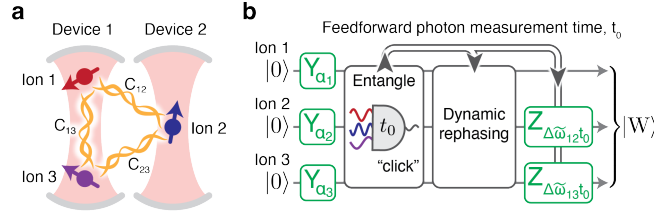


Figure 3.13: Tripartite entanglement heralding sequence. (a) Experimental schematic of three-ion entanglement: two ions are housed within the same cavity and the third in a separate node. (b) Pulse sequence used to generate and herald W state via a single-photon detection, dynamic rephasing, and phase feedforward.

where the subscript indices refer to individual ions ($i, j, k \in 1, 2, 3$) and k denotes the third ion that is traced out. This observable isolates the quantum coherence between ions i and j when the third ion is in the $|0\rangle_g$ state, as expected for a W-type state.

In the three-ion experiments, $\alpha_1 = 0.079$, $\alpha_2 = 0.078$, and $\alpha_3 = 0.079$. The coherence oscillates with a frequency set by the optical frequency separation between each ion pair due to the accumulated phase differences at the time of heralding. By performing Fourier transform of the measured X-basis parity signal for each ion pair, after tracing over the third ion, we extract frequency-resolved coherence signatures.

Figure 3.14(c) shows the Fourier-transformed parity signals for all three ion pairs. Distinct peaks in each spectrum correspond to the ion-ion coherence oscillation frequencies and confirm successful creation of a coherent three-ion state. The extracted pairwise frequency offsets are $2\pi \times 508$ MHz, $2\pi \times 475$ MHz, and $2\pi \times 34$ MHz, which align with the measured spectral separations between the three optical transitions.

3.5.4 Quantum State Tomography of W State

To deterministically herald a three-ion W state across two quantum network nodes, we extend the single-photon heralding protocol with dynamic optical rephasing and two sequential phase corrections. Ions 1 and 3 reside in Device 1, while Ion 2 is located in Device 2. All three ions are initialized into the ground state $|0\rangle_g$ and driven into weak spin superpositions prior to optical excitation. Upon the detection of a single photon, signaling a successful heralding event, the system is projected into a superposition of single-excitation states across the three ions. However, due to frequency differences and spectral diffusion, the resulting entangled state

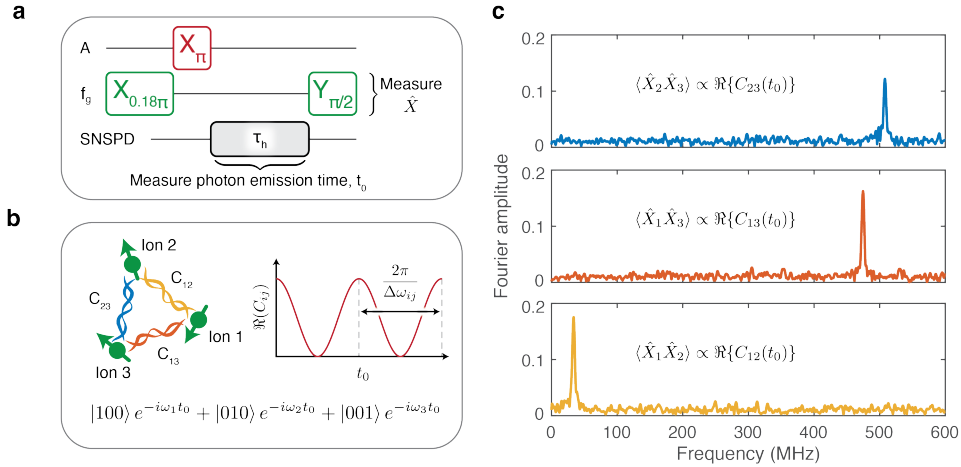


Figure 3.14: Parity measurements for the three-ion W state after heralding. (a) Experimental pulse sequence used to measure time-dependent X-basis parity following heralded three-ion entanglement. (b) Schematic illustration of coherence between different ion pairs within the W state. Each coherence term is extracted by tracing over the third ion. (c) Fourier-transformed spectra of the measured parity signals for all three ion pairs. The peaks indicate the coherent phase evolution frequencies corresponding to optical frequency differences between each ion pair.

accumulates deterministic and fluctuating phases that must be corrected before the final spin state is read out.

Immediately after heralding, dynamic optical rephasing is applied to all three ions. This process cancels the random phase accumulation caused by spectral diffusion by re-exciting the ions at a time equal to the detected photon emission time, ensuring phase refocusing of the optical transition. Once optical coherence is restored, two feedforward phase corrections are performed to compensate for known deterministic phase differences between specific ion pairs due to static optical frequency offsets.

The first phase correction is applied between Ion 1 and Ion 3 using the ac Stark shift control technique described in Section 3.5.1. The required optical pulse amplitude is calibrated using parity oscillations between Ion 1 and Ion 3 at several reference power levels. Given the linear scaling between pulse amplitude and induced Stark shift, the appropriate compensation phase can be calculated and applied in real time.

The second phase correction addresses the relative phase between Ion 1 and Ion 2. This is implemented by adjusting the microwave drive phase applied to Ion 2. Since the ac Stark pulse applied to Ion 1 in the previous step also changes its spin phase, this additional rotation must be taken into account when calculating the microwave

phase shift needed to cancel detuning-induced precession between Ion 1 and Ion 2.

To prevent phase drifts during data acquisition, we perform optical beatnote calibration between the two photon collection paths every 15 minutes. Using this calibration, we update the AOM phases to maintain long-term phase stability of the optical paths.

After completing the phase corrections, we perform full three-qubit quantum state tomography by measuring in 27 combinations of Pauli bases (X, Y, Z). With a heralding window size of 300 ns, we observe a heralding rate of 2 Hz and reconstruct the density matrix shown in Figure 3.15. The fidelity of the reconstructed state with respect to the ideal W state is 0.592 ± 0.007 .

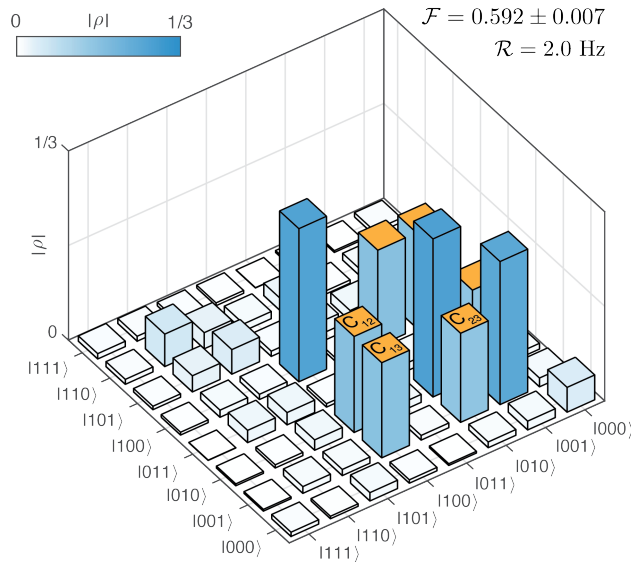


Figure 3.15: Quantum state tomography of the heralded three-ion W state. The density matrix is reconstructed from 27 Pauli basis measurements using a 300 ns photon detection window. The resulting state has a fidelity of 0.592 ± 0.007 and an entanglement heralding rate of 2 Hz.

The raw measurement data used to generate the density matrix are presented in the table below, where N_{abc} denotes the number of measurement outcomes with Ion 1 in $|a\rangle$, Ion 2 in $|b\rangle$, and Ion 3 in $|c\rangle$.

Ion 1 Basis	Ion 2 Basis	Ion 3 Basis	N_{000}	N_{001}	N_{010}	N_{011}	N_{100}	N_{101}	N_{110}	N_{111}
X	X	X	513	164	166	196	208	179	233	475
X	X	Y	316	464	188	185	231	185	398	329
X	X	Z	516	235	199	173	215	179	483	209
X	Y	X	393	167	405	243	235	379	182	327
X	Y	Y	335	213	185	443	394	187	203	333
X	Y	Z	349	195	358	211	358	196	337	175
X	Z	X	517	192	252	163	209	492	196	231
X	Z	Y	338	384	230	237	392	295	221	229
X	Z	Z	337	310	349	65	362	312	372	67
Y	X	X	345	168	186	359	414	200	192	301
Y	X	Y	374	185	363	153	147	341	222	368
Y	X	Z	378	204	343	193	307	215	330	225
Y	Y	X	205	189	374	412	375	333	205	178
Y	Y	Y	514	187	202	213	185	186	190	529
Y	Y	Z	506	209	210	169	178	188	511	251
Y	Z	X	344	357	220	226	394	308	217	209
Y	Z	Y	485	183	216	207	200	464	187	225
Y	Z	Z	344	280	323	71	409	323	348	64
Z	X	X	437	181	201	443	241	158	185	219
Z	X	Y	362	329	365	318	215	214	211	201
Z	X	Z	378	349	375	316	343	60	319	73
Z	Y	X	348	294	350	336	233	209	218	202
Z	Y	Y	484	225	189	476	240	210	173	232
Z	Y	Z	409	347	369	306	314	92	355	77
Z	Z	X	390	311	354	352	343	356	99	69
Z	Z	Y	358	310	324	335	324	319	87	75
Z	Z	Z	221	501	533	111	594	113	136	26

Readout errors are corrected using a calibrated 3-qubit readout matrix, which maps actual populations n_{abc} to observed populations p_{abc} . The readout transformation matrix is measured to be:

$$\begin{pmatrix} p_{000} \\ p_{001} \\ p_{010} \\ p_{011} \\ p_{100} \\ p_{101} \\ p_{110} \\ p_{111} \end{pmatrix} = \begin{pmatrix} 0.910 & 0.059 & 0.052 & 0.003 & 0.058 & 0.004 & 0.003 & 0.000 \\ 0.037 & 0.889 & 0.002 & 0.051 & 0.002 & 0.057 & 0.000 & 0.003 \\ 0.016 & 0.001 & 0.874 & 0.057 & 0.001 & 0.000 & 0.056 & 0.004 \\ 0.001 & 0.015 & 0.036 & 0.854 & 0.000 & 0.001 & 0.002 & 0.054 \\ 0.034 & 0.002 & 0.002 & 0.000 & 0.886 & 0.058 & 0.050 & 0.003 \\ 0.001 & 0.033 & 0.000 & 0.002 & 0.036 & 0.865 & 0.002 & 0.049 \\ 0.001 & 0.000 & 0.032 & 0.002 & 0.015 & 0.001 & 0.851 & 0.055 \\ 0.000 & 0.001 & 0.001 & 0.032 & 0.001 & 0.015 & 0.035 & 0.831 \end{pmatrix} \begin{pmatrix} n_{000} \\ n_{001} \\ n_{010} \\ n_{011} \\ n_{100} \\ n_{101} \\ n_{110} \\ n_{111} \end{pmatrix},$$

where $\{p_{abc}\}$ and $\{n_{abc}\}$ are the measured and actual populations, respectively, associated with Ion 1 in the $|a\rangle$ state, Ion 2 in the $|b\rangle$ state and Ion 3 in the $|c\rangle$ state.

After applying maximum likelihood estimation with readout correction constraints (positive semidefiniteness, Hermiticity, and unit trace), we obtain the reconstructed density matrix expressed as $\hat{\rho} = \hat{\rho}_{\text{real}} + i\hat{\rho}_{\text{imag}}$, with the corresponding real and imaginary components given explicitly. The matrix ordering follows the state vector basis:

$$\hat{\rho}_{\text{real}} = \begin{pmatrix} 0.044 & -0.013 & 0.009 & -0.003 & 0.005 & -0.009 & -0.003 & -0.005 \\ -0.013 & 0.252 & 0.156 & -0.000 & 0.164 & -0.001 & -0.011 & 0.002 \\ 0.009 & 0.156 & 0.278 & 0.007 & 0.168 & -0.002 & 0.006 & 0.003 \\ -0.003 & -0.000 & 0.007 & 0.046 & -0.002 & 0.017 & 0.021 & 0.000 \\ 0.005 & 0.164 & 0.168 & -0.002 & 0.270 & 0.006 & -0.000 & 0.004 \\ -0.009 & -0.001 & -0.002 & 0.017 & 0.006 & 0.043 & 0.028 & 0.005 \\ -0.003 & -0.011 & 0.006 & 0.021 & -0.000 & 0.028 & 0.057 & -0.009 \\ -0.005 & 0.002 & 0.003 & 0.000 & 0.004 & 0.005 & -0.009 & 0.009 \end{pmatrix},$$

$$\hat{\rho}_{\text{imag}} = \begin{pmatrix} 0.000 & -0.008 & 0.004 & 0.000 & -0.003 & -0.011 & 0.003 & 0.006 \\ 0.008 & 0.000 & 0.014 & 0.008 & 0.037 & 0.011 & -0.003 & -0.003 \\ -0.004 & -0.014 & 0.000 & 0.012 & 0.021 & 0.018 & -0.000 & -0.002 \\ -0.000 & -0.008 & -0.012 & 0.000 & -0.006 & 0.013 & 0.004 & -0.000 \\ 0.003 & -0.037 & -0.021 & 0.006 & 0.000 & 0.007 & 0.013 & -0.003 \\ 0.011 & -0.011 & -0.018 & -0.013 & -0.007 & 0.000 & 0.006 & -0.003 \\ -0.003 & 0.003 & 0.000 & -0.004 & -0.013 & -0.006 & 0.000 & -0.001 \\ -0.006 & 0.003 & 0.002 & 0.000 & 0.003 & 0.003 & 0.001 & 0.000 \end{pmatrix}.$$

The matrix ordering follows the state vector basis:

$$\left(|000\rangle \ |001\rangle \ |010\rangle \ |011\rangle \ |100\rangle \ |101\rangle \ |110\rangle \ |111\rangle \right)^T, \quad (3.25)$$

where we adopt a convention of $|\text{Ion 1, Ion 2, Ion 3}\rangle$ for labelling. The absolute value of the reconstructed matrix is visualized in Figure 3.15.

To explore the rate–fidelity tradeoff, we repeat the tomography procedure for varying photon detection window widths. The resulting fidelity and heralding rate as functions of the acceptance window size, ranging from 100 ns to 2.9 μ s, are

plotted in Figure 3.16. The entanglement fidelity decreases from 0.586 ± 0.012 to 0.460 ± 0.003 as the heralding window size increases from 100 ns to $2.9 \mu\text{s}$, while the corresponding entanglement rate increases from 0.66 Hz to 10.5 Hz.

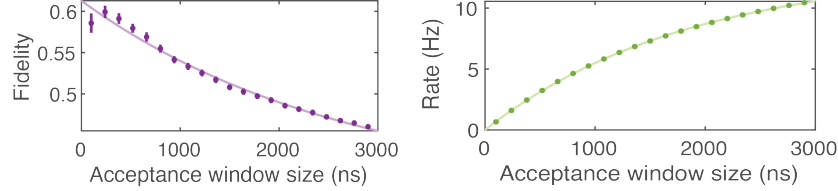


Figure 3.16: Three-ion W state heralding rate and fidelity as a function of photon detection window size. Solid lines are exponential fits.

3.6 Multiplexed Remote Entanglement

With multiple spectrally distinct ions within a single quantum node, we extend the utility of our system beyond tripartite entanglement to implement multiplexed entanglement heralding. This approach utilizes local multi-qubit resources to overcome entanglement heralding overheads and increase the overall entanglement rate [43, 44, 139, 140]. The key advantage is the ability to overcome entanglement bottleneck caused by photon transmission time between distant nodes. In our case, the entanglement rate is limited by the initialization time. By parallelizing the initialization process, we can increase the entanglement rate by using multiple pairs of ions.

To realize independent and efficient control of each ion in both devices, we employ three double-pass acousto-optic modulator (AOM) configurations for each device to enable flexible addressing of both F transitions for ions with large frequency separations (500–600 MHz). For the A transition, we exploit the zeroth- and first-order diffracted beams from AOM5 and AOM9. Within a single device, zeroth-order light from AOM5 and AOM9 is used to selectively pump Ion 1 and Ion 2, respectively, while first-order diffracted beams address Ion 3 and Ion 4. To ensure precise ion addressing and avoid cross-talk, the frequencies of AOM3 and AOM4 are set such that zeroth-order beams are off-resonant with all unaddressed ions when first-order beams are in use. This strategy affords independent optical control over all four ions (Ion 1–4) across both devices, as illustrated in Figure 3.2.

3.6.1 Experimental Sequence Details

The experimental sequence for performing multiplexed entanglement heralding follows the same protocolic structure as the bipartite and tripartite schemes described

previously, with a few key modifications to support multiple ion pairs operated in parallel. The extended sequence is shown in Figure 3.17, where the timing control and sequence flow for two pairs of ions, Ion 3/4 and Ion 1/2, are outlined.

During the qubit initialization stage, the optical π pulses for Ion 3 and Ion 4 are applied immediately following those for Ion 1 and Ion 2, but prior to the excited-state microwave π pulses. Because the optical lifetime of ^{171}Yb in YVO_4 is relatively long (1–2 μs) compared to the optical π -pulse duration (~ 140 ns), the state preparation remains highly efficient and does not suffer from state decay between pulse sequences.

In the entanglement attempts, Ion 3/4 and Ion 1/2 entanglement heraldings are executed sequentially within the same experimental repetition. A global qubit α -pulse is applied to all four ions, after which optical π pulses are first applied to Ion 3 and Ion 4. This is followed by a heralding window. Regardless of outcome, two dynamical decoupling (DD) pulses are applied to all four ions to preserve coherence and balance spin evolution. After the DD interval, optical π pulses are applied to Ion 1 and Ion 2, followed by a second heralding window. This is again followed by two global dynamical decoupling pulses.

If a photon detection is registered during the Ion 1/2 heralding window, dynamic optical rephasing and feedforward phase compensation (based on the detected photon's emission time) are applied to Ion 1 and Ion 2. If instead the detection occurs during the Ion 3/4 heralding window, the corresponding feedforward procedures are applied to that pair. If no photons are detected in either window, the sequence concludes, the ions are re-initialized, and the entanglement attempt restarts with the next trial.

The entanglement attempt duration for a single ion pair (either Ion 1/2 or Ion 3/4) is 81 μs . When operating both pairs sequentially in the same trial, an additional 12 μs is required to accommodate the second entanglement attempt, bringing the total multiplexed cycle time to 93 μs . Assuming both ion pairs have the same success probability and act independently, since this single 93 μs cycle contains two independent entanglement attempts, the protocol yields a theoretical rate enhancement factor of $2 \times (81 \mu\text{s} / 93 \mu\text{s}) \approx 1.74$ compared to operating a single pair with an 81 μs cycle time. This demonstrates that multiplexing two ion pairs nearly doubles the entanglement generation rate.

