Hybrid quantum systems offer the opportunity to combine the benefits of different qubit types while avoiding some of their pitfalls. Task-dependent qubit selection allows the usage of long-lived qubits for memory and qubits with rapid gate speeds for operations. For optical systems, a photon bus can be used to remotely link these systems via photon-heralded entanglement. To successfully generate entanglement, the two different qubit systems must emit identical photons, requiring spectro-temporal engineering of at least one qubit’s photon wavepacket. While significant progress has been made toward efficient quantum-frequency conversion\textsuperscript{[10–12]} post-emission temporal photon pulse-shaping\textsuperscript{[13–14]} techniques for the narrow-band photons from both trapped ions and solid-state defects is an outstanding challenge.

We have identified two disparate, complementary qubit systems in which high-fidelity photon-mediated entanglement should be possible by direct control over the photon emission process. Trapped ions are a well-studied qubit system with high operational fidelities and long coherence times\textsuperscript{[15–17]} but relatively slow initialization and gate speeds\textsuperscript{[18]}. Electron spins in semiconductors have rapid initialization and gate speeds\textsuperscript{[19–21]}, but have shorter coherence times. A hybrid system consisting of ions and electrons bound to donor defects would have the ability to use ions for quantum memory and defects for gate operations, producing a system more rapid and reliable than either qubit alone.

Yb\textsuperscript{+} and the ZnO donor were chosen as the target systems for their shared transition near 369 nm: the $^2P_{1/2}$ to $^2S_{1/2}$ transition in $^{171}$Yb\textsuperscript{+} and the In neutral donor bound exciton (D\textsuperscript{0}X) to neutral donor (D\textsuperscript{0}) transition in ZnO (Fig. 1). In:ZnO is analogous in structure to the better-known P:Si qubit system\textsuperscript{[22]} however ZnO is a direct band gap semiconductor enabling efficient donor coupling to photons. While the two transition frequencies are quite close ($\delta = 340 \text{ GHz}$), the excited state lifetimes differ by a factor of 6 resulting in a large temporal mismatch. Prior semiconductor spin - trapped ion entanglement schemes addressed similar temporal mismatch by using coherent scattering\textsuperscript{[13]} or sacrificing fidelity\textsuperscript{[15]}. Here we demonstrate that pulse shaping can be a powerful tool to attain high-fidelity entanglement and show that an entanglement rate of 21 kHz and fidelity of 94% is feasible.

A heralded entanglement scheme based on weak excitation, single-photon detection and which-path erasure can be used to entangle the two systems, similar to the proposal by Cabrillo et al\textsuperscript{[23]} Fig. 1 depicts the relevant energy levels and excitation/decay pathways for the donor and ion. Here D\textsuperscript{0} system is in the Voigt (B $\perp \hat{k}$) geometry but the Faraday geometry could also be utilized. The donor is coupled to an optical cavity detuned by $\Delta$ from the D\textsuperscript{0}X-D\textsuperscript{0} transition.

The diagram of the experiment is shown in Fig. 2. The Yb\textsuperscript{+} and In donor are first initialized using optical pumping...
FIG. 2. Trapped ion system (left) and ZnO system (right). A transfer cavity phase-locks the two 369 nm excitation lasers. The two acousto-optic modulators (AOM) are synchronized and programmed to output the calculated pulse shapes for their respective qubits. Photons collected from the two qubits interfere on the beam splitter (BS) via inputs A and B. Successful entanglement is heralded by the detection of a single photon by photodetectors (PD) at outputs C, D.

to |F = 0, m_F = 0⟩ and |m_z = −1/2⟩, respectively, producing the initial state |Ψ_i⟩ = |0⟩_{Yb} ⊗ |0⟩_{In} ⊗ |vac⟩ = |0; 0; vac⟩. Next, each system is excited to |e⟩_{In} or |e⟩_{Yb}, using resonant or near-resonant pulsed excitation. Here, we assume the weak excitation limit (excitation probability p_1,x < 10%, x = {Yb, In}). The state of the ZnO donor and ion is now given by

$$|Ψ⟩_{i} = β_1 |0; 0; vac⟩ + β_2 |1; 1; ζ_{Yb}, ζ_{In}⟩ + β_3 |0; 1; ζ_{In}⟩ + β_4 |1; 0; ζ_{Yb}⟩,$$