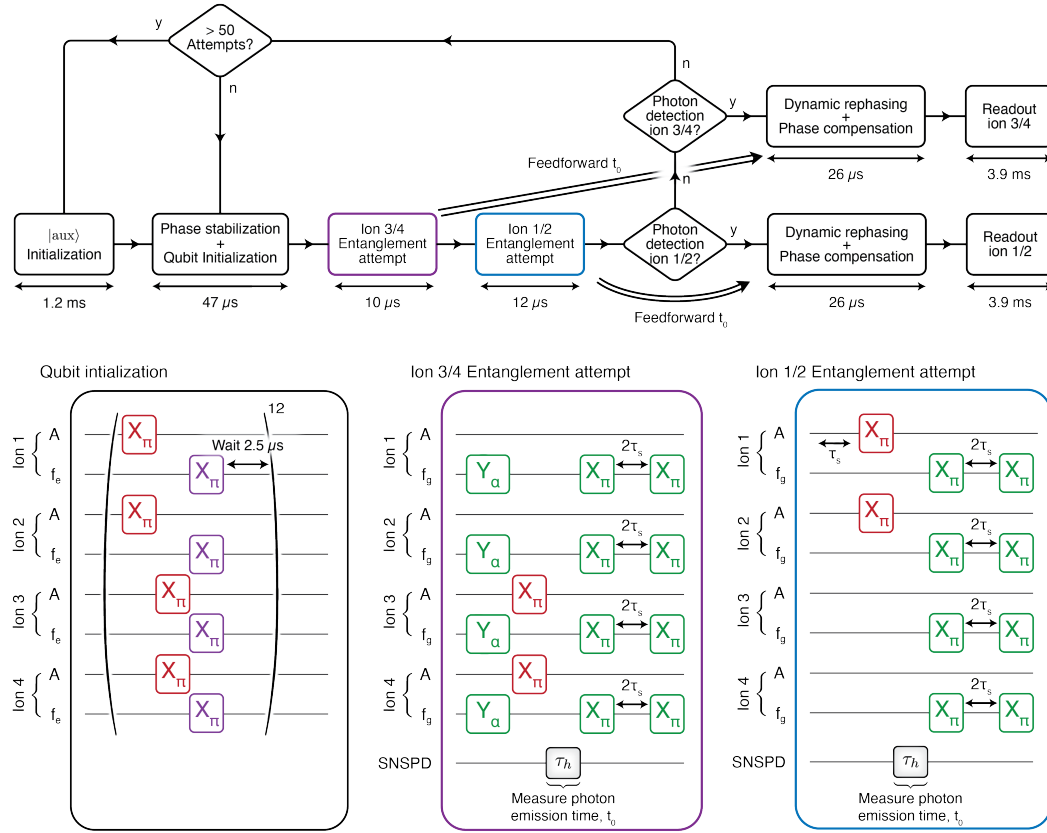


Figure 3.17: Details of the multiplexed entanglement heralding sequence. Top: flow chart of the experimental protocol. Sequential entanglement attempts are launched first on Ion 3/4, followed by Ion 1/2. Depending on which pair successfully emitted a heralded photon, the appropriate rephasing and phase correction sequences are conditionally applied. Bottom: subpanels highlight protocol elements distinct from standard bipartite heralding, including parallel ion initialization and sequential entanglement attempts.

3.6.2 Multiplexed Rate and Fidelity

To evaluate the performance of multiplexed remote entanglement heralding, we select Ion 1 and Ion 2 (separated by 36.5 MHz) as Pair 1, and Ion 3 and Ion 4 (separated by 104 MHz) as Pair 2, as labeled in Figure 3.4. Each ion pair is first characterized independently using the bipartite entanglement heralding protocol described in Section 3.4, with results shown in Figure 3.18(a).

The experiments are conducted using resonant qubit microwave pulses, optimized for independent pair measurements. At a heralding window size of 500 ns, Pair 1 yields a fidelity of 0.745 ± 0.005 and an entanglement rate of 3.8 Hz. Pair 2, in comparison, exhibits a fidelity of 0.656 ± 0.006 and a rate of 3.7 Hz. The lower

fidelity of Pair 2 arises from background ions contributing dark counts during the detection window. The slightly improved performance of Pair 1 compared to earlier results in Section 3.4 is due to the use of a two-SNSPD setup here, as opposed to the delay-line interferometric detection setup described in Section 3.2. The delay-line system requires frequent fiber re-splicing when ion frequency drifts, which reduced fidelity.

We then perform multiplexed entanglement heralding with both ion pairs operating concurrently. Using the same 500 ns heralding window, we measure an average fidelity of 0.682 ± 0.004 . The fidelity for Pair 1 under multiplexed operation is 0.700 ± 0.005 , while Pair 2 yields 0.668 ± 0.006 . The total entanglement event rate increases to 7.1 Hz, with 4.1 Hz contributed by Pair 1 and 3.0 Hz from Pair 2. This corresponds to a $1.9\times$ rate enhancement compared to operating a single ion pair alone—demonstrating the effectiveness of multiplexing for boosting entanglement throughput. Pauli basis measurements (X, Y, and Z) on the entangled output states are shown in Figure 3.18(b).

When comparing the expected entanglement rate and experiment entanglement rate, we note that the total multiplexed entanglement rate is slightly higher than predicted. With the pulse sequence, we estimate that the entanglement rate for Pair 1 should be $3.8 \text{ Hz} \times 0.87 = 3.3 \text{ Hz}$, and for Pair 2, $3.7 \text{ Hz} \times 0.87 = 3.2 \text{ Hz}$, where the 13% rate reduction accounts for additional entanglement attempt in the multiplexed configuration. The observed rate for Pair 2 is consistent with this prediction and can be explained by modest alignment drift in the optical setup over time. In contrast, the observed Pair 1 entanglement rate exceeds expectations.

We attribute this difference to implementation details in the pulse sequence. Because all ions within the same device share a common microwave control, we apply global qubit pulses whose frequencies lie between those of the individual ion's qubit transitions. In this case, the microwave pulse is slightly detuned from resonance for both ions in the pair. To evaluate its effect, we simulate the population dynamics of all four ions under the multiplexed entanglement attempt sequence, incorporating the detuned microwave pulses and local Overhauser field variations. As shown in Figure 3.19, the detuned pulses result in increased population for Ion 1 and Ion 2 prior to the start of the heralding window. This increased excitation probability explains the elevated heralding rate and the slight reduction in fidelity observed in the experimental results for Pair 1 compared to its standalone measurements.

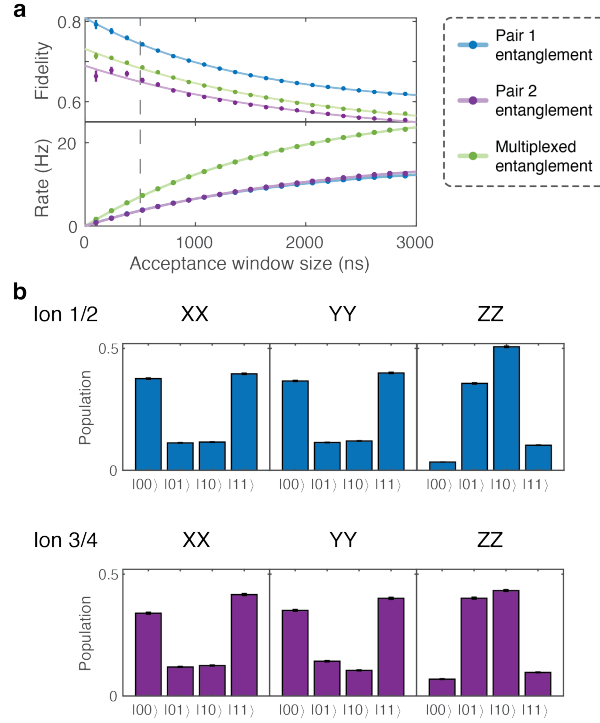


Figure 3.18: Multiplexed remote entanglement heralding results. (a) Fidelity and heralding rate versus photon detection window size. Blue and purple curves correspond to standalone entanglement characterization of Pair 1 and Pair 2, respectively. Green data correspond to multiplexed entanglement results. At fixed detection window size, the entanglement rate is nearly doubled while fidelity remains close to the average of the two pairs. (b) Pauli X, Y, and Z measurements of the multiplexed entangled states, extracted with a 500 ns detection window.

3.7 Outlook

In this chapter, we have experimentally demonstrated the use of multiple spectrally distinct ^{171}Yb ions within a single nanophotonic cavity for remote entanglement heralding. By leveraging these local multi-qubit resources, we implemented advanced entanglement distribution protocols, including tripartite entanglement and multiplexed bipartite entanglement heralding. These demonstrations highlight the scalability and versatility of our quantum node architecture.

To enable more complex quantum networking protocols and further enhance the performance of entanglement-based operations, improvements in both the coherence and fidelity of nuclear spin storage and the fidelity and rate of remote entanglement generation are required. Strategies for increasing fidelity and entanglement rates, including suppression of spontaneous emission during rephasing, reduction of opti-

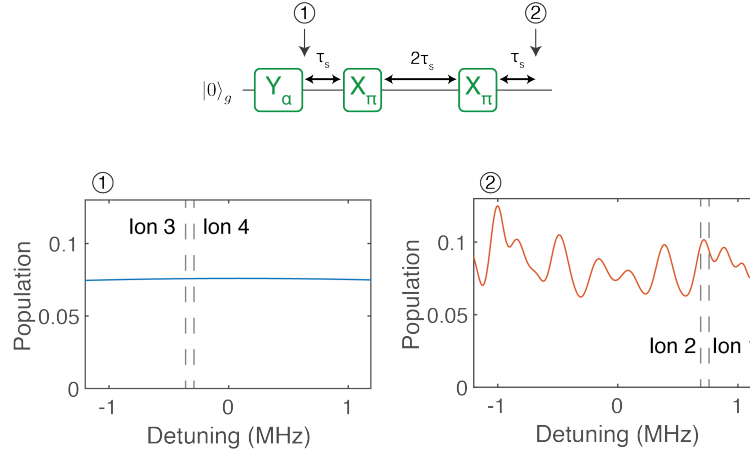


Figure 3.19: Simulated population for all four ions under the multiplexed entanglement pulse sequence. Left: effect of the pulse sequence on Ion 3 and Ion 4 (Pair 2). Right: response of Ion 1 and Ion 2 (Pair 1), showing increased population in the excited manifold due to detuned qubit pulses, which contributes to the enhanced heralding rate.

cal pulse errors, and mitigation of dark counts, have been discussed in Section 3.4. These improvements are particularly critical for applications such as entanglement swapping and quantum repeaters.

Looking beyond current capabilities, increasing the number of spectrally resolvable ions within a single device offers a clear path to expanding the available qubit resources at each node. Greater spectral occupancy would not only support higher parallelism in entanglement distribution but also enable local quantum processing among resident ions either through dipolar interaction [104] or remote entanglement within the same node. Recent work has shown that co-doping additional rare-earth species into the host crystal can significantly increase the spectral diversity and density of addressable emitters [87]. Exploring this direction offers a promising route toward scalable, multiplexed quantum networks.

Chapter 4

CREATING DECOHERENCE-PROTECTED GREENBERGER–HORNE–ZEILINGER STATE USING NUCLEAR SPINS

Controllable and interacting multi-spin systems hold great promise for advancing quantum information processing, quantum simulation, and quantum sensing [119–121, 141]. Solid-state nuclear spins are particularly attractive for these applications due to their exceptionally long coherence times, making them ideal for quantum information storage [142, 143]. These highly coherent nuclear spins have enabled pioneering advances in quantum information science and many-body physics [144, 145]. However, the weak interactions between nuclear spins — arising from their small magnetic moments — pose challenges in efficiently generating highly entangled states, an essential resource for quantum error correction and quantum networking [9, 29].

Electronic spins embedded in nuclear spin baths (so-called central spins) offer a solution by enabling stronger interactions due to their larger magnetic moments and proximity [146, 147]. This enhanced interaction strength facilitates efficient control of nuclear spins via central spins, effectively overcoming the weak coupling between nuclear spins. Such central-spin platforms have led to notable achievements across various systems, including demonstrations of many-body-localized discrete time crystals, fault-tolerant operations, and nuclear magnonic quantum registers [119, 121, 148].

Despite these achievements, a fundamental trade-off persists between the interaction strength and nuclear spin decoherence induced by the central spin. In particular, optical readout and initialization of central spins introduce decoherence to nuclear spins that cannot be mitigated by dynamical decoupling [54, 149, 150]. Furthermore, decoherence from the central spins and central spin coherence times limit the number of accessible nuclear spins. Nearby nuclear spins with stronger interactions can also induce decoherence on the central spins [151]. To fully harness the potential of nuclear spins for quantum applications, it is crucial to develop methods that enable stronger coupling between nuclear and central spins while simultaneously mitigating central spin and nuclear spin-induced decoherence (Figure 4.1a) [152].

One effective approach to address this trade-off is to leverage decoherence-protected states, quantum states engineered to be resilient against specific noise sources, such as magnetic field fluctuations [153–156]. Specifically, if nuclear spins experience common-mode noise from the central spin, highly entangled states such as Greenberger–Horne–Zeilinger (GHZ) states can provide decoherence protection [157]. By satisfying two key requirements, common-mode noise from central spins and the preparation of highly-entangled states, quantum information can be stored in decoherence-protected subspaces while maintaining strong coupling to central spins (Figure 4.2).

YVO_4 crystals containing dense nuclear spin ensembles with weakly doped ^{171}Yb ions, which serve as individually optically addressable qubits, present such a deterministic central spin system that can address these challenges [101]. Each ^{171}Yb ion controls four ^{51}V nuclear spins (register) that are frequency-detuned from the surrounding ^{51}V nuclear spin bath. Unlike sparse nuclear spin baths (such as ^{13}C weakly coupled to defects in diamond and SiC) [102, 158, 159] or dense nuclear spin bath (such as quantum dot systems) [160], our system features exactly four ^{51}V nuclear spins that form a many-body system symmetrically positioned around each ^{171}Yb ion. This symmetric arrangement ensures that the nuclear spin experience common-mode noise from the central spin and enables efficient, simultaneous control of the nuclear spins.

In this Chapter, we utilize ^{171}Yb to prepare ^{51}V GHZ states and perform high-resolution spectroscopy to sense different nuclear spin states. This capability facilitates exploration of decoherence-protected subspaces that are insensitive to common-mode noise and enables quantum information storage in ^{51}V GHZ states. This advancement is crucial for future scaling of quantum networks built on this platform.

4.1 Central Spin Qubits and Nuclear Spin Controls

The central spin platform employed in this Chapter consists of ^{171}Yb ions doped in Yttrium orthovanadate (YVO_4) crystals. This system features a 675 MHz clock transition (defining the qubit states $|0\rangle_g$ and $|1\rangle_g$) at zero magnetic fields and a 984.5 nm optical transition [106]. The optical transition is Purcell enhanced by integration with nanophotonic crystal cavities, enabling efficient optical control and readout (See Chapter 2) [91]. Previous demonstrations with this platform have achieved multiplexed remote entanglement distribution between ^{171}Yb ions in

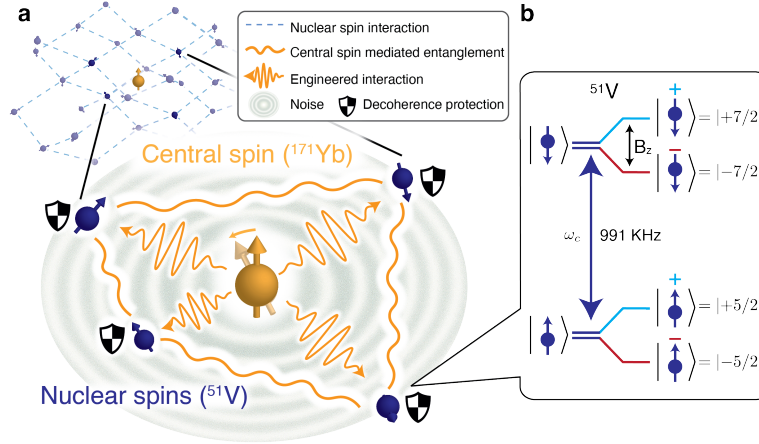


Figure 4.1: Nuclear-spin control in central spin systems. (a) Schematic of a central spin (orange, ^{171}Yb) system with nuclear spins (blue, ^{51}V). The crystal structure is shown on the top left. Weak interactions between nuclear spins are depicted by blue dash lines. The central part of the lattice is enlarged and shown below the lattice schematic. A central spin interacts with and controls (orange arrow) the nuclear spins via hyperfine interactions, facilitating entanglement between nuclear spins (orange wavy lines). When nuclear spins are prepared in specific configurations, the nuclear spin states are protected from central spin noise (background waves). (b) The energy level structure shows the c transition of the ^{51}V nuclear spin, separated by 991 KHz. At zero magnetic field, ^{51}V nuclear spins are doubly degenerate, consisting of $|\pm m_I\rangle$ with opposite Zeeman sensitivities.

separate devices and successful polarization and storage using W states of the four nuclear-spin register (See Chapter 2 and 3) [80, 101].

The nuclear spin environment surrounding each ^{171}Yb consists primarily of ^{51}V nuclei, which possess spin-7/2 and doubly degenerate energy levels composed of plus and minus m_I (i.e., $|\pm \frac{m_I}{2}\rangle$) states at zero magnetic field (Figure 4.1b). The ^{51}V nuclear spins are governed by the quadrupole Hamiltonian [123]:

$$\hat{H}_V = Q \hat{I}_z^2, \quad (4.1)$$

where $Q = 2\pi \times 165(171)\text{ KHz}$ for the register (bath) spins and $\hat{I}_z$ is the z-component of ^{51}V nuclear spin operators.

The four ^{51}V register spins are positioned equidistantly around each ^{171}Yb ion and interact through an induced dipolar interaction [101]. The ^{171}Yb - ^{51}V system

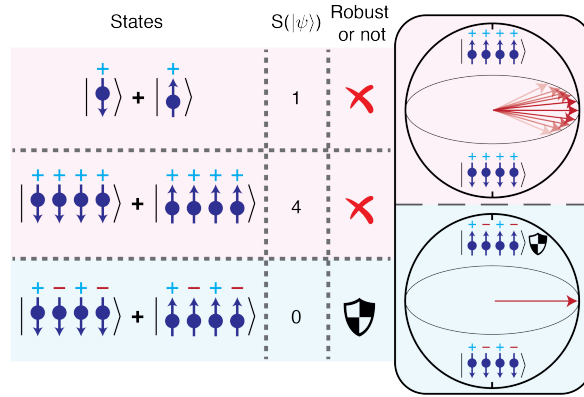


Figure 4.2: Table of highly entangled states and their sensitivity to common-mode magnetic field noise. The left column lists examples of the states. The middle column $S(|\psi\rangle)$ quantifies the common-noise sensitivity of the quantum state, normalized to that of a single-spin superposition. $S(|\psi\rangle) = 0$ indicates a protected state. The last column shows whether the superposition state is protected from the common noise field. States with zero noise sensitivity are robustly protected from common-mode noise. On the right, schematics of the dynamics of superposition states for non-decoherence-protected (top) and decoherence-protected (bottom) cases are shown on the Bloch sphere.

dynamics can be described by the Hamiltonian (See Section 2.4.1):

$$\begin{aligned} \hat{H}_{\text{sys}} = & \hat{H}_{\text{Yb}} + \sum_{i \in \text{register}} [\hat{H}_{\text{V},i} \\ & + \hat{S}_z (B^{\text{OH}} + B^{\text{RF}}(t)) (a_x \hat{I}_x^{(i)} + a_z \hat{I}_z^{(i)})], \end{aligned} \quad (4.2)$$

where $\hat{H}_{\text{Yb}}$ and $\hat{H}_{\text{V},i}$ include the energy levels and magnetic field dependent Hamiltonian terms that exclusively involve ^{171}Yb and ^{51}V , respectively. $\hat{S}_z$ is the ^{171}Yb qubit operator along z-axis, $\hat{I}_x$ is the ^{51}V register spin 7/2 operators along x-axis, B^{OH} is magnetic field noise on ^{171}Yb that is caused by nuclear spin baths (Overhauser field), $B^{\text{RF}}(t)$ is applied RF field, and a_x and a_z are coupling coefficients. For this work, we restrict our focus to the $|\pm\frac{5}{2}\rangle$ and $|\pm\frac{7}{2}\rangle$ subspace. We define $|+\frac{5}{2}\rangle = |+\uparrow\rangle$, $|-\frac{5}{2}\rangle = |-\uparrow\rangle$, $|+\frac{7}{2}\rangle = |+\downarrow\rangle$, and $|-\frac{7}{2}\rangle = |-\downarrow\rangle$. The frequency separation between the two states is denoted as ω_c , which is $2\pi \times 991$ KHz.

We engineer controlled rotation gates between ^{171}Yb and ^{51}V register spins by induced dipolar interactions (Figure 4.3) [126, 142]. An oscillating magnetic dipole of ^{171}Yb , which depends on the ^{171}Yb state, is induced by an oscillating RF field. The oscillating dipole creates driving fields on ^{51}V (Section 4.1.1). Since ^{51}V

nuclear spins occupy symmetric locations around ^{171}Yb , this creates a one (^{171}Yb) to all (^{51}V) controlled rotation gate. The effective Hamiltonian between ^{171}Yb and ^{51}V , expressed in the rotating frame of ^{51}V nuclear spins, is:

$$\hat{H}_{\text{eff}} = A_x \sum_{i \in \text{register}} B_{\omega_c}^{\text{RF}} \hat{S}_z \hat{I}_x^{(i)}, \quad (4.3)$$

where A_x is the coupling coefficient, $B_{\omega_c}^{\text{RF}}$ is the amplitude of the resonant RF field, and $\hat{I}_x$ is the ^{51}V register spin operator in the $|\uparrow\rangle$ and $|\downarrow\rangle$ basis. Note that here the $\pm$ signs of the register spin states are ignored because they are indistinguishable at zero magnetic fields. This effective Hamiltonian enables controlled rotation of all four ^{51}V register spins based on the ^{171}Yb state, forming the foundation for preparing and manipulating register spins.

4.1.1 Details of Controlled Rotation Gates

To engineer a deterministic gate between ^{171}Yb and ^{51}V , it is crucial to decouple the random Overhauser-induced noise field (B^{OH}) arising from the ^{51}V nuclear spin bath. This decoupling prevents stochastic interactions (second term in Equation 2.17) and ^{171}Yb dephasing (first term in Equation 2.17).

Here we designed a controlled rotation gate with ^{171}Yb as the controlled qubit and ^{51}V nuclear spins as the target qubits by applying an oscillating RF field at the ^{51}V transition frequency. The oscillating RF field induces a ^{171}Yb state dependent oscillating dipole, which generates RF fields and drives the ^{51}V spins. Importantly, the applied RF fields do not directly drive the ^{51}V nuclear spins since the RF fields are oriented in the z direction and therefore only induce detunings.

During the RF pulse sequence, XY8 dynamical decoupling pulses are applied to the ^{171}Yb at the node of the RF pulses. When an ^{171}Yb π pulse is applied, the interaction direction reverses (Equation 4.2); accordingly, we flip the phase of the oscillating RF field to maintain continuous driving of the ^{51}V spins (Figure 4.3). This protocol effectively cancels the Overhauser-induced noise field, leading to Equation 4.3 and generating the desired controlled rotation gate.

4.2 Yb-V GHZ State and ^{51}V Number Sensing

The controlled rotation gate enables simultaneous manipulation of all four ^{51}V , allowing efficient generation and coherence studies of highly entangled states. By combining $\pi/2$ controlled rotation gates and single qubit operations on ^{171}Yb , we

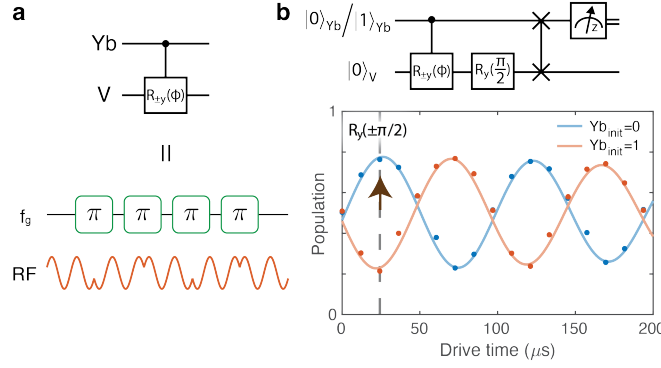


Figure 4.3: Controlled rotation gate sequence and experimental demonstration. (a) Gate sequence for implementing controlled rotation gates. ^{171}Yb π pulses are applied at the node of the RF oscillations, and the RF phase is flipped when a ^{171}Yb π pulse is applied. (b) Experimental results of demonstrating the controlled rotation gate. In the experiments, ^{171}Yb is initialized in different states, and ^{51}V is measured in the X basis after driving. The results show that the driving phase of ^{51}V depends on the initial state of ^{171}Yb . Experimental data, represented by blue and red dots, are fitted by oscillatory functions with gaussian decays (solid lines).

can prepare a Yb-V GHZ state of the form $(|0\rangle |\uparrow\uparrow\uparrow\uparrow\rangle + |1\rangle |\downarrow\downarrow\downarrow\downarrow\rangle)/\sqrt{2}$ (Figure 4.4) [157, 161].

The GHZ preparation begins with ^{171}Yb and ^{51}V initialized into $|0\rangle$ and $|\downarrow\rangle$, respectively. After a $\pi/2$ controlled rotation gate, a ^{171}Yb $\pi/2$ pulse prepares a ^{171}Yb superposition state, $(|0\rangle + |1\rangle)/\sqrt{2}$. Subsequently, a $\pi/2$ controlled rotation gate rotates all four ^{51}V nuclear spins depending on the ^{171}Yb state, thus creating a maximally entangled GHZ state between ^{171}Yb and ^{51}V . The state is characterized by applying the inverse preparation sequence followed by ^{171}Yb readout while varying the time between initialization and readout. Parity oscillations arise from the differential phase accumulation of the two GHZ state components during free evolution (Figure 4.4). The oscillation frequency is enhanced by a factor equal to the population imbalance $|N_1 - N_0| \times \omega_c$, where N_1 and N_0 represent the number of nuclear spins in the $|\uparrow\rangle$ and $|\downarrow\rangle$, respectively. These oscillations occur at $4\omega_c$ verifying that four spins are presented in the GHZ state [162]. However, by examining the Fourier spectrum of the parity oscillations (Figure 4.5), additional harmonics are observed, which arise from imperfect initialization of the nuclear spin ensemble. For example, the peak at $2\omega_c$ arises primarily from the state $|\uparrow\downarrow\downarrow\downarrow\rangle$.

The distinct oscillation frequencies associated with different nuclear spin configurations enable a post-selection protocol for improving ^{51}V state preparation fidelity.

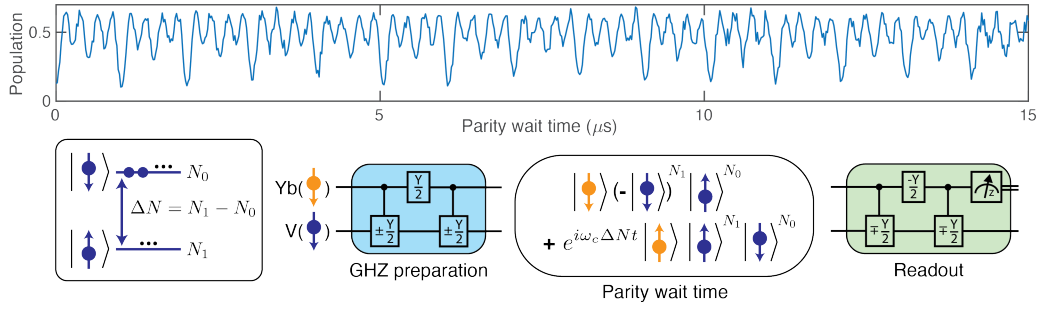


Figure 4.4: Yb-V GHZ state generation. The experimental results and sequence for Yb-V GHZ state generation utilizing $\pi/2$ controlled rotation gates. During the parity wait period, the state accumulates phase at $\Delta N = N_1 - N_0$ times ω_c . Here, N_0 and N_1 represent the number of Vanadium nuclear spins that are initially in $|\downarrow\rangle$ or $|\uparrow\rangle$ (bottom left), respectively. The sequence concludes with a reverse sequence to map the phase of the GHZ states to ^{171}Yb and optical readouts are performed on ^{171}Yb . The top left panel illustrates the measured GHZ state parity oscillations as the wait time is varied between the Yb-V GHZ preparation and readout sequence.

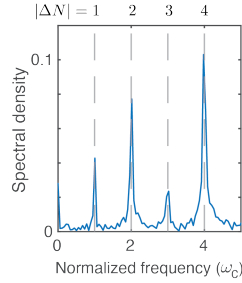


Figure 4.5: Fourier transform of the parity oscillations shown in Figure 4.4, revealing peaks at integer multiples of the Vanadium transition frequency, which arise from different initial states.

Different harmonic components exhibit different ^{171}Yb population outcomes when measured at specific parity wait times t_1 (Figure 4.6, see Section 4.2.1). By carefully choosing the parity wait time and applying appropriate single-qubit rotations before ^{171}Yb readout, we can selectively enhance desired nuclear spin configurations while suppressing unwanted ones. For instance, preparing all V in $|\downarrow\rangle$ and post-selecting at $t_1 = 1/4\omega_c$ selects the $4\omega_c$ component and rejects $2\omega_c$ component when applying an X gate before ^{171}Yb readout. Experiment results (Figure 4.7, $4\omega_c$ case) show a larger contribution of the $4\omega_c$ component and a suppression of $2\omega_c$ compared to the non-post-selected case (Figure 4.5), indicating successful post-selection. This method provides a way to sense $|N_1 - N_0|$ and improve the initialization fidelity of

^{51}V . Multiple stages of post-selection can further enhance the initialization fidelity (Figure 4.8).

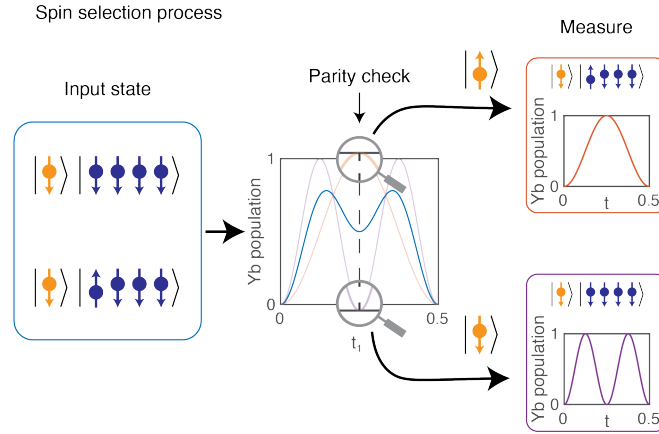


Figure 4.6: Vanadium spin number sensing. ^{51}V spin number sensing using the Yb-V GHZ state generation sequence. Phase accumulation depends on the initial ^{51}V state (left) and wait time (t_1), with the accumulated phase proportional to ΔN (middle). The blue curve shows the parity oscillations of the mean of the two initial states on the left. The purple curve represents the parity oscillation with spins initialized into $|\downarrow\downarrow\downarrow\downarrow\rangle$, while the red curve represents the parity oscillation with spins initialized into $|\uparrow\downarrow\downarrow\downarrow\rangle$. Specific harmonics can be selected by reading out ^{171}Yb at different wait times (right).

4.2.1 V Spin Number Sensing and Post-selection Details

The Yb-V GHZ state generation protocol, illustrated in Figure 4.4, enables the selection of different states by varying the wait time between the generation and readout sequences. The detailed sequence used to probe this spin selection process is shown in Figure 4.9. In this sequence, ^{171}Yb is initialized in the $|0\rangle$ state and ^{51}V is prepared in the $|\downarrow\rangle^{N_0} |\uparrow\rangle^{N_1}$ state. After the GHZ state preparation sequence (see Figure 4.4), ^{171}Yb and ^{51}V become entangled, resulting in the state:

$$\frac{1}{\sqrt{2}}(|0\rangle (-|\downarrow\rangle)^{N_1} |\uparrow\rangle^{N_0} + |1\rangle |\downarrow\rangle^{N_0} |\uparrow\rangle^{N_1}). \quad (4.4)$$

After a free evolution time t_1 and the subsequent readout sequence, the final state is given by:

$$\frac{1 + e^{i\Delta N\omega_c t_1}}{2} |0\rangle |\downarrow\rangle^{N_0} |\uparrow\rangle^{N_1} + \frac{-1 + e^{i\Delta N\omega_c t_1}}{2} |1\rangle |\uparrow\rangle^{N_0} (-|\downarrow\rangle)^{N_1}, \quad (4.5)$$

where ^{171}Yb exhibit parity oscillations that depend on ΔN .

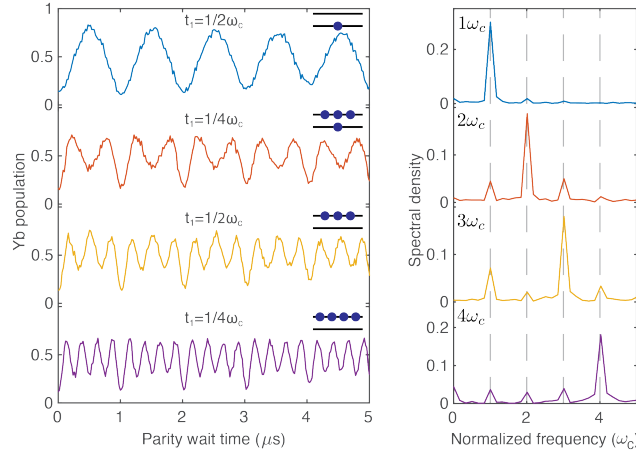


Figure 4.7: Experimental result of harmonic selection through post-selection. The left panels show the parity oscillations after post-election. The insets show the main contributing states, which is determined by different ^{51}V initialization maximizing the contribution of selected harmonics. The post-selection times (t_1) of different harmonics are shown at the middle top for each harmonics. The right panels show the corresponding Fourier spectrum. ^{51}V nuclear spins are initialized differently for different harmonic measurements. For example, a single spin is initialized into $|\uparrow\rangle$ for $1\omega_c$ post-selection (Section 4.2.1).

To selectively enhance specific harmonics contributions (Figure 4.6), both the initial state and the wait time t_1 are tailored for each case. For $1\omega_c$, ^{51}V is first prepared to ^{51}V $|3/2\rangle$ state using ZenPol on the b and c transitions. A subsequent swap on the b transition with ZenPol prepares a single spin excitation in $|\uparrow\rangle$ state [101]. The wait time is set to $t_1 = 1/\omega_c/2$ to suppress $2\omega_c$ and $4\omega_c$ contributions. For $2\omega_c$, ^{51}V is fully initialized into $|\downarrow\rangle$, and the $2\omega_c$ signal arises from the initialization infidelity. $t_1 = 1/\omega_c/4$ is chosen to suppress the $4\omega_c$ component. For $3\omega_c$, ^{51}V is fully initialized into $|\downarrow\rangle$ and a π pulse on b transition is applied to shelve residual population in $|\uparrow\rangle$ to $|3/2\rangle$. t_1 is set to be $1/\omega_c/2$. For $4\omega_c$, ^{51}V is fully initialized into $|\downarrow\rangle$ and $t_1 = 1/\omega_c/4$.

For two-stage spin selection (Figure 4.8), harmonics from $1\omega_c$ to $3\omega_c$ can be suppressed by employing two distinct wait times: $t_1 = 1/\omega_c/4$ and $t_2 = 1/\omega_c/2$. In all the spin selection measurements, to maximize the readout fidelity and the effect of readout optical pulses on the ^{51}V states, only 20 optical π pulses are applied to readout ^{171}Yb $|1\rangle$ state.

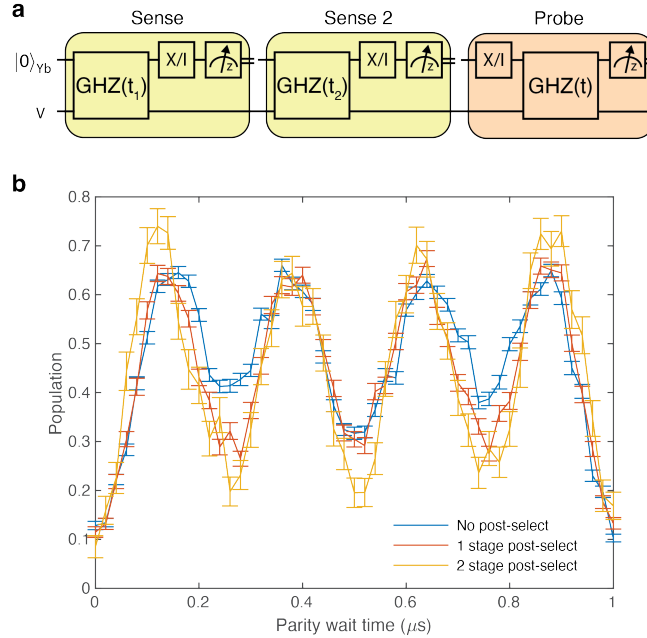


Figure 4.8: Two-stage ^{51}V spin number selection. (a) Experimental sequence for two-stage ^{51}V spin number selection. The time interval t_1 is set to $1/4/\omega_c$ to eliminate the $2\omega_c$ contribution, and t_2 is set to $1/2/\omega_c$ to suppress the $1\omega_c$ and $3\omega_c$ contributions. (b) Experimental results of demonstrating spin number selection at different stages. The contrast of the parity oscillations is enhanced with the implementation of two-stage spin number selection.