where the emitted photons on paths A and B of Fig. 2 [2, ζ_{Yb}] = \sum_ω \xi_{Yb,ω} a_ω^† |vac⟩ and |ζ_{In}⟩ = \sum_ω \xi_{In,ω} b_ω^† |vac⟩ are given by a sum over all modes ω (ω’) with coefficients ξ_{Yb,ω} (ξ_{In,ω’) and creation operators a_ω^† (b_ω^†). The coefficients β emerge from the excitation (p_1,x) probabilities of the two systems, the phase gained from excitation laser phases (ϕ_{x,L}), and the distance travelled by the collected photon (ϕ_{x,d}):

$$β_1 = \sqrt{(1 - p_{1,Yb})(1 - p_{1,In})} e^{i(ϕ_{Yb,L} + ϕ_{In,L})}$$
$$β_2 = \sqrt{p_{1,In}(1 - p_{1,Yb})} e^{i(ϕ_{Yb,L} + ϕ_{In,d})}$$
$$β_3 = \sqrt{p_{1,Yb}(1 - p_{1,In})} e^{i(ϕ_{Yb,d} + ϕ_{In,L})}$$
$$β_4 = \sqrt{p_{1,In}p_{1,Yb}} e^{i(ϕ_{Yb,d} + ϕ_{In,d})}$$

By phase locking the laser pulses, we can ignore ϕ_{x,L}.

Collected photons from both systems interfere on the beamsplitter, which erases which-path information. Entanglement is heralded by the detection of a single photon at one of the two photodetectors. With the appropriate choice for p_{1,Yb}, p_{1,In}, and the collection efficiency from each system (supplemental material), photon detection in path D projects the ion-donor qubits onto the renormalized entangled state

$$|Ψ⟩ = \frac{1}{\sqrt{2}} \left( |0; 1; ζ_{In}⟩ - ie^{iΔφ} |1; 0; ζ_{Yb}⟩ \right),$$

where Δφ is determined by the optical path length difference. Similar expression can be derived for detector C. Tracing over all photon modes, we get the reduced Yb+ − In density matrix

$$ρ^{Yb,In}_{Yb,In} = \frac{1}{2} |0; 1⟩⟨0; 1| + \frac{1}{2} |1; 0⟩⟨1; 0| +$$
$$\frac{1}{4} \left( ie^{iΔφ} ⟨ζ_{Yb} | ζ_{In}⟩ |0; 1⟩⟨1; 0| + c.c. \right),$$

where ⟨ζ_{Yb} | ζ_{In}⟩ = \sum_ω ξ_{Yb,ω}^* ξ_{In,ω}° is the overlap of the photons from the Yb+ and ZnO systems.

Factors which affect the entanglement fidelity are photon overlap, false identification of both-system excitation as a single-system excitation, and atomic recoil from the ion interacting with the excitation laser. Accounting for these sources of error, the final fidelity is:

$$F = \frac{1}{2 + c_1} [1 + F_{dyn} \Re(⟨ζ_{Yb} | ζ_{In}⟩)]$$

where c_1 depends on the excitation probabilities and detection efficiencies of both systems (supplemental material) and F_{dyn} is related to the photon recoil effect. Motion of the trapped ion due to photon recoil during the absorption/emission process can shift the frequency of the photon and reduce fidelity of the entangled state. Note that for the ZnO donor, absorption/emission are recoilless due to the Mössbauer effect. For a Doppler-cooled ^{171}Yb+ in a 1 MHz trap in geometry where the ion is excited by a laser pulse parallel to the light collection direction, the expected F_{dyn} is 96%. Additional factors that may further decrease the fidelity include photodetector dark counts, background luminescence from ZnO, D’X spectral diffusion, and uncertainty in the phase stabilization.