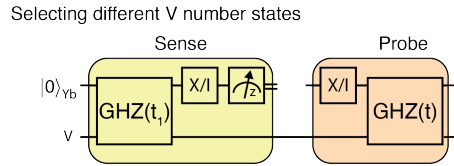


Figure 4.9: Experimental sequence for selecting different ^{51}V number states. The detailed sequence for GHZ block is shown in Figure 4.4.

4.3 Sensing the Decoherence Protected Subspace

In the previous section, the GHZ state consists of both ^{171}Yb and ^{51}V , therefore its coherence is limited by the relatively short ^{171}Yb Ramsey coherence time ($8\ \mu\text{s}$). To study the ^{51}V decoherence dynamics, we apply an alternative GHZ state preparation and measurement sequence (Figure 4.10), which circumvents the ^{171}Yb coherence limit. Specifically, an entangled state between ^{171}Yb and ^{51}V is prepared, but the subsequent measurement sequence does not depend on the ^{171}Yb coherence, ensuring a coherence measurement that is only affected by the nuclear spins (Section 4.3.1).

^{51}V nuclear spins experience magnetic field noise from both the bath nuclear spins and ^{171}Yb . Bath nuclear spins create magnetic field noise with different field strengths on different ^{51}V register spins; whereas, due to the symmetrical position of ^{51}V register spins around ^{171}Yb , the ^{51}V register spins experience common-mode noise from ^{171}Yb . Due to the short distance between ^{171}Yb and ^{51}V register spins, ^{171}Yb creates a noise field on ^{51}V that is stronger than the magnetic field noise directly from bath nuclear spins (Section 4.3.3, [101]), and ^{51}V register spin coherence is limited by common-mode magnetic field noise generated by ^{171}Yb .

To understand the decoherence effect of ^{171}Yb magnetic field noise on different GHZ states, we need to consider the relative number of nuclear spins initially in the $|+\downarrow\rangle$ and $|-\downarrow\rangle$ states. Specifically, spins with opposite Zeeman sensitivity ($|+m_I\rangle$ and $| -m_I\rangle$) acquire quantum phases of equal magnitude but opposite sign in a common magnetic field, so their contributions cancel. Figure 4.2 lists the resulting relative sensitivities for several nuclear-spin GHZ states.

To explore this magnetic field sensitivity, we apply a DC magnetic field via an externally-induced ^{171}Yb magnetic dipole moment (see Figure 4.10 and Section 4.3.1 for details). We probe the resulting frequency shift by taking a Fourier transform of the time-dependent GHZ-state parity oscillations. This measurement is repeated at different magnetic field values, and the Fourier transforms are horizontally stacked in Figure 4.11. Focusing specifically on the four-spin case ($|\uparrow\uparrow\uparrow\uparrow\rangle + |\downarrow\downarrow\downarrow\downarrow\rangle$), we find that the resonance at 3.96 MHz splits into five distinct frequencies associated with different initial nuclear spin states (Figure 4.10). Most notably, initial states consisting of an equal number of opposite-sensitivity magnetic sublevels ($|+\downarrow + \downarrow - \downarrow - \downarrow\rangle$) experience no net frequency shift and are therefore insensitive to common-mode noise.

To verify the insensitivity of this decoherence-protected subspace to common-mode noise fields, we artificially introduced a fluctuating magnetic field, randomly sampled from a Gaussian distribution with a specified standard deviation (Figure 4.12a). Each trace in Figure 4.12a contains all possible $|+m_I\rangle$ and $| -m_I\rangle$ combinations. As the standard deviation increases (from faint to solid traces in Figure 4.12a), the non-decoherence-protected subspace (consisting of initial states such as $|+\downarrow + \downarrow + \downarrow - \downarrow\rangle$ and $|+\downarrow + \downarrow + \downarrow + \downarrow\rangle$) exhibits faster decay due to sensitivity to common-mode noise. The decay is limited by both uncorrelated nuclear Overhauser field noise and common-mode noise from the ^{171}Yb . In contrast, the decoherence-protected subspace maintained unchanged coherence decay, limited

solely by nuclear Overhauser field noise. For each trace, the two dominant decay timescales are extracted and plotted in Figure 4.12b, where the long decay has a timescale of $164 \pm 8 \mu\text{s}$. This demonstrates the insensitivity of the ^{51}V GHZ state decoherence-protected subspace to the common-mode noise field, which is crucial for operating ^{171}Yb while maintaining coherent quantum information storage on ^{51}V .

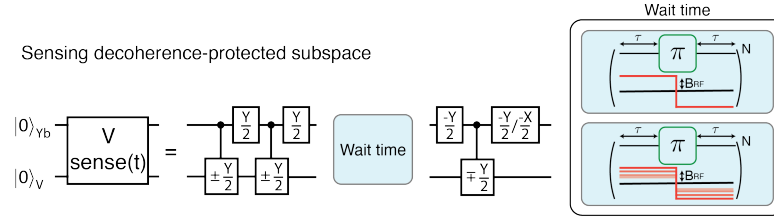


Figure 4.10: Experimental sequence for sensing the decoherence-protected subspace. This sequence is used to generate Figure 4.11 and 4.12. The right-hand side illustrates the sequences used to generate DC fields and artificial noise on ^{51}V .

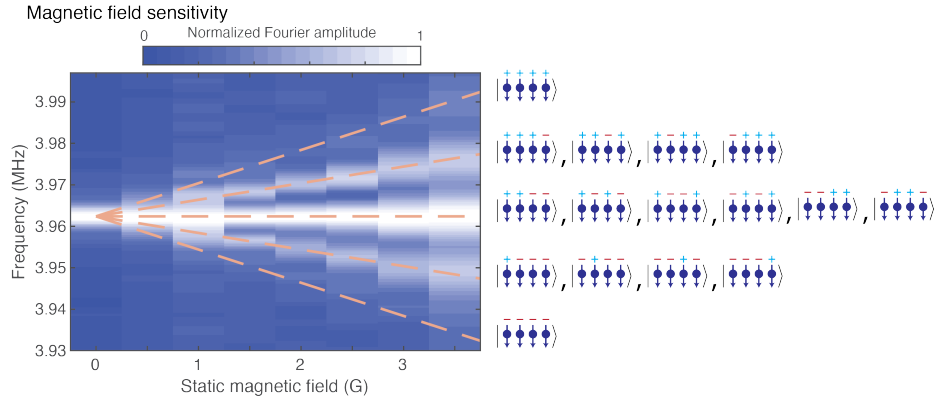


Figure 4.11: Sensing of the decoherence protected subspace of ^{51}V . Experimental results of ^{51}V GHZ states under different DC magnetic fields, applied through Yb using square wave RF pulses interleaved with Yb π pulses. Each vertical axis data is obtained by Fourier transforming the time-domain measurement results. The $4\omega_c$ resonance exhibits field-dependent splitting. The orange lines are fitted lines for the split peaks. The magnetic field-dependent and independent peaks originate from the non-decoherence-protected and decoherence-protected subspaces, respectively. The states that contribute to each specific peak are shown on the right of the plot.

4.3.1 Detailed gate sequence for sensing the decoherence-protected subspace

In this section, we developed a pulse sequence to probe ^{51}V GHZ coherence using only three controlled rotation gates (Figure 4.10). During the wait period,

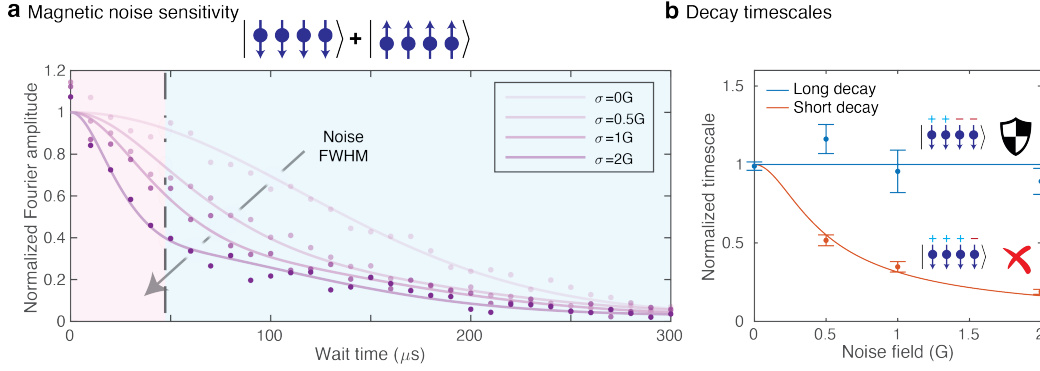


Figure 4.12: Decoherence-protected subspace of GHZ states. (a) Testing the decoherence-protected subspace by applying arbitrary common noise fields (σ) through ^{171}Yb . Here, we apply common noise fields through ^{171}Yb . The noise field is sampled using a Gaussian distribution with different standard deviations for different curves. Nuclear spins are fully initialized into $7/2$ state and the decay is fitted by Gaussian decays with different timescales and fixed amplitude ratios (solid lines), where the amplitude ratios are determined by the probability of being in specific states. The colored dots are the experimental data. The pink and blue regions indicate non-decoherence protected decay and decoherence-protected decay, respectively. With increasing noise field, the coherence decay of the decoherence-protected subspace is unaffected, but the coherence decay of the non-decoherence-protected subspace becomes faster. (b) The fitted short and long timescale decays for different noise fields. The fitted timescale is normalized to the long timescale ($164\text{ }\mu\text{s}$). The longer decay is fitted with a constant line. The short decay is fitted by considering a fixed gaussian decay from the long decay and a gaussian decay with a timescale proportional to $\frac{1}{\sigma}$.

^{171}Yb remains entangled with ^{51}V ; however, by measuring in a specific basis, the measurement outcome becomes independent of ^{171}Yb coherence.

Assuming ^{171}Yb and ^{51}V are initialized to $|0\rangle$ and $|\downarrow\rangle^{N_0} |\uparrow\rangle^{N_1}$, respectively, the joint state during the wait time is given by:

$$\begin{aligned} & \frac{1}{2} (|0\rangle (|\uparrow\rangle^{N_0} (-|\downarrow\rangle^{N_1}) - e^{i\Delta N\omega_c t} |\downarrow\rangle^{N_0} |\uparrow\rangle^{N_1}) \\ & + |1\rangle (|\uparrow\rangle^{N_0} (-|\downarrow\rangle^{N_1}) + e^{i\Delta N\omega_c t} |\downarrow\rangle^{N_0} |\uparrow\rangle^{N_1})). \end{aligned} \quad (4.6)$$

If the wait time exceeds ^{171}Yb Ramsey coherence time (about $8\text{ }\mu\text{s}$), the state decoheres into a mixed state, allowing $|0\rangle$ and $|1\rangle$ components to be treated independently. After applying the first two gates of the readout sequence, the states corresponding

to $|0\rangle$ during the wait time evolve into:

$$\begin{aligned} & \frac{1}{2}[(|0\rangle + e^{i\Delta N\omega_c t} |1\rangle) |+\rangle^{N_0} (-|-\rangle)^{N_1} \\ & + (-e^{i\Delta N\omega_c t} |0\rangle - (-1)^{N_0+N_1} |1\rangle) |-\rangle^{N_0} |+\rangle^{N_1}], \end{aligned} \quad (4.7)$$

while the states corresponding to $|1\rangle$ become:

$$\begin{aligned} & \frac{1}{2}[(|0\rangle + e^{i\Delta N\omega_c t} |1\rangle) |+\rangle^{N_0} (-|-\rangle)^{N_1} \\ & (e^{i\Delta N\omega_c t} |0\rangle + (-1)^{N_0} |1\rangle) |-\rangle^{N_0} |+\rangle^{N_1}]. \end{aligned} \quad (4.8)$$

Finally, by measuring along the X-axis of ^{171}Yb through the last $\pi/2$ pulse when ΔN is even, or along the Y-axis when ΔN is odd, we can extract the coherence time of ^{51}V GHZ states.

4.3.2 Generation of DC fields on ^{51}V

To measure the magnetic field sensitivities of GHZ states, we apply DC magnetic fields to ^{51}V as shown in Figure 4.11. Since we operate at zero magnetic fields for ^{171}Yb , we employ a specialized pulse sequence to generate effective DC fields on ^{51}V (Figure 4.10).

The sequence consists of a square wave RF field applied to ^{171}Yb , with ^{171}Yb π pulses synchronized to occur at the nodes (zero-crossing points) of the RF field. This design exploits the coupling between ^{171}Yb and ^{51}V , which depends on the ^{171}Yb S_z operator (Equation 4.2).

The key principle is that while phase accumulation from the direct oscillating magnetic field applied to ^{51}V sums to zero over each RF cycle, the magnetic field induced by ^{171}Yb accumulates constructively. This occurs because the ^{171}Yb π pulses flip the sign of the ^{171}Yb -induced magnetic field at precisely the same moments when the external RF field changes sign. The result is an effectively constant (DC) magnetic field experienced by ^{51}V throughout the sequence.

An additional benefit of this approach is that the ^{171}Yb π pulses cancel the Overhauser-induced ^{171}Yb dipole field, thereby enhancing the overall sensitivity of the measurement sequence.

4.3.3 ^{51}V coherence limits

The coherence of ^{51}V register spins is limited by two distinct magnetic field noise sources: dipolar interactions with the ^{51}V bath and ^{51}V bath induced ^{171}Yb dipole, which generates common-mode noise.

The magnetic noise arising from nuclear dipolar interactions with the ^{51}V bath is calculated as:

$$B(\mathbf{r}_V) = \sum_{i \in \text{bath}} \frac{\mu_0 \mu_N g_{Vz}}{4\pi(\mathbf{r}_V - \mathbf{r}_i)} (3(n^{(Vi)} - 1)m_I^{(i)}), \quad (4.9)$$

where $\mathbf{r}_V$ is the position of the register nuclear spin and $n^{(Vi)}$ is the z-direction cosine of the ^{51}V - ^{51}V displacement vector. This interaction produces magnetic field fluctuations with a standard deviation of 0.35 G at each register spin position.

For an ^{51}V nuclear spin, this bath-induced noise limits the coherence to approximately 680 μs . When considering a four-spin GHZ state, the uncorrelated nature of the field noise at different register sites reduces the collective coherence to 340 μs , scaling as $T_2^N = T_2^1/\sqrt{N}$ for N spins, where the superscript denotes the number of spins.

The magnetic field noise from ^{171}Yb introduces correlated noise across the register spins, calculated from the second-order perturbation term in Equation 2.17. This limits single-spin coherence to 51 μs , shorter than the bath-limited timescale.

For multi-spin entangled states, the decoherence rate depends on the specific spin configuration due to the common-mode nature of the ^{171}Yb noise. The most probable state ($|+\downarrow+\downarrow+\downarrow-\downarrow\rangle$) exhibits a coherence time of approximately 26 μs , consistent with the short-timescale decay observed in Figure 4.14.

4.4 V GHZ State Storage and Applications

One of the key applications of this decoherence-protected subspace is auxiliary quantum storage while performing operations on the central spin qubit, e.g., generating spin-photon entanglement.

This requires a gate set for faithfully mapping quantum information between the ^{171}Yb central spin and the ^{51}V nuclear spin decoherence-protected subspace. We develop a sequence that can swap ^{171}Yb states, with a basis of $|0\rangle$ and $|1\rangle$, to ^{51}V states, with a basis of $|\downarrow\downarrow\downarrow\downarrow\rangle$ and $|\uparrow\uparrow\uparrow\uparrow\rangle$ (Figure 4.13a, Figure 4.14). We demonstrate this protocol by swapping six Bloch cardinal states onto this GHZ-state basis and back to ^{171}Yb to characterize the storage (Figure 4.13a). The average fidelity after swapping back is 0.71, which is limited by the ^{51}V initialization and controlled rotation gate fidelities.

Combined with the ability of multiplexed remote entanglement distribution, the decoherence-protected nuclear spin storage can be used to perform teleported gates

and entanglement purification. A protocol for achieving this is shown in Figure 4.13b and discussed in Section 4.4.3.

4.4.1 Detailed gate sequence for ^{51}V storage

We developed a gate sequence for storing quantum information in the ^{51}V basis states $|\downarrow\downarrow\downarrow\downarrow\rangle$ and $|\uparrow\uparrow\uparrow\uparrow\rangle$ (Figure 4.15). A ^{51}V Z-rotation gate is incorporated into the sequence to enable swapping of quantum states between ^{171}Yb and ^{51}V . The Z rotation is generated by applying dynamical decoupling pulses to ^{171}Yb while allowing phase evolution on ^{51}V .

At the start of the sequence, ^{171}Yb and ^{51}V are initialized into $a|0\rangle + b|1\rangle$ and $|\downarrow\downarrow\downarrow\downarrow\rangle$, respectively. Before the Z rotation gate, the joint state is:

$$a|0\rangle|\uparrow\uparrow\uparrow\uparrow\rangle + a|1\rangle|\downarrow\downarrow\downarrow\downarrow\rangle - b|0\rangle|\downarrow\downarrow\downarrow\downarrow\rangle + b|1\rangle|\uparrow\uparrow\uparrow\uparrow\rangle. \quad (4.10)$$

After applying a Z rotation gate and a controlled rotation gate, the state evolves to:

$$ia(|0\rangle - i|1\rangle)|+++\rangle - b(|0\rangle - i|1\rangle)|---\rangle. \quad (4.11)$$

Finally, an X rotation on ^{171}Yb , followed by the controlled rotation, completes the sequence, resulting in:

$$|0\rangle(ia|\downarrow\downarrow\downarrow\downarrow\rangle - b|\uparrow\uparrow\uparrow\uparrow\rangle). \quad (4.12)$$

The inverse of the gate sequence can be used to retrieve the quantum information back into ^{171}Yb . The coherence of the stored superposition state, shown in Figure 4.14, exhibits a two-timescale decay: a fast decay associated with the non-decoherence-protected subspace, and a slower decay corresponding to the decoherence-protected subspace.