Photon collection efficiency primarily affects the protocol’s probability of success. For trapped ions, light collection is challenging due to the high-vacuum environment and the need to isolate ions from decoherence-inducing surfaces. Typical light collection efficiency is 2-4% utilizing off-the-shelf long working distance microscope objectives, while optics based on in-vacuum lenses and custom high-NA objectives are capable of collecting up to 10% of the emitted photons. Further enhancement is possible by integrating a metallic parabolic mirror as an RF electrode of the ion trap. Ions are trapped at the focus of the mirror, so that the emitted photons are collimated upon reflection from the mirror with an expected 32% overall coupling efficiency into a single-mode optical fiber. As we show below, the parabolic mirror trap also provides a novel mechanism for polarization filtering. Longer term, integrated-photonics platforms may provide a path toward high-NA collection from scalable arrays of ions.
donor-cavity coupling strength, $\kappa$ is the cavity decay rate and $\Gamma_{\text{In}}$ is the spontaneous decay rate in the “bad cavity” limit necessary for the pulse-shaping procedure described below, lie in a band of readily achievable $Q/V$ ratios with today’s nanophotonic fabrication techniques. Due to intrinsic band-edge absorption, the high quality factor region in Fig. 3 may not be achievable at $D^0X-D^0$ transitions, thus low mode volume cavities with moderate quality factors should be targeted. While nanophotonic fabrication in ZnO is relatively immature compared to other quantum defect host crystals, small mode volume ZnO nanowire cavities have enabled UV lasers and ZnO cavities fabricated by focused ion beam milling, a method that has been used to achieve high cooperativity in rare-earth doped systems, exhibit quality factors up to 1000. In the limit that the cavity photon loss rate $\kappa$ is dominated by coupling to the output mode, over 50% collection efficiency into a waveguide for planar geometry cavities, or into an objective lens for nanowire cavities is possible.

As shown in Eq. 5 for high fidelity entanglement, the frequency, polarization, and temporal shape of the photons emitted by the two systems must be matched to maximize $\mathcal{R}(\langle \xi_{\text{Yb}} | \xi_{\text{In}} \rangle)$. The type of donor used affects the amount of frequency shift required to match the emission frequency of Yb$^+$. Of the three primary donor candidates, Al, Ga and In, the In $D^0X$ transition is closest to the Yb$^+$ transition, $v_{\text{In}} = 1 1.5 + 0.34$ THz. The donor will be integrated in an optical cavity detuned from the relevant transition by ~200 GHz. The remaining frequency shift will be attained via the dc Stark effect. Electric field tuning in a similar quantum dot trion system has shown that several meV of tuning is possible.

Decay from $|e\rangle_{\text{Yb}} (^2P_{1/2} | F = 1, m_F = 0 \rangle)$ can occur along three different channels, producing either a $\sigma^\pm$ Raman photon or a $\pi$ Rayleigh photon (see Fig. 4). A pure polarization state is required for polarization matching with the photon emitted by the ZnO donor. While the use of a high-NA collection optic increases the photon collection efficiency, it can pose problems for polarization purity. However, the parabolic mirror can be utilized to filter out the undesired $\pi$ polarized photons when the optical axis is oriented along the quantization axis defined by the applied magnetic field. In this geometry, the $\pi$-polarized photons reflected off the mirror have a radial polarization pattern, which completely destructively interferes when focused into a single-mode optical fiber. The $\sigma$-polarized photons, on the other hand, have an elliptical polarization upon reflection from the mirror. The eccentricity increases with radial distance from the center, with perfectly circular polarization at the center of the reflected beam and linear polarization at the edge. The linear component is filtered out by destructive interference in the optical fiber.

In the Voigt geometry, with the applied magnetic field perpendicular to the crystal axis, the branching ratio between the ZnO donor Raman transitions $|e\rangle_{\text{In}} \rightarrow |0\rangle_{\text{In}}$ and $|e\rangle_{\text{In}} \rightarrow |1\rangle_{\text{In}}$ is approximately $1:4:1:4$ for a cavity with large $V$ and high $Q$ (e.g. ring resonator), the cavity resonance will be narrower than the Zeeman splitting of D$^0$, allowing for selective coupling of the desired Raman transition. For high $V$, the size of the cavity is large compared to the excitation beam diameter, so polarization selection can be attained by selectively exciting a small area of the cavity, where only one dipole moment is coupled to the cavity mode. For cavities with low $Q$ and $V$, polarization and frequency selection can be achieved via cross polarization and spectral filtering.

Matching the temporal profiles of the emitted photons poses a greater challenge. The $^2P_{1/2}$ Yb$^+$ state lifetime is 8.1 ns, while that of $D^0X$ state in ZnO is only 1.4 ns. Post-emission pulse shaping is not feasible because the ZnO and Yb photons are too narrow band for these dispersive methods. Instead, the photons emitted by the ZnO donor can be pulse-shaped at their creation by modulating the intensity of the excitation pulse. The ZnO cavity is constructed with parameters within the “bad cavity” regime ($\kappa \gg g^2/\kappa \gg \Gamma_{\text{In}}$). The large cavity decay rate ensures that we are not in the strong coupling regime, so the donor excitation follows the optical pulse, while the high cooperativity ensures that the donor decays via Raman emission into the cavity.

While it is possible to obtain an analytic expression for the ideal excitation pulse shape for maximum photon overlap, in this work we limit ourselves to experimentally attractive Gaussian pulses and performed numerical simulations to determine photon temporal overlap, given the practical cavity
considerations discussed above. The donor defect is modeled as a three level system with initial state $|0\rangle_{In}$ (Fig. 4) connected to the excited state $|e\rangle_{In}$ by an excitation pulse of Rabi frequency $\Omega_{In}(t)$ and detuning $\Delta$. We neglect the effect of the other excited state level. The cavity is coupled to the $|e;0\rangle \leftrightarrow |1;1\rangle$ transition with detuning $\Delta$ and coupling strength $g$. Photons from this transition have a spontaneous radiative decay rate of $\Gamma_{In}$. Photons escape the cavity at the cavity decay rate $\kappa$. The equations of motion for the population amplitudes are [10,12]

$$\frac{d}{dt}a_{In}(t) = \frac{1}{2} \left( \begin{array}{cc} \Omega_{In}(t) & 0 \\ 0 & 0 \end{array} \right) a_{In}(t),$$ (6)

where $a_{In}(t) = [a_{0,In}(t), a_{e,In}(t), a_{1,In}(t)]^T$.

The Yb$^+$ is modeled in a similar manner but without a cavity. The ground state $|0\rangle_{Yb}$ is coupled to the excited state $|e\rangle_{Yb}$ by the Rabi pulse $\Omega_{Yb}(t)$. Decay of the excited state occurs with the cavity $\Gamma_{Yb}$. The equations of motion are:

$$i \frac{d}{dt} \begin{pmatrix} a_{0,Yb}(t) \\ a_{e,Yb}(t) \end{pmatrix} = \frac{1}{2} \begin{pmatrix} \Omega_{Yb}(t) & 0 \\ 0 & -i\Gamma_{Yb} \end{pmatrix} \begin{pmatrix} a_{0,Yb}(t) \\ a_{e,Yb}(t) \end{pmatrix}$$ (7)

The emission rates of the photons from the ZnO and Yb$^+$ systems are $\kappa|a_{1,In}(t)|^2$ and $\Gamma_{Yb}|a_{e,Yb}(t)|^2$, respectively [11] with temporal wavefunctions given by normalizing the population amplitudes $a_{1,In}(t) \rightarrow A_{1,In}(t)$ and $a_{e,Yb}(t) \rightarrow A_{e,Yb}(t)$. By controlling the Rabi frequencies $\Omega_{In}(t)$ and $\Omega_{Yb}(t)$, it is possible to engineer the real component of the photon overlap $\int_{0}^{\infty} A_{e,Yb}(t) A_{1,In}(t) dt = \langle \xi_{Yb} | \xi_{In} \rangle$ to ~0.99 for practical experimental parameters using the control pulses shown in Fig. 5. The optimized pulse is restricted to a Gaussian pulse shape with adjustable rise time $\sigma_1$, fall time $\sigma_2$, time to pulse max $\tau$, hold time $t_h$, maximum pulse height $\Omega_{max}$, and phase factor $e^{i(\sigma_1 + \theta)}$. Setting either pulse to achieve a desired excitation probability $p_{1,\tau}$, we iteratively sweep the pulse parameters for the other system to obtain local maxima in the overlap.