4.4.2 Protocol for deterministic preparation into the decoherence-protected subspace

We develop a protocol for deterministically preparing ^{51}V nuclear spins into the decoherence-protected subspace ($|+\downarrow+\downarrow-\downarrow-\downarrow\rangle$). While experimental demonstration of this protocol remains challenging due to the low fidelity of nuclear spin control sequences, the theoretical framework provides a pathway for achieving decoherence-protected state preparation.

The protocol begins with a GHZ storage sequence (Figure 4.15), where ^{171}Yb is prepared in a superposition state (Figure 4.16). During the storage period, a DC magnetic field is applied to the ^{51}V spins. The resulting ^{51}V GHZ states accumulate

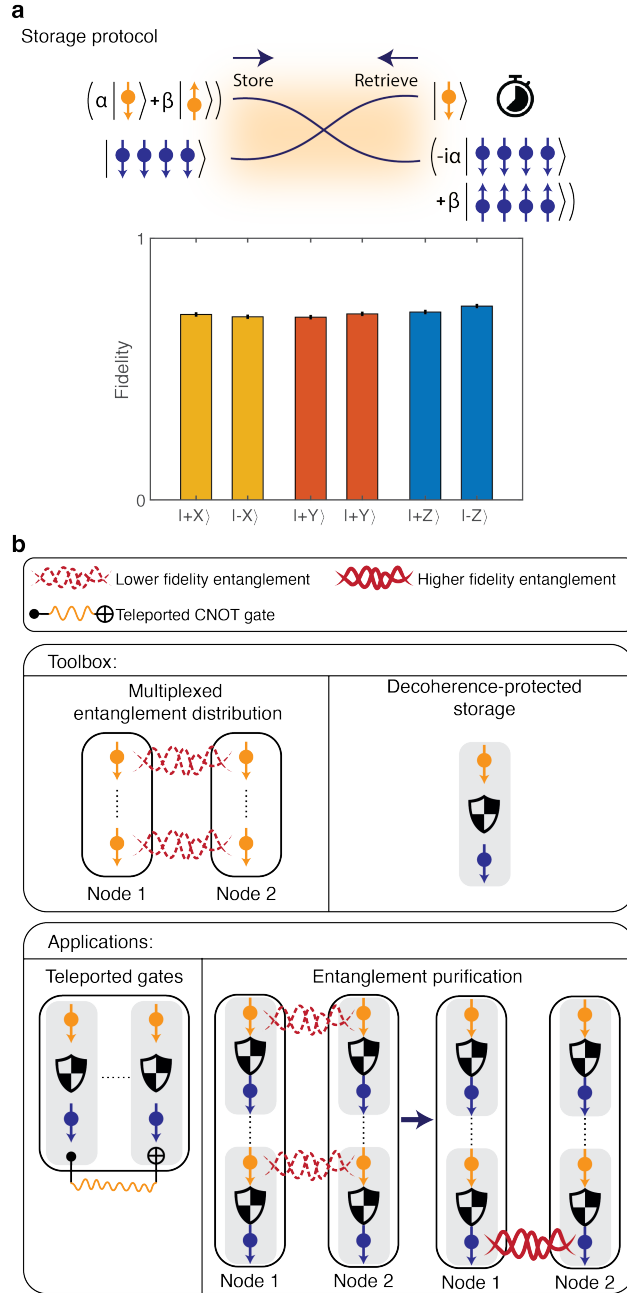


Figure 4.13: Applications of decoherence-protected nuclear spin storage in a multiplexed quantum network node. (a) Experimental demonstration of swapping information from ^{171}Yb $|0\rangle$ and $|1\rangle$ to ^{51}V $|\downarrow\downarrow\downarrow\downarrow\rangle$ and $|\uparrow\uparrow\uparrow\uparrow\rangle$ with a schematic shown at the top. Experimental results of swapping the state into ^{51}V and swapping back to ^{171}Yb are shown at the bottom. ^{171}Yb is prepared in six cardinal Bloch states and the retrieved states are characterized through quantum state tomography. (b) Toolbox and applications of a decoherence-protected nuclear spin storage in a multiplexed quantum network node. The toolbox includes multiplexed remote entanglement distribution and decoherence-protected storage. With these capabilities, teleported gates and entanglement purification can be performed.

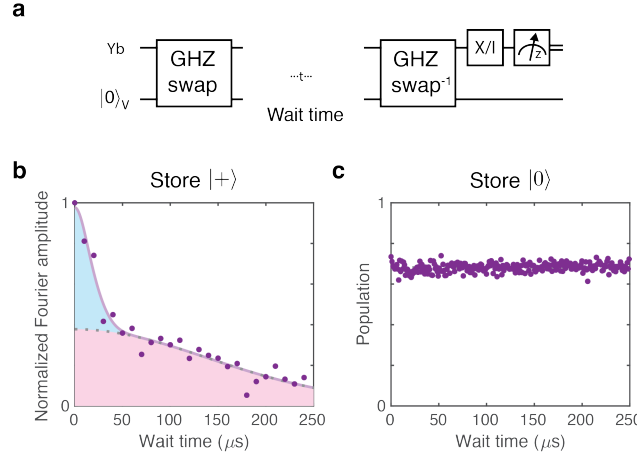


Figure 4.14: Storage time in ^{51}V $|\downarrow\downarrow\downarrow\downarrow\rangle$ and $|\uparrow\uparrow\uparrow\uparrow\rangle$ bases. (a) Experimental sequence for measuring storage time. The GHZ swap sequence, as described in Figure 4.15, is used to swap the ^{171}Yb state onto ^{51}V , which is then retrieved after certain waiting period. (b) Storage time for the $|+\rangle$ state. The Y-axis shows the $4\omega_c$ Fourier transformed amplitude of the parity oscillations. Experimental data, represented by purple dots, are fitted by Gaussian decay curves (solid lines). The short decay has a timescale of $27 \pm 4 \mu\text{s}$, and the long decay is $200 \pm 40 \mu\text{s}$. (c) Storage time for the $|0\rangle$ state. The decay timescale corresponds to the ^{51}V T_1 time 0.5 second, which is much longer than the probing period.

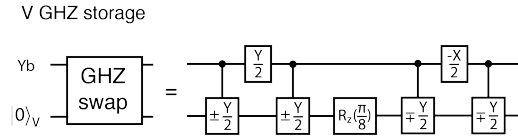


Figure 4.15: Experimental sequence for performing swap between $|0_g\rangle/|1_g\rangle$ and $|\downarrow\downarrow\downarrow\downarrow\rangle/|\uparrow\uparrow\uparrow\uparrow\rangle$.

phase shifts that depend on their magnetic field sensitivity. By carefully selecting the wait time and magnetic field strength, we can induce different phase shifts between the two components of the GHZ states for states with different magnetic field sensitivities. After the state swaps back to ^{171}Yb , optical readouts can be performed to distinguish different states.

After applying two sensing sequences, we can determine the current ^{51}V state. If the state already resides in the decoherence-protected subspace, it is accepted without further manipulation.

When all four spins share the same sign, we apply ZenPol sequence twice to flip two spins to $|\uparrow\rangle$. Subsequently, controlled rotation π pulses are applied sequentially

to the b and a transitions of ^{51}V to prepare the state in $|1/2\rangle$. A magnetic field is then applied to split $|+1/2\rangle$ and $|-1/2\rangle$ states, enabling driving of the transition between the two states. Once ^{51}V is driven to the opposite sign, controlled rotation π pulses are applied consecutively on the a and b transitions, followed by ZenPol sequences to prepare the final $|\downarrow\rangle$ state. This procedure deterministically converts both $|+\uparrow+\uparrow+\uparrow+\uparrow\rangle$ and $|-\uparrow-\uparrow-\uparrow-\uparrow\rangle$ into the target state $|+\downarrow+\downarrow-\downarrow-\downarrow\rangle$.

When three spins share the same sign, we apply a single ZenPol sequence to flip one spin to $|\uparrow\rangle$. The same procedure described above is then applied to flip the sign of this single spin. This approach succeeds in preparing the decoherence-protected state 75% of the time, while the remaining 25% results in a state with four spins of identical signs. An additional round of ^{51}V sensing determines whether the system has reached the decoherence-protected subspace. If not, then the procedure for four spins that share the same sign is applied to prepare ^{51}V in the decoherence-protected subspace.

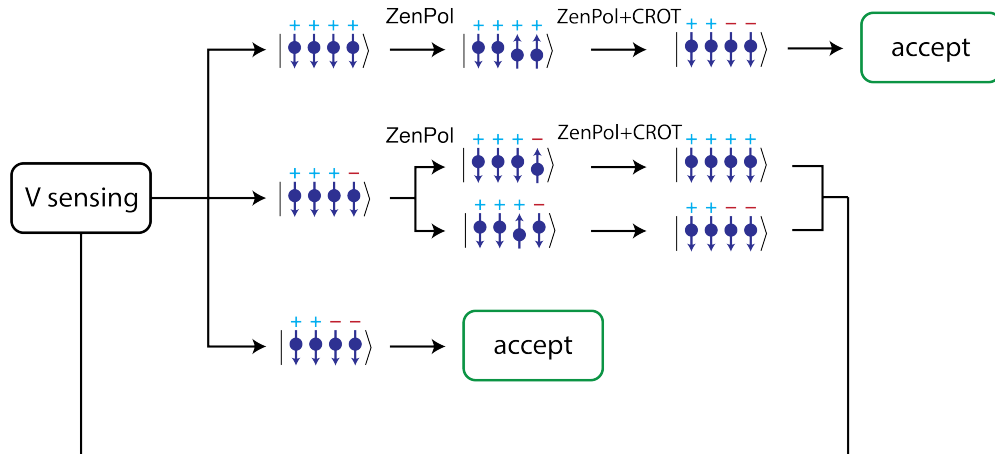


Figure 4.16: Protocol for deterministic initialization into the decoherence-protected subspace. A ^{51}V sensing sequence is first applied with a DC magnetic field to sense the state of the nuclear spins. If ^{51}V is not in the decoherence-protected subspace, ZenPol is applied to flip either one or two ^{51}V spins depending on the sensed state. After flipping ^{51}V , controlled rotation gates on the a and b transitions of ^{51}V are applied to prepare the flipped ^{51}V to $|1/2\rangle$. With an applied DC field, controlled rotation gate can flip the sign of the $|1/2\rangle$ state. Then ZenPol or controlled rotation gates can be applied to polarize the spins to $|7/2\rangle$. Depending on the initial state, this leads to preparation of a decoherence-protected state, or another round of ^{51}V sensing can be applied.

4.4.3 Entanglement Purification Simulations

As discussed in Figure 4.13, we propose using multiple emitters, each coupled to a decoherence-protected storage within a node, to implement enhanced entanglement purification across two nodes. This section details the entanglement purification procedures and simulation parameters.

The entanglement purification procedure begins with multiplexed entanglement distribution across two nodes (Figure 4.17), exploiting the availability of multiple optically addressable qubits for entanglement distribution [43, 101]. Following each successful entanglement distribution event, entanglements are stored in nuclear spins for decoherence protection.

Once two pairs of successful entanglement distributions are established, teleported gates [29] are performed between nuclear spins within the same node. This is achieved by remotely entangling the corresponding ^{171}Yb ions to execute the entanglement purification protocol [38]. By measuring one entangled pair in the Z basis, post-selection can be applied to the remaining pair, thereby improving the entanglement fidelity according to:

$$F' = \frac{F_1 F_2 + \frac{1}{9}(1 - F_1)(1 - F_2)^2}{F_1 F_2 + \frac{1}{3}F_1(1 - F_2) + \frac{1}{3}F_2(1 - F_1) + \frac{5}{9}(1 - F_1)(1 - F_2)}, \quad (4.13)$$

where F' is the purified fidelity, and F_1 and F_2 are the fidelities of the unpurified pairs.

The simulation (Figure 4.17b) demonstrates that increasing the number of ions within a single node enables entanglement purification at lower link efficiencies (η_{link}). The link efficiency is defined as the ratio of the entanglement generation rate (γ_{ent}) to the decoherence rate.

The simulation assumes an entanglement attempt duration of $10 \mu\text{s}$ when the two nodes are not separated, with a protocol overhead of $8 \mu\text{s}$ [140] that can be improved through multiple emitters and entanglement multiplexing. The distance between two nodes is assumed to be 5 km. All single-qubit gates of the emitter and storage, as well as two-qubit gates between emitter and nuclear spin storage, are assumed to be perfect. Remote entanglement between two central spins within the same node is also assumed to be perfect.

The central spins are assumed to have 10 ms coherence times, while nuclear spins have coherence times equal to $\eta_{\text{link}}/\gamma_{\text{ent}}$. Since ^{171}Yb emits at 984.5 nm, the fiber

attenuation is assumed to be 3.5 dB per km. The remote entanglement fidelity is assumed to be 0.8, and the readout time is assumed to be 100 μ s.

The simulation compares two scenarios:

- Single emitter configuration: Each node contains only one emitter, with entanglement purification performed using entanglement distribution between single emitters and nuclear spin storage [163].
- Multiple emitter configuration: Each node contains multiple emitters with enhanced entanglement rates through multiplexing [44, 45]. Entanglement purification is performed using two pairs of nuclear spins.

Using the parameters and assumptions detailed above, we find that the multiple-ion configuration enables improved entanglement fidelity at lower values of η_{link} , demonstrating the advantage of multiplexed quantum network nodes with decoherence-protected storage.

4.5 Simulation of GHZ States

We simulate the Yb-V system using the effective Hamiltonian described in Section 2.4.1. Specifically, we apply rotating wave approximation and transform Equation 2.14 to ^{171}Yb rotating frame. Overhauser fields from ^{51}V bath are assumed to remain static throughout the pulse sequences. We sample different bath configurations by assuming fully thermalized bath spins for each Monte-Carlo simulation repetition. The register dynamics are simulated in a reduced Hilbert space by considering only ω_c manifold. ^{171}Yb π pulses are modeled with finite pulse durations.

The initial states of ^{51}V spins are sampled from a probability distribution, where each ^{51}V register spin has probability ϵ_1 of being in $|\uparrow\rangle$ and probability ϵ_2 of being outside of ω_c manifold. We note that this constitutes an assumption about the system's initial state. Under this framework, we cannot identify values of ϵ_1 and ϵ_2 that would deterministically prepare ^{51}V in specific states such as $|\downarrow\downarrow\downarrow\uparrow\rangle$. Nevertheless, this assumption is necessary to avoid an underdetermined system that would result from introducing free parameters for each possible initial state when fitting the Fourier spectrum of Yb-V GHZ state parity oscillations (Figure 4.5).

In this work, we present only simulation results for Yb-V GHZ state parity oscillations because our simulation framework cannot capture infidelities arising from controlled rotation gates [101]. This limitation prevents us from fully reproducing

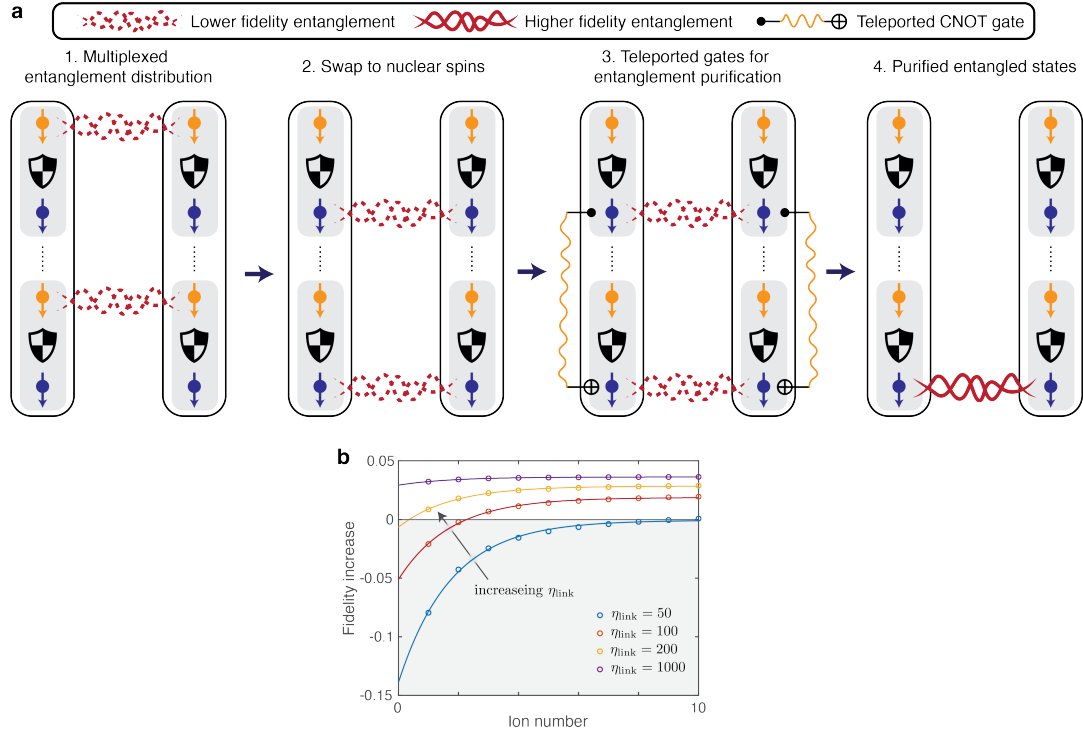


Figure 4.17: Applications of decoherence-protected nuclear spin storage in a multiplexed quantum network node. (a) Protocol for entanglement purification in a multiplexed quantum network node. Orange spins represent optically addressable central spins, and the blue spins indicate decoherence-protected nuclear spin storage. First, multiplexed entanglement attempts are performed between nodes. Then, successfully entangled pairs are swapped to the decoherence-protected nuclear spin storage. Once two successful pairs of entanglement are established, entanglement purification is performed using teleported gates mediated by central spin qubits. Lastly, the purified entangled state can be used for further purification or other applications. Dashed lines between spins represent lower-fidelity entangled pairs. Solid lines between spins indicate purified or higher fidelity entangled pairs. (b) Simulations of how the multiplexing and nuclear spin storage improve entanglement purification. The figure illustrates the fidelity increase achieved through a single round of entanglement purification, plotted as a function of ion number. Simulations (colored dots) were conducted using different link efficiencies, defined as the ratio of entanglement rate to coherence decay rate. Solid lines are fitted as guided lines.

the experimental results through simulation. Future studies must address the discrepancies between experimental and simulation results. Figure 4.18a shows the comparison between simulated and experimental parity oscillations of the Yb-V GHZ state. The simulation is optimized to reproduce the ratio between different harmonics (Figure 4.18b). The optimal values are $\epsilon_1 = 0.14$ and $\epsilon_2 = 0.09$,

respectively.