The probability of successful entanglement is

$$P_{\text{suc}} = [p_{1,\text{Yb}} p_{2,\text{Yb}} (1 - p_{1,\text{In}}) + p_{1,\text{In}} p_{2,\text{In}} (1 - p_{1,\text{Yb}})] \eta$$ (8)

where $\eta$ is the quantum efficiency of the detector, which can be as high as ~80% using superconducting nanowire single photon detectors (SNSPD’s) [11] for photons at 369 nm. With a parabolic mirror ion trap, collection efficiency for Yb$^+$ systems is 32%; the ZnO system is set to 34% collection efficiency to match the coefficients in Eq. 2 to create a maximally entangled state. Excitation probabilities depend on the pulse shaping requirements, and need to be kept low (<10%) to minimize error. For good fidelity while still maintaining a reasonably high success probability, we use excitation probabilities around 5%.

Each experimental run begins with ~1 $\mu$s of optical pumping, followed by the ~10 ns excitation pulse. If a single photon is detected, then the state readout is performed, taking ~10 $\mu$s and limited by the ioSP [12]. We find a success probability of ~2.7%, leading to an entanglement generation rate of 21 kHz. Practically, this rate will be further decreased by the interferometer phase stabilization and defect frequency stabilization steps [12].

With all experiments using this type of protocol, there is a tradeoff between success probability and fidelity [17,18]. One can always increase the success probability by increasing the excitation probability, but this degrades the fidelity according to Eq [5]. Further, in order to be useful, the entanglement rate needs to be comparable to the rate of decoherence. While the demonstrated coherence time for trapped ytterbium ions is 10$^3$ [23] (10 minutes), the spin echo time $T_2$ of ensemble donor bound excitons in ZnO is only 50 $\mu$s. However, the fundamental limit of $T_2$ is the longitudinal spin relaxation time $T_1$ which exceeds 100 ms [33] and may allow for improvement through chemical and isotope purification [25].

In summary, a ZnO donor defect qubit and a single trapped Yb$^+$ ion can be remotely entangled via a photonic link at 369 nm. Pulse shaping techniques can be used to alter the temporal profile of the photon emitted by the donor to attain the temporal wavefunction overlap of 0.99 with the photon emitted by the trapped ion, leading to an entangled state fidelity of 94% with realistic parameters.

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1. J. D. Siverns, J. Hannegan, and Q. Quraishi, “Neutral-atom wavelength-
B. Blinov, D. Moehring, L.-M. Duan, and C. Monroe, “Observation of atom-atom entanglement by single-photon detection,” Physical Review Letters 114, 091101 (2019).

M. H. Huang, S. Mao, H. Feick, H. Yan, Y. Wu, H. Kind, E. Weber, R. Rauso, and P. Y. Yu, “Room-temperature ultraviolet nanowire nanolasers,” Science 292, 1897–1899 (2001).

E.-C. Chang, K.-B. Hong, Y.-Y. Lai, Y.-H. Chou, S.-C. Wang, and T.-C. Lu, “Zeno-based microwaves sculpted by focus ion beam milling,” Nanoscale Research Letters 11, 319 (2016).

T. Zhong, J. M. Kindem, J. G. Bartholomew, J. Rochman, I. Craciun, V. Verma, S. W. Nam, F. Marsili, M. D. Shaw, A. D. Beyer, and A. Faraon, “Optically addressing single rare-earth ions in a nanophotonic cavity,” Physical Review Letters 121, 183603 (2018).

M. Arcari, I. Söllner, A. Javadì, S. Lindskov Hansen, S. Mahmoodian, J. Liu, H. Thyrrestrup, E. H. Lee, J. D. Song, S. Stobbe, and P. Lodahl, “Near-unity coupling efficiency of a quantum emitter to a photonic crystal waveguide,” Physical Review Letters 113, 093603 (2014).

P. Senellart, G. Solomon, and A. White, “High-performance semiconductor quantum-dot single-photon sources,” Nature Nanotechnology 12, 1026–1039 (2017).

B. Meyer, H. Alves, I. D. Hofmann, W. Kriegseis, D. Forster, F. Bertram, J. Christen, A. Hoffmann, M. Straubl, M. Dworzak, et al., “Bound exciton and donor–acceptor pair recombinations in ZaO,” physical status solidi (b) 241, 231–260 (2004).

A. J. Bennett, R. B. Patel, J. Skiba-Szymanska, C. A. Nicoll, I. Ferrar, D. A. Ritchie, and A. J. Shields, “Giant stark effect in the emission of single semiconductor quantum dots,” Applied Physics Letters 97, 031104 (2010).

T. Kim, P. Maunz, and J. Kim, “Efficient collection of single photons emitted from a trapped ion into a single-mode fiber for scalable quantum-information processing,” Physical Review A 84, 063423 (2011).

X. Linpeng, M. L. Viitaniemi, A. Vishnuradhan, Y. Kozuka, C. Johnson, M. Kawasaki, and K.-M. C. Fu, “Coherence properties of shallow donor qubits in zn o,” Physical Review Applied 10, 064061 (2018).

M. R. Wagner, J.-H. Schulze, R. Kirsie, M. Cobet, A. Hoffmann, C. Rauch, A. V. Rodina, B. K. Meyer, U. Röder, and K. Thonke, “η 7 valence band symmetry related hole fine splitting of bound excitons in zn observed in magneto-optical studies,” Physical Review B 80, 205203 (2009).

X. Liu, A. W. Bruch, Z. Gong, J. Lu, J. B. Surya, L. Zhang, J. Wang, J. Yan, and H. X. Tang, “Ultra-high-q UV microring resonators based on a single-crystalline aln platform,” Optica 5, 1279–1282 (2018).

D. Yum, and K. Kim, “Single-qubit quantum memory exceeding ten-minute coherence time,” Nature Photonics 11, 646–650 (2017).

M. Kawasaki, and K.-M. C. Fu, “Coherence properties of shallow donor qubits in zn o,” Physical Review Applied 10, 064061 (2018).

M. Wagner, G. Callesen, J.-H. Schulze, R. Kirsie, M. Cobet, I. Ostapenko, S. Roth, C. Nestielt, M. Kaiser, et al., “Bound excitons in ZnO Structural defect complexes versus shallow impurity centers,” Physical Review B 84, 055133 (2011).

I. J. Wright, M. Karpiński, C. Söller, and B. J. Smith, “Spectral imaging and fluorescence collection with a parabolic mirror trap,” Correlation 80, 319 (2016).

S. W. Nam, and J. Kim, “High-speed low-crosstalk detection of a single-photon quantum dot resonance fluorescence in an integrated cavity-waveguide device,” Optica 7, 380–385 (2020).

S. Olsmschenk, D. Hayes, D. Matsukevich, P. Maunz, D. Moehring, K. Young, and C. Monroe, “Measurement of the lifetime of the 6p 12s level of Yb,” Physical Review A 80, 022502 (2009).

M. Wagner, G. Callesen, J.-H. Schulze, R. Kirsie, M. Cobet, I. Ostapenko, S. Roth, C. Nestielt, M. Kaiser, et al., “Bound excitons in ZnO Structural defect complexes versus shallow impurity centers,” Physical Review B 84, 053133 (2011).

S. W. Nam, and J. Kim, “High-speed low-crosstalk detection of a single-photon quantum dot resonance fluorescence in an integrated cavity-waveguide device,” Optica 7, 380–385 (2020).

S.-Y. Baek, O. Kwon, and Y.-H. Kim, “Temporal shaping of a heralded single-photon wave packet,” Physical Review A 77, 013829 (2008).

C. Law and H. Kimble, “Deterministic generation of a bit-stream of single-photon pulses,” Journal of Modern Optics 44, 2067–2074 (1997).

G. S. Vasilev, D. Ljunggren, and A. Kuhn, “Single photons made-to-measure,” New Journal of Physics 12, 063024 (2010).

S. Crain, C. Cahall, G. Vrijsen, E. E. Wollman, M. D. Shaw, V. B. Verma, S. W. Nam, and J. Kim, “High-speed low-crosstalk detection of a 171-Yb+ qubit using superconducting nanowire single photon detectors,” Communications Physics 2, 97 (2019).

Y. Wang, M. Um, J. Zhang, S. An, M. Lyu, J.-N. Zhang, L.-M. Duan, D. Yum, and K. Kim, “Single-qubit quantum memory exceeding ten-minute coherence time,” Nature Photonics 11, 646–650 (2017).

J. Tribollet, “Theory of the electron and nuclear spin coherence times of shallow donor spin qubits in isotopically and chemically purified zinc oxide,” The European Physical Journal B 72, 531 (2009).
I. SUPPLEMENTAL MATERIAL

A. Maximally entangled state and fidelity

We first define the state $|\Psi_1\rangle$ of Yb$^+$ and the state $|\Psi_2\rangle$ of the In donor. We begin by optically pumping both systems into the ground state

$$|\Psi_1\rangle = |0\rangle_{Yb}$$

$$|\Psi_2\rangle = |0\rangle_{In}.$$ (9a)

We now apply an excitation pulse to both species with $p_{1,x} \ll 1$ so that the probability of both systems being excited during the same experimental run is small.