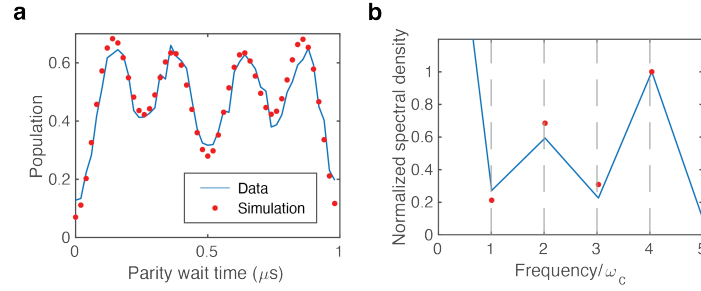


Figure 4.18: Simulation of Yb-V GHZ parity oscillations. (a) Comparison between experimental data (blue curve) and simulation results (red dots) for Yb-V GHZ state parity oscillations. (b) Comparison between experimental data (blue curve) and simulation results (red dots) for the Fourier spectrum of Yb-V GHZ state parity oscillations.

4.6 Summary and Outlook

In this Chapter, we prepare and characterize various quantum states within the four ^{51}V register spin system via tailored gate sequences. We demonstrate preparation of highly entangled GHZ states consisting of both ^{171}Yb and ^{51}V spins and their application in sensing the number of ^{51}V nuclear spins in specific states. Additionally, we devise a sequence for sensing $|+m_I\rangle$ and $|-m_I\rangle$ of ^{51}V nuclear spins, revealing a decoherence-protected subspace in our symmetric four-spin system. We also develop and validate a sequence capable of storing information within this subspace. With our symmetric and deterministic nuclear spin register, these results provide a pathway to overcome the trade-off between nuclear spin control speed and decoherence from non-unitary central spin evolution.

Looking ahead, it is essential to control $|+m_I\rangle$ and $|-m_I\rangle$ manifolds independently to initialize into the decoherence-protected subspace deterministically. This can be achieved by sensing the states through the application of DC magnetic fields. For states that are sensitive to the applied magnetic fields, either one or two ^{51}V spins are driven to $|\uparrow\rangle$. Then, through driving the ^{51}V transitions, we can deterministically initialize into the decoherence-protected subspace (Section 4.4.2, Figure 4.16). Enhancing the gate fidelity and ^{51}V initialization fidelity is also crucial for implementing these nuclear spin memories in quantum networking experiments [164]. One possible way of improving the gate fidelity is to reduce the gate time. The gate time of the controlled rotation gate is currently limited by the power we can

apply to the chip without heating the system. One solution is to fabricate chips with superconducting coplanar waveguides rather than gold to reduce heating [165]. To further expand the platform's capabilities, we aim to explore driving different ^{51}V nuclear spin shells. This can further increase the Hilbert space we are working with and create opportunities for studying many-body physics in the nuclear spin platform [166, 167]. These advancements pave the way for pushing the boundaries of quantum control and sensing in solid-state systems.

Chapter 5

$^{171}\text{Yb}:\text{YVO}_4$ AS A QUDIT SYSTEM FOR QUANTUM NETWORKING

A qudit operates in a d -dimensional Hilbert space ($d > 2$), contrasting with the 2-dimensional space of qubits. This higher dimensionality confers several computational advantages: qudits can encode more information per quantum unit, thereby reducing circuit depth and gate complexity in quantum algorithms while decreasing hardware overhead in quantum computing implementations [168]. In quantum communication protocols, higher-dimensional quantum states demonstrate superior performance characteristics. Qudit-based quantum key distribution systems achieve enhanced key generation rates and exhibit greater tolerance to quantum bit errors compared to their qubit counterparts [169]. Qudits also serve as essential resources for advanced quantum information applications. They facilitate efficient generation of high-dimensional photonic cluster states [170]. Additionally, qudits enable implementation of novel quantum networking protocols [171, 172]. These capabilities position qudits as critical components in scalable quantum network architectures, offering enhanced efficiency and robustness compared to conventional qubit-based systems.

Several experimental platforms have demonstrated successful qudit implementations. Some examples include trapped ion systems utilizing multiple Zeeman sub-levels [173], superconducting transmon qubits with access to higher energy levels [174], solid-state qubits [175], and photonic systems employing path or time-bin encoding [176, 177]. Rare-earth-ion-doped crystals have emerged as particularly promising candidates due to their long coherence times and well-defined energy level structures [80, 178].

The majority of previous work on the $^{171}\text{Yb}:\text{YVO}_4$ system has focused on the two-level qubit subspace formed by the ground state levels ($|0\rangle_g$ and $|1\rangle_g$) [80, 91, 101]. While this qubit implementation has demonstrated exceptional versatility with optical access, the complete hyperfine structure of this system provides access to four distinct energy levels, expanding the computational capabilities. Specifically, the auxiliary doublet $|aux\rangle_g$, which would be degenerate in an ideal crystal environment, exhibits a splitting of several tens of MHz due to local site asymmetry. These

split levels form clock transitions, whose frequencies are first-order insensitive to magnetic field fluctuations, providing inherent protection against environmental noise. Transitions between the $|0\rangle_g$, $|1\rangle_g$ states and the $|aux\rangle_g$ states also constitute clock transitions, collectively enabling a robust four-level qudit. Given that qudit-based quantum networking protocols are an emerging area of active research [179], the following sections will present the experimental platform and comprehensive characterization demonstrating that $^{171}\text{Yb}:\text{YVO}_4$ represents a promising candidate for qudit-based quantum networking applications.

5.1 System Hamiltonian and Device Requirements

While $^{171}\text{Yb}:\text{YVO}_4$ possesses a four-level structure ideal for qudit encoding, these levels have remained inaccessible due to the control requirements. Universal control of the four-level ground state manifold demands magnetic field components that cannot be generated by standard planar devices aligned with the crystal's high-symmetry axes. As discussed in Section 2.3.3, the $|aux\rangle_g$ states in $^{171}\text{Yb}:\text{YVO}_4$ exhibit a splitting of 10-100 MHz due to crystal field asymmetry. This splitting is postulated to be caused by the asymmetry in the hyperfine $\mathbf{A}_g$ tensor between the crystal a-axes. For instance, consider the hyperfine tensor:

$$\mathbf{A}_g = 2\pi \times \begin{bmatrix} 0.625 & 0 & 0 \\ 0 & 0.725 & 0 \\ 0 & 0 & -4.82 \end{bmatrix} \text{GHz} \quad (5.1)$$

, where the asymmetry between the x- and y-axes results in a 50 MHz splitting of the $|aux\rangle_g$ doublet. The resulting non-degenerate auxiliary states have eigenvectors:

$$|aux\rangle_{g0} = (|\uparrow_{\text{Yb}}\uparrow_{\text{Yb}}\rangle + |\downarrow_{\text{Yb}}\downarrow_{\text{Yb}}\rangle)/\sqrt{2}, \quad (5.2)$$

$$|aux\rangle_{g1} = (|\uparrow_{\text{Yb}}\uparrow_{\text{Yb}}\rangle - |\downarrow_{\text{Yb}}\downarrow_{\text{Yb}}\rangle)/\sqrt{2}. \quad (5.3)$$

The magnetic field requirements for driving transitions between these states present a design challenge. Transitions between $|aux\rangle_{g0}$ and $|aux\rangle_{g1}$ require magnetic fields along the crystal c-axis, while transitions between the primary qubit states $|0, 1\rangle_g$ and the auxiliary states $|aux\rangle_{g0,g1}$ require magnetic field components along the crystal a-axes. To address this requirement, we developed a novel photonic crystal cavity design where the device z-axis is oriented between the crystal a- and c-axes (Figure 5.1(a)). This geometry represents an elegant solution, enabling a planar coplanar waveguides to generate magnetic field components along both crystal directions, providing access to all four qudit levels.

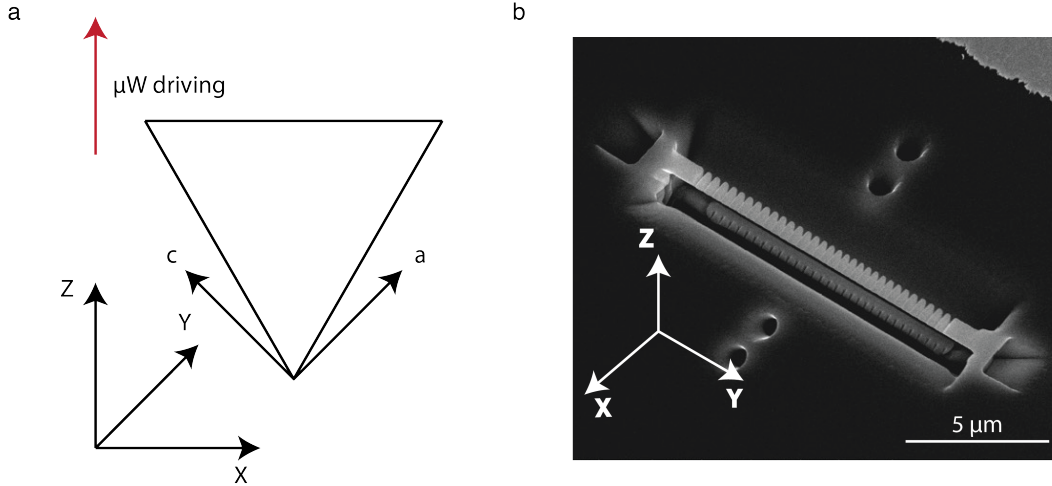


Figure 5.1: Device platform for four-level qudit operation. (a) Schematic showing crystal axes orientation relative to the device geometry. (b) SEM image of the fabricated photonic crystal cavity device.

The birefringent properties of YVO_4 enable cavity mode engineering despite the non-aligned crystal axes. The crystal exhibits different refractive indices along the a - and c -axes (1.96 and 2.16 at 980 nm, respectively), allowing us to design cavity modes aligned with specific crystal directions even when the crystal and device axes are misaligned. For optimal cyclicity of the A transition, the cavity field must be aligned along the crystal c -axis rather than the a -axis. Through minor modifications of our established c -cut device design, we identified design parameters that maintain cavity modes along the crystal c -axis while preserving similar simulated Q-factors and mode volumes to our previous c -cut implementations. Using the fabrication procedures described in Section 2.2.1, we successfully fabricated devices achieving Q-factors comparable to our c -cut devices (Figure 5.1(b)).

5.2 Experimental Setup

The 45° -cut device is operated in a ^3He fridge at base temperature (0.5 K). The optical experimental setup remains identical to that described in Chapter 4, utilizing the same laser stabilization, detection, and control systems established for qubit operations.

For qudit operation, additional microwave driving capabilities are required to access transitions between the qubit and auxiliary manifolds. The frequency separations between the qubit states ($|0\rangle_g$, $|1\rangle_g$) and auxiliary manifolds ($|\text{aux}\rangle_{g0}$, $|\text{aux}\rangle_{g1}$) are

approximately 2050 MHz and 2700 MHz, respectively. To coherently drive these transitions, we implemented a heterodyne mixing setup that combines a 3.3 GHz frequency source (Holzworth HS9002A) with an arbitrary waveform generator (AWG) channel (Tektronix 5204AWG). The mixed signal is subsequently amplified using a Mini-circuits ZX05-U432H-S+ amplifier and filtered through a Mini-circuits ZBSS-2250-S+ filter to suppress image frequency components.

The microwave driving system incorporates a switch that alternates between the qubit-auxiliary transition drive and the excited state microwave drive operating at 3.37 GHz. Transitions within the auxiliary manifold ($|\text{aux}\rangle_{g0} \leftrightarrow |\text{aux}\rangle_{g1}$) are driven directly through the AWG, utilizing the same driving scheme as the primary qubit transitions. This configuration provides complete control over all four qudit levels and enables coherent single-qudit operations (Figure 5.2).

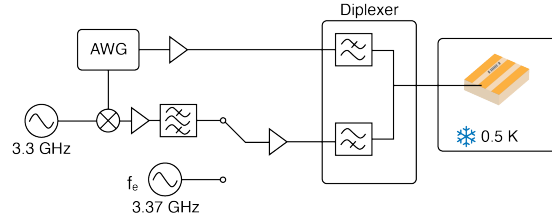


Figure 5.2: Microwave driving setup for four-level qudit control. The heterodyne mixing system enables coherent driving of transitions between qubit and auxiliary manifolds.

5.3 Qudit Characterization and Coherence

The coherence properties of the four-level qudit system were characterized using the 45°-cut devices described above. The following sections summarize the key performance metrics that establish the viability of this platform for quantum networking applications.

5.3.1 Optical Properties

Individual ^{171}Yb ions were identified through photoluminescence scans around the A transition wavelength (Figure 5.3(a)). A well-isolated ion was selected for detailed characterization, as indicated in the spectrum. Using the nanophotonic crystal cavity, we measured a Purcell-enhanced optical lifetime of $1.612 \pm 0.009 \mu\text{s}$ (Figure 5.3(b)), similar to the emission lifetimes observed in our previous *c*-cut devices (Chapter 3).

Optical cyclicity, which is critical for high-fidelity state readout, was characterized through repeated optical excitations. The ion was first initialized to $|1\rangle_g$, followed by 1000 consecutive optical readout cycles. The population remaining in $|1\rangle_g$ after N optical π pulses was determined by histogramming photon counts as a function of pulse number. The measured optical cyclicity of 0.99493 ± 0.00007 is slightly lower than our c -cut devices (~ 0.997) but remains sufficiently high for reliable optical readout operations.

The reduced cyclicity in the 45° -cut geometry can be mitigated by leveraging the additional microwave driving capabilities. Since the $|0\rangle_g \leftrightarrow |0\rangle_e$ transition is forbidden, transferring population from $|\text{aux}\rangle$ manifold to $|1\rangle_g$ through microwave driving after optical readouts can effectively increase the apparent cyclicity when operating within the qubit subspace, providing operational advantages of the 45° -cut geometry.

5.3.2 Qudit properties

Following optical characterizations, the microwave properties of the four-level qudit system were characterized through spin coherence measurements. Figure 5.4(a) illustrates the complete energy level structure, comprising the qubit manifold ($|0\rangle_g$ and $|1\rangle_g$) and the auxiliary manifold ($|\text{aux}\rangle_{g0}$ and $|\text{aux}\rangle_{g1}$).

To identify transitions between the qubit and auxiliary manifolds, we performed optically detected magnetic resonance (ODMR) measurements (Figure 5.4(b)). The ^{171}Yb ion is first prepared in the $|1\rangle_g$ state through F transition pumping and qubit initialization. Subsequently, 1000 optical A transition π pulses are applied to deplete the population from the qubit manifold into the auxiliary states. Microwave driving is then applied at various frequencies, and resonant transitions are identified by the recovery of population to the qubit manifold, detected through optical readout. For transitions that bring auxiliary states to $|0\rangle_g$, an additional qubit π pulse transfers the population to $|1\rangle_g$ for optical detection.

This ODMR approach successfully identified all four transitions connecting the qubit and auxiliary manifolds, enabling coherent spin Rabi oscillations between these states (Figure 5.4(c)). Notably, different transitions exhibit varying Rabi frequencies under similar driving power, which we attribute to the orientation of the hyperfine tensor asymmetry relative to the coplanar waveguide geometry. In the ^{171}Yb reference frame, x-directed drives couple $|\text{aux}\rangle_{g1} \leftrightarrow |0\rangle_g$ and $|\text{aux}\rangle_{g0} \leftrightarrow |1\rangle_g$ transitions, while y-directed drives couple $|\text{aux}\rangle_{g0} \leftrightarrow |0\rangle_g$ and $|\text{aux}\rangle_{g1} \leftrightarrow |1\rangle_g$

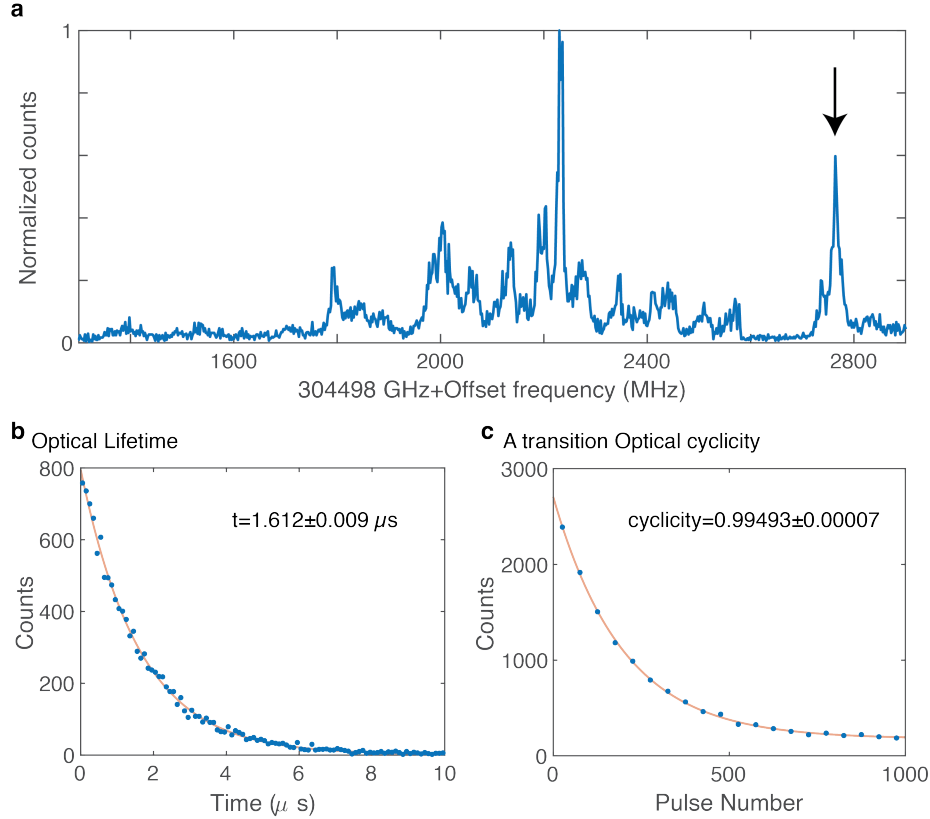


Figure 5.3: Optical characterization of ^{171}Yb ions in 45° -cut devices. (a) Photoluminescence spectrum around the A transition showing individual ion peaks; the highlighted peak corresponds to the ion used for subsequent measurements. (b) Optical lifetime measurement revealing Purcell-enhanced emission with $\tau = 1.612 \pm 0.009 \mu\text{s}$. (c) Optical cyclicity measurement yielding $C = 0.99493 \pm 0.00007$.

transitions. The relative orientation between the ion's hyperfine frame and the waveguide determines the observed Rabi frequency variations.

The capability to coherently control qubit-auxiliary transitions enables enhanced initialization protocols. By replacing F transition pumping with A transition excitation combined with excited state microwave driving, interleaved with qubit-auxiliary microwave transfers, we achieve improved depopulation of auxiliary states and reduced initialization time. This optimization reduces overall sequence duration and enhances fidelity for remote entanglement protocols. Due to the approximately 7-fold reduction in the g-factor along the crystal a-axis compared to the c-axis, stronger microwave fields are required for efficient qubit-auxiliary transitions.

The coherence properties of the transition within the auxiliary manifold were characterized by standard pulse sequences. As expected, the $|\text{aux}\rangle_{g0} \leftrightarrow |\text{aux}\rangle_{g1}$ transition

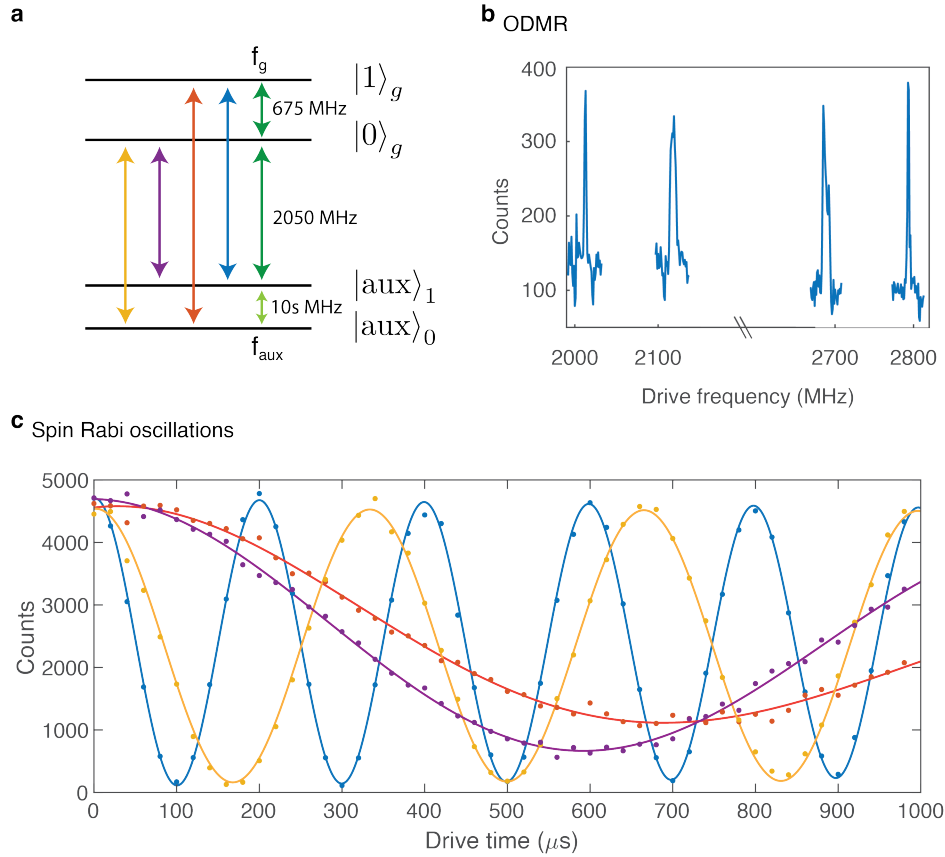


Figure 5.4: Microwave characterization of the four-level qudit system. (a) Energy level structure showing qubit and auxiliary manifolds. (b) ODMR spectrum identifying transitions between qubit and auxiliary states. (c) Coherent Rabi oscillations for all four qubit-auxiliary transitions.

exhibits reduced coherence compared to the primary qubit transition due to increased magnetic field sensitivity ($\Delta \propto B^2/\omega_{ij}$, where Δ represents detuning and ω_{ij} is the transition frequency between qubit states or auxiliary states).

Figure 5.5(a) shows the detuned spin Ramsey coherence, with experimental data well-described by simulations incorporating static Overhauser field noise from vanadium nuclear spins. Spin echo measurements reveal a coherence time of about $13.2 \mu\text{s}$ with pronounced modulations attributed to strong ^{171}Yb - ^{51}V coupling (Figure 5.5(b)).

To extend coherence times, we implemented XY8 dynamical decoupling sequences with fixed interpulse spacing of $5.856 \mu\text{s}$ and variable pulse number. This approach extends the auxiliary transition coherence time to $2.27 \pm 0.15 \text{ ms}$ (Figure 5.5(c)). Finally, the spin lifetime of the auxiliary transition was measured to be $15.4 \pm 1.7 \text{ ms}$

(Figure 5.5(d)), establishing the fundamental limit for auxiliary state storage.

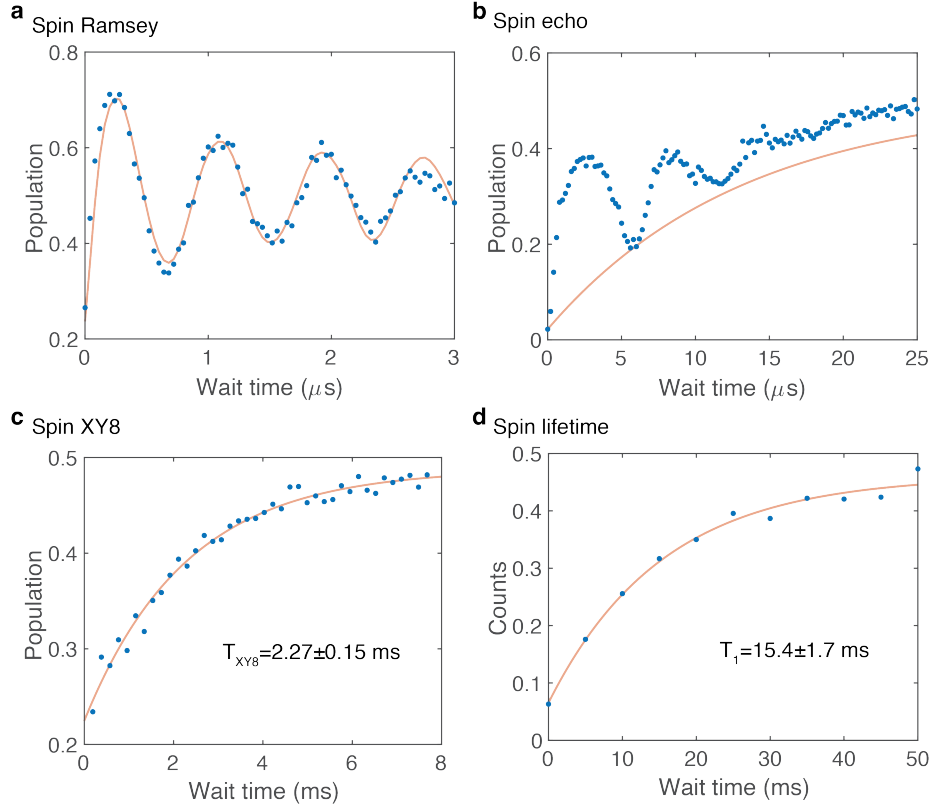


Figure 5.5: Coherence characterization of the auxiliary transition f_{aux} . (a) Ramsey dephasing measurements (blue points) with simulation including vanadium Overhauser noise (red line). (b) Spin echo decay (blue points) overlaid with an $13.2 \mu\text{s}$ exponential decay (red line). (c) XY8 dynamical decoupling extending coherence to $2.27 \pm 0.15 \text{ ms}$. (d) Spin lifetime measurement yielding $15.4 \pm 1.7 \text{ ms}$.

5.4 Nuclear Spin Control

A key distinction between the auxiliary and qubit manifolds lies in their magnetic field sensitivity, with the auxiliary manifold exhibiting significantly higher susceptibility to magnetic field noise. This enhanced sensitivity also results in a larger induced dipole moment compared to the qubit manifold (see Chapter 4), with the enhancement factor given by $\omega_{0,1} / \omega_{\text{aux}0,\text{aux}1}$, where $\omega_{0,1}$ and $\omega_{\text{aux}0,\text{aux}1}$ represent the transition frequencies of the qubit and auxiliary manifolds, respectively.

This enhanced coupling enables more efficient nuclear spin control through the auxiliary states. Figure 5.6(a) demonstrates Rabi frequencies exceeding 20 kHz achieved by applying the controlled rotation sequences described in Chapter 4 using auxiliary manifold states. The ability to drive ^{51}V nuclear spins with higher Rabi

frequencies at the same power levels, mediated through the ^{171}Yb auxiliary states, effectively addresses heating issues that arise when implementing multiple nuclear spin control pulses in sequence.

However, we observe that the oscillation decay rate increases with higher Rabi frequencies (Figure 5.6(b)), presenting a trade-off between control speed and coherence preservation. The physical origin of the shorter decay at higher driving powers remains under investigation.

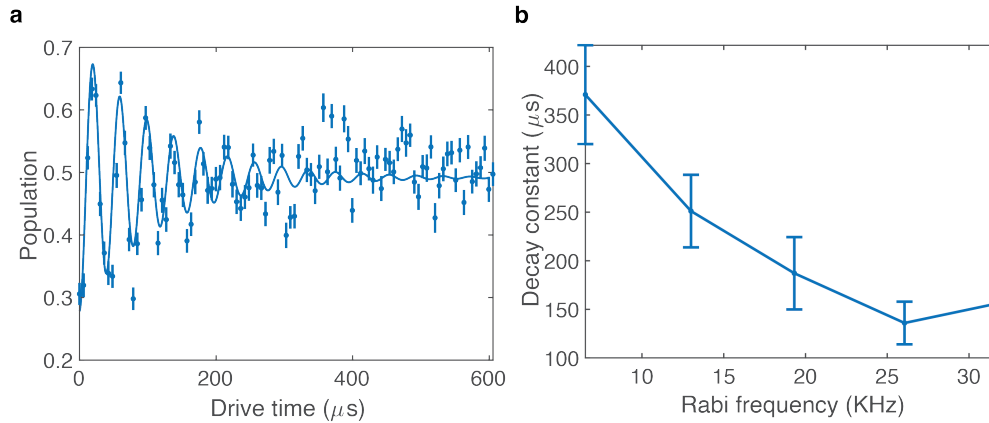


Figure 5.6: ^{51}V direct driving using auxiliary manifold. (a) Direct driving with varying drive time. (b) Rabi frequencies and decay constants of the Rabi oscillations.

5.5 Outlook

In this chapter, we have presented the design, fabrication, and coherent control of a novel photonic crystal cavity enabling four-level qudit operation in $^{171}\text{Yb}:\text{YVO}_4$. Through combined optical and microwave control, we demonstrated access to the full ground-state hyperfine manifold and characterized the qudit-level coherence and control fidelity. These capabilities establish $^{171}\text{Yb}:\text{YVO}_4$ as a promising solid-state platform for exploring qudit-based quantum networking.

To advance beyond single-qudit demonstrations and enable entanglement between spatially separated qudits, further improvements in coherence are essential. In particular, the echo coherence of transitions involving the auxiliary manifold presently limits the fidelity of prospective qudit–qudit entanglement protocols. One promising pathway to mitigate this limitation is the controlled tuning of the auxiliary state splitting via engineered strain fields [180]. Crystal strain can potentially increase the splitting of the $|\text{aux}\rangle_g$ states. A larger energy separation would reduce their sensitivity to magnetic field fluctuations and enhance coherence times.

An alternative approach involves moving to more favorable host materials. For example, CaWO_4 has been proposed as an attractive platform for rare-earth doping due to its reduced nuclear spin bath and coherent optical and spin transitions [181]. The use of host materials with lower nuclear spin noise could allow longer coherence times.

From a functionality perspective, a key milestone will be the demonstration of qudit–photon entanglement [182], a prerequisite for distributing qudit states across a quantum network. This capability will unlock the implementation of higher-dimensional quantum protocols [171, 172]. Adapting existing entanglement protocols for qudit states thus represents a clear next step.

In addition to their role in networking protocols, the 45° -cut photonic crystal cavities developed for qudit control also enable new opportunities for nuclear spin memory. In particular, they allow access to the first-shell ^{51}V nuclear spins, which are located directly above and below the ^{171}Yb ions. These nuclear spins couple to the electronic spin primarily via an Ising-type interaction and are expected to offer long spin lifetimes and strong electron–nuclear coupling. Critically, driving these first-shell spins requires magnetic fields along the crystal a -axis, a requirement that is naturally fulfilled in the 45° -cut geometry. This makes the device platform suited for investigating controlled interactions with proximal ^{51}V spins and implementing high-performance nuclear spin quantum memories [101, 142].

Together, these advances chart a clear path toward expanding the role of qudits within solid-state quantum networks. By combining higher-dimensional encoding, coherent optical control, and spin-based memory, $^{171}\text{Yb}:\text{YVO}_4$ continues to demonstrate the flexibility and scalability needed for next-generation quantum networking systems.

CONCLUSION AND OUTLOOK

In this thesis, we have demonstrated both nuclear spin control and remote entanglement distribution between ^{171}Yb ions in separate devices, establishing fundamental building blocks for quantum networking. Our work significantly extends the capabilities of quantum nodes by achieving several key advances: multiplexed remote entanglement between two pairs of spectrally distinguishable ^{171}Yb ions (Chapter 3), probing decoherence-protected GHZ states using nuclear spins (Chapter 4), and characterizing qudit coherence properties for future qudit quantum networking experiments (Chapter 5). While these results demonstrate a promising pathway toward building scalable quantum networks using ^{171}Yb doped in YVO_4 , several limitations remain that must be addressed to enable scaling to multiple nodes and long-distance quantum networking.

6.1 Nuclear Spin Storage Enhancement

One of the primary limitations stems from our current nuclear spin storage implementation. We utilize an ensemble of vanadium nuclear spins for information storage, and while coherent control of ensemble nuclear spins has been demonstrated, the control fidelity remains limited. This constraint prevents the implementation of multiple swap gates between ^{171}Yb ions and the additional nuclear spin storage. Furthermore, the current nuclear spin storage coherence time of approximately 1 ms requires improvement to enable entanglement swapping and purification between nodes.

The primary limiting factors of nuclear spin control remain unclear. Including $|aux\rangle_g$ states in our theoretical simulations and incorporating higher-order contributions could provide deeper insights into these limitations. Alternative approaches include exploring other nuclear spin defects for quantum information storage [102] or utilizing additional ^{171}Yb ions as storage elements. If multiple ^{171}Yb ions can be positioned in proximity, dipolar interactions could enable two-qubit gates between them. Realizing this capability would require precise implantation within small regions of fabricated devices, allowing several atoms to occupy the small mode volume where they can interact strongly [103].

6.2 Improving Entanglement Rate and Fidelity

The current platform's entanglement rate and fidelity present another significant limitation. The entanglement rate is primarily constrained by finite detection efficiency, with low cavity-to-waveguide coupling and waveguide-to-fiber coupling resulting in limited overall detection efficiency (2.5%). Several approaches could address these coupling inefficiencies, including tapered fiber coupling [137] and anti-reflective coatings to reduce surface reflections.

Cavity-to-waveguide coupling could be enhanced by reducing radiative and scattering losses from the cavities. Currently, the fabricated Q factor falls below the designed value, with performance limited primarily by scattering losses. Fabrication improvements offer a direct path to better cavity performance. While we currently employ gallium ion beam fabrication, transitioning to neon or helium ion beams could produce photonic crystal cavities with smoother surfaces, thereby reducing scattering losses. The availability of thin-film YVO₄ would enable more scalable fabrication methods with reduced turnaround times [183], potentially yielding superior devices with higher quality factors and coupling efficiencies. Improved coupling efficiency and quality factors would directly enhance entanglement fidelity (see Chapter 3).

6.3 Long-Distance Networking Capabilities

Extending networking capabilities to long distances requires converting 980 nm single photons to the 1550 nm telecommunications wavelength. Quantum frequency conversion has been successfully demonstrated with SiV and NV centers to telecom wavelengths [58, 184], providing a proven methodology that could be adapted to extend our networking distance capabilities.

6.4 Future Research Directions

Several exciting research avenues exist in the field. Co-doping with different rare-earth ions presents opportunities for both additional quantum networking resources and exploration of many-body physics [78, 87, 122]. Investigation of alternative host materials with reduced nuclear spin environments could yield longer coherence times [79, 93, 181].

The high flexibility offered by different platforms, combined with the exceptional multiplexing capabilities enabled by inhomogeneous broadening, positions rare-earth ions in solid-state hosts as excellent candidates for future quantum networking developments. The foundation established in this thesis provides a solid platform

for addressing the identified limitations and advancing toward practical, scalable quantum networks.

While challenges remain in scaling to multi-node architectures and long-distance implementations, the identified improvement pathways provide directions for future development. The combination of enhanced fabrication techniques, improved coupling methods, alternative storage approaches, and wavelength conversion capabilities offers a comprehensive roadmap for advancing this quantum networking platform toward practical applications.

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Appendix A

HYBRID QUANTUM PHOTONIC INTERFACE FOR $^{171}\text{Yb}^{3+}:\text{YVO}_4$

Previously, we demonstrated that $^{171}\text{Yb}^{3+}$ ions inside YVO_4 have spin coherence times exceeding 10 ms, over 99.9% single-qubit gate fidelities, 95% post-selected optical readout and initialization fidelity, and additional nuclear spin qubit control which are essential properties for building a quantum network [91, 101]. In rare-earth ion platforms, coherent 4f-4f optical transitions are only weakly allowed inside crystals, therefore it is essential to couple ions to cavities with large quality factors and small mode volumes to increase the emission rate through Purcell enhancement. However, the nanofabrication of monolithic nanophotonic cavities for common rare-earth host materials is limited due to the unavailability of high-quality thin films and selective etching chemistries (aside from a few examples [92, 185, 186]). Here we demonstrate a near-infrared hybrid platform comprising a separately fabricated photonic crystal cavity that is subsequently transferred onto the host crystal (Figure A.1) [95, 187]. This platform possesses a smaller mode volume compared to Fabry-Perot microcavities [188, 189], and unlike the previous approach of focused ion beam milling [91, 109], does not lead to crystal damage.

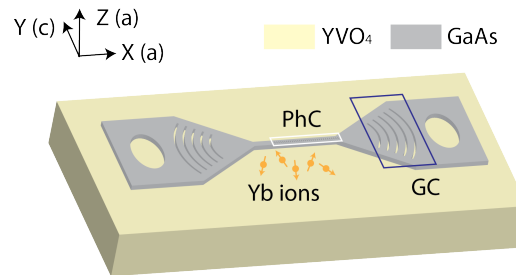


Figure A.1: The device schematic of hybrid device platform for coupling to $^{171}\text{Yb}:\text{YVO}_4$. The GaAs photonic crystal cavity and YVO_4 crystal are shown in gray and yellow, respectively. Yb ions are shown with orange arrows and are located inside YVO_4 . The crystal axes are shown next to the coordinate axes. The grating coupler (blue rectangle) is used for coupling light to a free-space setup. The photonic crystal cavity is indicated with a white rectangle.