The states of both systems are given by:

$$|\Psi_1\rangle = \sqrt{p_{1,Yb}} e^{i\phi_{1,Yb}} |1\rangle_{Yb} + \sqrt{1-p_{1,Yb}} e^{i\phi_{1,Yb}} |0\rangle \langle \text{vac} |$$

$$|\Psi_2\rangle = \sqrt{p_{1,In}} e^{i\phi_{1,In}} |1\rangle_{In} + \sqrt{1-p_{1,In}} e^{i\phi_{1,In}} |0\rangle \langle \text{vac} |$$

where $\phi_{1,x}$ denotes the phase of the laser at species $x$, $\phi_{2,x}$ denotes the phase of the emitted photon after traveling a distance $D_x$, $|\text{vac}\rangle$ is the vacuum state. $|\xi_{Yb}\rangle = \sum_{a_0} \xi_{Yb,a_0} a_0^\dagger |\text{vac}\rangle$ and $|\xi_{In}\rangle = \sum_{a_0} \xi_{In,a_0} b_0^\dagger |\text{vac}\rangle$ are the temporal wavefunctions of emitted photons from each system. The temporal wavefunctions are given by a sum over all modes $a_0$ with coefficients $\xi_{Yb,a_0}$ and raising operators $a_0^\dagger$ ($b_0^\dagger$). We phase lock the laser systems to set $\phi_{1,Yb} = \phi_{1,In} = 0$. Assuming we collect a single photon with efficiency $p_{2,x}$ from either system, we obtain the state (not normalized)

$$|\Psi_{1,2}\rangle = \sqrt{p_{1,Yb}(1-p_{1,In})} p_{2,Yb} |1,0\rangle \sum_{a_0} \xi_{Yb,a_0} a_0^\dagger |\text{vac}\rangle + \sqrt{p_{1,In}(1-p_{1,Yb})} p_{2,In} e^{i\phi_{1,In}} |0,1\rangle \sum_{a_0} \xi_{In,a_0} b_0^\dagger |\text{vac}\rangle$$

where $\Delta \phi = \phi_{2,Yb} - \phi_{2,In}$ is the difference in the phase of the emitted photon between the two systems.

Here, we note that to obtain a maximally entangled state we want to set

$$p_{1,Yb}(1-p_{1,In}) p_{2,Yb} = p_{1,In}(1-p_{1,Yb}) p_{2,In}.$$ (13)

Since we use $p_{1,x}$ to achieve good temporal overlap, and typically $p_{2,Yb} < p_{2,In}$, this is accomplished by lowering $p_{2,In}$. Now, at the beamsplitter we choose the transformation $a_0^\dagger \rightarrow (c_0^\dagger + i d_0^\dagger)/\sqrt{2}$, $b_0^\dagger \rightarrow (d_0^\dagger + i c_0^\dagger)/\sqrt{2}$, where $c_0^\dagger$, $d_0^\dagger$ are the raising operators of the respective paths A and B and $c_0, d_0$ are the raising operators in paths C and D, as depicted in Fig. 2 of main text.

We also have that $i(\phi_{2,Yb} - \phi_{2,In}) = i\Delta \phi$ where $\Delta \phi$ is the difference in phase between photons traversed from each system. To account for the reflection of one of the two paths in the beamsplitter, we set a phase difference of $\pi/2$ between $|1,0\rangle$ and $|0,1\rangle$ states.