Achieving significant enhancement in a hybrid photonic platform requires using a material with several important features. It needs to be transparent at the target wavelength, the refractive index needs to be significantly higher than the target crystal and high-quality thin-film material should be readily available. The optical transition of $^{171}\text{Yb}^{3+}:\text{YVO}_4$ at 984.5 nm precludes the use of silicon. Hence, we choose GaAs as the photonic layer due to its transparency in the near infrared, its large refractive index ($n_{\text{YVO}_4} = 2.17$, $n_{\text{GaAs}} = 3.52$) and the availability of wafer-scale thin films. High-quality factor photonic crystal cavities have been fabricated at this wavelength using ebeam lithography, thus enabling mass production [190, 191]. Recently, it has been demonstrated that the GaAs platform is amenable for scaling up deterministic solid-state photon-emitter interfaces [192]. In addition, wafer bonding techniques enable the creation of large scale photonic integrated circuits [193]. In the following sections, we will present the design and fabrication of these hybrid devices, and experimentally demonstrate optical control of single ^{171}Yb ions.

A.1 Device Design and Fabrication

Yb ions inside an a-cut YVO_4 crystal are coupled to a 1-dimensional photonic crystal cavity with fundamental TE mode [111], where the electric field is mostly aligned with the c-axis of the crystal. The photonic crystal design is based on unit cells with elliptical holes to engineer a bandgap at 984.5 nm. A localized cavity mode is formed by quadratically tapering 20 central periods of a 44-period photonic crystal (Figure A.2a,b). This is optimized to reduce radiation loss while maintaining a small mode volume. We engineer preferential cavity–waveguide coupling on one side by removing 11 mirror periods. The design of GaAs photonic crystal is chosen to enable high field penetration into YVO_4 without decreasing the quality factor significantly. We chose an effective index corresponding to the geometric mean of YVO and GaAs. Depending on the loss mechanisms a systematic study might yield better performance. Figures A.2b,c show the simulated y-directed electric field (E_y) that will be aligned with the Yb optical dipole moment along the c axis, along with the pertinent cavity dimensions. The simulated squared electric field magnitude ($|E|^2$) at the YVO_4 –GaAs interface is 40% of the maximum $|E|^2$ inside the GaAs layer. The effective mode volume is $\sim 1.7 (\lambda/n_{\text{YVO}_4})^3$ (normalized according to the strongest electric field in YVO_4 right at the surface) and the simulated radiatively-limited quality factor is over 1×10^6 . $|E_y|^2$ evanescently decays with distance from the interface, and the 1/e depth is 42 nm. To couple light from the cavity into a fiber, we utilize a fully-etched grating coupler [194]. The grating coupler efficiency

is optimized by modifying the grating dimensions leading to a simulated efficiency of $\sim 25\%$.

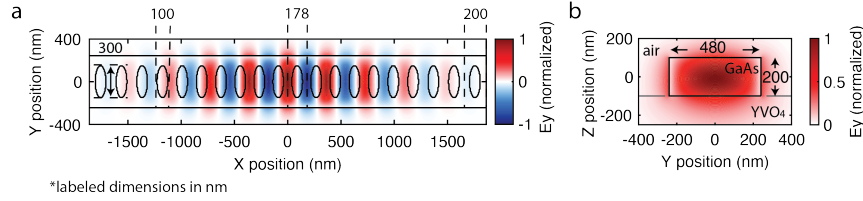


Figure A.2: Hybrid device simulations. (a), (b) Simulated E_y field of the fundamental TE mode in the XY and YZ planes, respectively. Note the evanescent decay of electric field inside YVO_4 .

Devices are fabricated on an epitaxial GaAs wafer using a standard electron-beam lithography procedure with an acceleration voltage of 100kV (Raith EPBG 5000) [190]. A 500-nm positive-tone resist (ZEP) is patterned and subsequently developed using n-Amyl acetate (ZED, Zeon corp.). The resist serves as the etch mask for an optimized inductively-coupled plasma reactive-ion etcher with Cl_2 -Ar chemistry. Devices are undercut by etching an AlGaAs sacrificial layer in HF and cleaned subsequently using H_2O_2 and KOH to remove any residues (Figure A.4a).

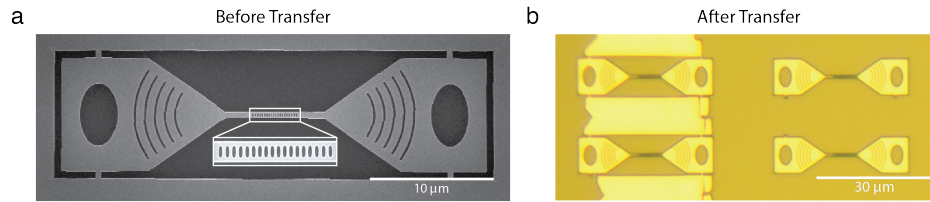


Figure A.3: Images of fabricated GaAs devices. (a) Scanning electron microscope image of the suspended GaAs photonic crystal cavity before transferring onto YVO_4 . (b) Optical image of the transferred device. The darker yellow substrate under the GaAs photonic crystal cavities is YVO_4 .

Finally, suspended GaAs photonic crystal cavities are transferred onto the YVO_4 surface using a stamping technique [95]. Devices are aligned with the a-axis and are perpendicular to the c-axis. We use a polydimethylsiloxane stamp covered by a thin film of polycarbonate to pick up devices and subsequently release them onto YVO_4 using a transfer stage. The success rate of this procedure is over 90%. The YVO_4 crystal used in this experiment was polished from an undoped boule (Gamdan Optics) with an Yb concentration of 0.14 ppm. The optical image after the transfer

is shown in Figure A.4b. The device used in these measurements has a quality factor of 5300 after transferring onto YVO_4 (Figure A.4). Typical quality factors of the devices before and after transfer are 15000 and 5000, respectively, and are likely limited by the imperfect device fabrication and surface absorption [191].

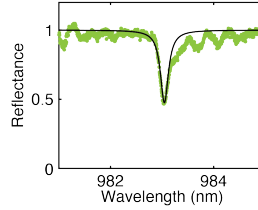


Figure A.4: Cavity reflection spectrum after transferring onto YVO_4 . Data are shown in green dots and overlaid with a fitted black line, which gives a quality factor of 5300.

A.2 Detection of Single Yb Ions in YVO_4

Experiments are performed in a Bluefors ^3He fridge (LD-He250) at 0.5 K and zero magnetic field. The experimental setup is depicted in Figure A.5a, where acousto-optic modulated laser pulses are sent through a 99:1 beamsplitter and focused onto the grating coupler through an aspheric lens doublet. The ion emission is sent back through the same fiber, and 99% of the light is directed to a superconducting nanowire single photon detector (SNSPD). The light coupling is optimized using an x-y-z nanopositioner (Attocube), and the device resonance is tuned to the ion emission frequency through nitrogen condensation.

Figure A.5b shows a resonant pulsed photoluminescence spectrum, where ion emission has been distinguished from the excitation in the time domain. The zero frequency offset corresponds to emission from Yb isotopes with no nuclear spin. At zero magnetic field, these isotopes contain degenerate Kramers doublets in both the ground and excited states [106]. The arrows indicate the ions used in this work.

To demonstrate single ion addressability, a single detector pulsed $g^2(t)$ measurement was performed on the ion indicated by the blue arrow (Figure A.5c). The resulting $g^2(0)$ is 0.26 ± 0.09 , lower than the two-ion limit of 0.5. The $g^2(t)$ measurement demonstrates a weak bunching feature, likely caused by spectral diffusion of the optical transition. By modeling spectral diffusion via rate equations, we derived and

fit the following function to the $g^2(t)$ data:

$$g^{(2)}(t) = [1 + (\tau_w/\tau - 1)\exp(-|t|/\tau)] \times [1 - \exp(-(r + 1/\tau_{\text{ion}})|t|)], \quad (\text{A.1})$$

where τ is the spectral diffusion rate, τ_w is the rate of ion diffusing out of the detection window, r is the pumping rate, and τ_{ion} is the lifetime of the emitter [195]. We can derive a spectral diffusion correlation time-scale of about 1 ms. The ion indicated by the red arrow has the shortest lifetime in this sample, measured to be $4.2 \pm 0.1 \mu\text{s}$ (Figure A.5d). This corresponds to a lifetime reduction of 64 times, which is consistent with the maximum lifetime reduction ($\beta F_p = 83$) predicted by the Purcell effect [114]. $\beta = 0.35$ is the branching ratio of the transition [91] and $F_{p,\text{max}} = (3/4\pi^2)(\lambda/n)^3(Q/V) = 237$. From the predicted maximum Purcell factor, measured lifetime, and the decay profile of the electric field strength given in Section A.1, we can derive maximum depths of the ions shown in Figure A.5d, which are 12 nm and 101 nm for the red and orange ion, respectively.

A.3 Coherent Optical Control of a Single ^{171}Yb Ion

While Yb isotopes without nuclear spin show bright emission without initialization, they suffer from poor coherence times due to first order susceptibility of transition frequencies to magnetic fields. Hereafter, we focus on ^{171}Yb ions which have an additional 1/2 nuclear spin giving a zero field hyperfine energy level structure shown in Figure A.6a. These ions exhibit optical and spin clock transitions between the $|0\rangle_g$, $|1\rangle_g$, $|0\rangle_e$, and $|1\rangle_e$ levels, which are first order insensitive to magnetic field noise yielding enhanced coherence properties [91]. The A-transition has its dipole moment along the c-axis of the crystal; it is copolarized with, and enhanced by, the cavity. Furthermore, it has no overlap with other Yb isotopes, making it suitable for optical readout. The C-transition has dipole moment perpendicular to the c-axis of the crystal and is used for optical pumping and initialization.

To verify that the ion indicated by the orange arrow is ^{171}Yb (Figure A.5b), we performed a pump-probe measurement, where we varied the pump laser frequency and measured the photoluminescence on the A-transition. When the pump laser is resonant with the C-transition, the ion will be partially initialized into $|1\rangle_g$ and have brighter emission (Figure A.6b). This initialization is performed prior to all subsequent measurements.

Resonant photoluminescence with varied excitation pulse length on the A-transition shows optical Rabi oscillation (Figure A.6c). In this measurement, initialization is

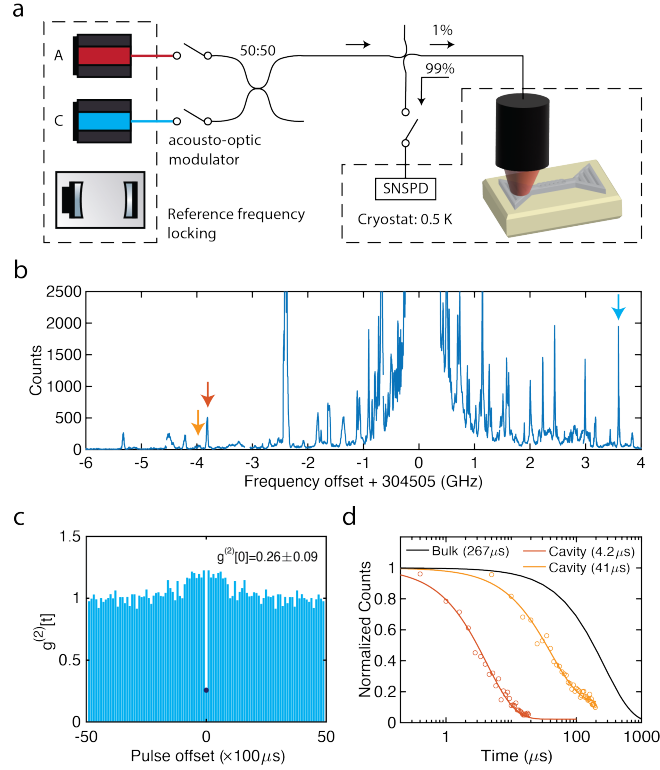


Figure A.5: Experimental setup and detection of single Yb ions. (a) Experimental setup. Optical pulses from two frequency locked lasers, labeled A and C according to the driven optical transition (Figure A.5a), are sent to the device located in a ^3He fridge at 0.5 K. Emitted photons are routed to an SNSPD in the same cryostat. Laser pulses are shaped using two double-pass acousto-optic modulators, and optical pulses are removed from the detection through time filtering and an acousto-optic modulator shutter setup. (b) Resonant photoluminescence spectrum. The large emission peak at 304505 GHz corresponds to Yb isotopes with no nuclear spin. Isolated peaks inside the spectrum are mostly single Yb ions, the arrows indicate ions measured in subsequent experiments. Data was taken with 10 second integration time and 50 kHz repetition rate for each point. The fitted linewidth of red and blue ions are 27.3 MHz and 15.9 MHz, respectively. (c) Pulsed $g^{(2)}(t)$ measurement of the ion indicated with a blue arrow shows $g^{(2)}(0) = 0.26 \pm 0.09$. (d) Lifetime measured through time-resolved photoluminescence of ions indicated by red ($4.2 \pm 0.1 \mu s$) and orange ($32 \pm 2 \mu s$) arrows are shown with corresponding colors. Experimental data are shown with dots and overlaid with fitted exponential decays. The fit to the orange curve also includes an additional contribution from background bulk ions due to its lower intensity. Bulk lifetime is shown with a black line.

periodically applied after each sequence of 50 readout pulses. Combined with the limited cyclicity of the readout transition, this acts to saturate the photon counts and create a flat top feature. This ion has a lifetime of $32 \pm 2 \mu s$, leading to an A-

transition cyclicity of 13 [91]. Characterization of the optical coherence properties is first performed with an optical Ramsey measurement, which includes two optical $\pi/2$ pulses with varied separation. We measured $T_2^* = 69 \pm 15$ ns corresponding to an effective linewidth of 4.6 MHz. The short-timescale optical frequency stability is measured using an echo consisting of an additional intermediate optical π pulse to rephase the coherence, yielding $T_2 = 330 \pm 50$ ns.

In these experiments, we were unable to perform optical coherence measurements for some of the ions that have shorter lifetimes. Ions in these hybrid devices with larger Purcell factors are closer to the surface and therefore more susceptible to surface defects. We postulate that the resulting charge noise could lead to excess spectral diffusion and deteriorated coherence properties. By contrast, in previously studied monolithic devices [91], well-coupled ions showed 1 μ s post-selected optical Ramsey and lifetime-limited optical echo (4 μ s) since they were centrally located and farther from surfaces. The next step would be to characterize ion statistics with different Purcell factors and understand the limiting factor for the optical coherence in these hybrid devices.

A.4 Conclusion and Outlook

In this work, we fabricate suspended GaAs photonic crystal cavities and transfer them onto YVO₄ with a $\sim 90\%$ success rate. We use a cavity with a quality factor of 5300 to address single ^{171}Yb ions, show a lifetime reduction of 64 times, and measure their optical coherence properties.

In the future, developing coherent microwave control of the spin transition will be essential. To improve the platform and increase count rates, refining fabrication of the photonic crystal cavity through surface passivation and further optimization of the lithographic procedure are crucial [191, 196]. Switching to a shallow-etched grating coupler can also increase the waveguide to free-space coupling efficiency [197], and the overall photon collection rate into the waveguide can be improved by optimizing the cavity-waveguide coupling [198]. If the optical coherence is limited by surface proximity, we could dope Yb ions at different depths and characterize their properties. Optimizing the choice of the host crystal can also significantly improve the optical coherence [79, 86]. Additional improvements could be attained by removing polishing-induced strain through plasma etching [199] and controlling the surface chemistry through systematic study of surface cleaning and preparation techniques [200]. Given the fabrication capability and the flexibility of the hybrid

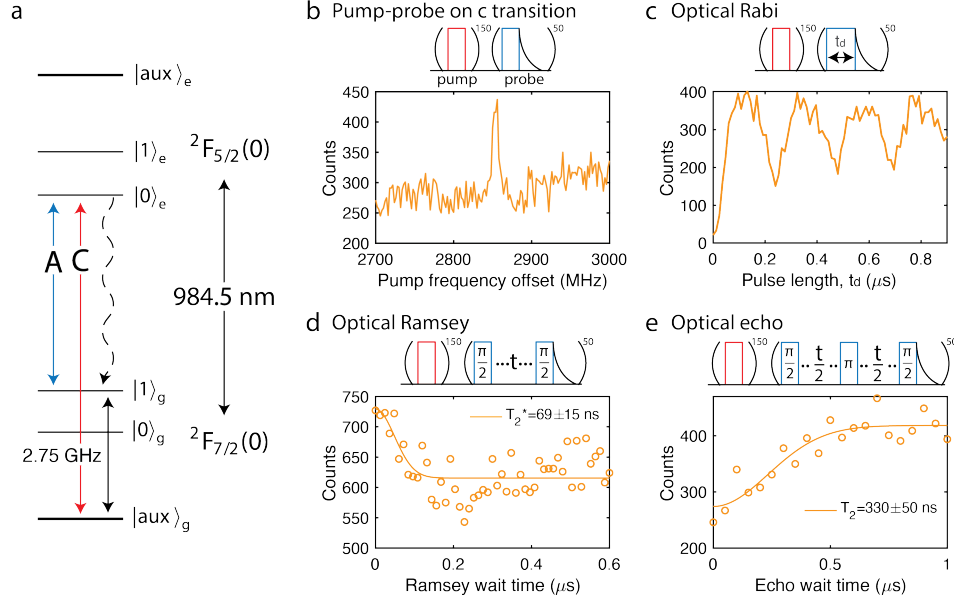


Figure A.6: Coherent optical control of a single ^{171}Yb ion. (a) Energy level structure of ^{171}Yb inside YVO_4 at zero magnetic field. Transition A (cavity enhanced) is shown in blue; transition C (non-enhanced) is shown in red. Cavity enhanced decay is shown with a dashed arrow. $|0\rangle_g$, $|1\rangle_g$, $|0\rangle_e$, $|1\rangle_e$ are magnetic field insensitive forming clock transitions. (b) Pump-probe measurement of the ^{171}Yb optical transitions. The pump frequency is varied around the C-transition with probe frequency fixed to the A-transition. In subsequent measurements, the ion is partially initialized by repeatedly pumping on this transition. The difference between the frequency noted in Figure A.6a and the measurement could be due to strain at the site of the ion. (c) Optical Rabi oscillation on the A-transition. (d) Optical Ramsey measurement on the A-transition gives $T_2^* = 69 \pm 15$ ns. (e) Optical echo measurement on the A-transition gives $T_2 = 330 \pm 50$ ns.

GaAs platform combined with the excellent properties of $^{171}\text{Yb}:\text{YVO}_4$, we think this approach can be a promising route to build a quantum network.