We then obtain the entangled state upon detection of a single photon

$$|\Psi_{1,2}\rangle = \frac{1}{\sqrt{2}} \left[ |1,0\rangle \sum_a \xi_{Yb,a} c_0^\dagger + i d_0^\dagger |\text{vac}\rangle - i e^{i\Delta \phi} |0,1\rangle \sum_a \xi_{In,a} d_0^\dagger + i c_0^\dagger |\text{vac}\rangle \right]$$

(14)

The density matrix can then be computed. Let us first assume the photon was detected on path D, and not on path C. Tracing over photon states in the path D, and over all photon frequencies $\omega$, we obtain

$$\rho_{Yb,In} = \frac{1}{2} \left[ |1,0\rangle \langle 1,0| + |0,1\rangle \langle 0,1| \right] - i e^{i\Delta \phi} (\xi_{Yb} \langle 0,1| \langle 1,0|) (15)$$

The same matrix can be found for the path C. Summing the density matrices we then find the complete density matrix including paths C and D

$$\rho_{Yb,In} = \frac{1}{2} \left[ |1,0\rangle \langle 1,0| + |0,1\rangle \langle 0,1| \right] - i e^{i\Delta \phi} (\xi_{Yb} \langle 0,1| \langle 1,0|)$$

(16)

where we have used the relations $\langle \xi_{In} | \xi_{In} \rangle = \langle \xi_{Yb} | \xi_{Yb} \rangle = 1$ and $\langle \xi_{Yb} | \xi_{In} \rangle = \langle \xi_{In} | \xi_{Yb} \rangle = \sum_a \xi_{In,a} \xi_{Yb,a}$.

Finally, we compute the fidelity using the target state $|\Psi_{\text{ent}}\rangle = \frac{1}{\sqrt{2}} (|1,0\rangle - i e^{i\Delta \phi} |0,1\rangle)$

$$F = \langle \Psi_{\text{ent}} | \rho_{Yb,In} | \Psi_{\text{ent}} \rangle = \frac{1}{2} \left[ 1 + R e (\langle \xi_{Yb} | \xi_{In} \rangle) \right]$$

(17)

B. Double Excitations

Here we will derive the parameter $c_1$ of Eq. 7 of the main text. In an experimental set-up, when a photon is detected on either detector, there is a non-zero probability that both qubits were excited but only one was detected. This probability is given by

$$p_{\text{double}} = [P_{1,Yb} P_{1,In} P_{2,Yb} (1 - P_{2,In})] + [P_{1,Yb} P_{1,In} P_{2,In} (1 - P_{2,Yb})]$$

(18)

where the two terms come from the probability of detecting one photon from either qubit that has decayed from its excited state. There is a phase of $\pi/2$ between these two photons as with the state in Eq. 7, and an additional phase factor.
determined by the total optical path length of the In system \( \phi_D = i k D_2 \). Following through the same process, we arrive at an entangled state

\[
|\Psi_{1,2}\rangle = \frac{1}{\sqrt{2+c_1^2}} \left[ (1,0) \sum_{\omega} \xi_{Yb,\omega} c_{1\omega}^\dagger + i d_{1\omega}^\dagger |\text{vac}\rangle 
- ie^{i\phi_2} \sum_{\omega} \xi_{In,\omega} \frac{d_{\omega}^\dagger + ic_{\omega}^\dagger}{\sqrt{2}} |\text{vac}\rangle 
+ c_1 (1,1) \left[ \sum_{\omega} \xi_{Yb,\omega} c_{1\omega}^\dagger + id_{1\omega}^\dagger |\text{vac}\rangle 
- ie^{i\phi_2} \sum_{\omega} \xi_{In,\omega} \frac{d_{\omega}^\dagger + ic_{\omega}^\dagger}{\sqrt{2}} |\text{vac}\rangle \right] \right) 
\]

where we have that

\[
c_1 = \frac{\sqrt{P_{\text{double}}}}{\sqrt{p_{1,Yb}(1-p_{1,In})p_{2,Yb}}}
= \frac{\sqrt{p_{1,In} p_{2,Yb}(1-p_{2,In}) + p_{2,In}(1-p_{2,Yb})}}{\sqrt{(1-p_{1,In})p_{2,Yb}}}.
\]

Since the target state has no \( |1,1\rangle \) component, when we calculate the fidelity, we obtain the same result as before, with the only modification being the prefactor \( \frac{1}{\sqrt{2+c_1^2}} \to \frac{1}{\sqrt{2+c_1^2}} \).

Including the effect of \( F_{\text{dyn}} \) from Cabrillo et al. on photon distinguishibility, we then obtain the equation

\[
F = \frac{1}{2 + c_1^2} (1 + F_{\text{dyn}} \text{Re}(\langle \xi_{Yb} | \xi_{In} \rangle)).
\]