On-chip spin-photon entanglement based on single-photon scattering

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On-chip spin-photon entanglement based on single-photon scattering

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The realization of on-chip quantum gates between photons and solid-state spins is a key building block for quantum-information processors, enabling, e.g., distributed quantum computing, where remote quantum registers are interconnected by flying photons [1–3]. Self-assembled quantum dots integrated in nanostructures are one of the most promising systems for such an endeavor thanks to their near-unity photon-emitter coupling [4] and fast spontaneous emission rate [5]. Here we demonstrate an on-chip entangling gate between an incoming photon and a stationary quantum-dot-spin qubit. The gate is based on sequential scattering of a time-bin encoded photon with a waveguide-embedded quantum dot and operates on sub-microsecond timescale; two orders of magnitude faster than other platforms [6–9]. Heralding on detection of a reflected photon renders the gate fidelity fully immune to spectral wandering of the emitter. These results represent a major step in realizing a quantum node capable of both photonic entanglement generation [10–13] and on-chip quantum logic, as demanded in quantum networks [14] and quantum repeaters [15].

In a future quantum network [16], remote quantum nodes could be connected by a large web of entangled photons. Traditionally these photonic states have been generated probabilistically by fusing smaller states, which typically requires an exponential overhead of ancillary photons [17]. The advent of a deterministic quantum interface between light and matter promises to radically change this notion [3]. For such systems, a flying photon is funneled into a waveguide or cavity and interacts efficiently with a quantum emitter that hosts a single spin [4]. Coherent manipulation of the spin state entangles it with the photon, forming the basis for deterministic quantum gates and, e.g., the generation of photonic cluster states for quantum computing [18].

So far, significant progress has been made towards this goal, particularly the realization of spin-photon entanglement [7, 10, 11, 13, 19–22], spin-spin entanglement [7, 12, 23], single-photon switching [24, 25], photon-photon gates [6] and spin-spin gates [8, 9] using various quantum emitters. Among these platforms, quantum dots (QDs) integrated in nanophotonic waveguides offer near-unity coupling to light (β ≈ 98%) [26] and a high photon-generation rate with near-unity purity and coherence [27]. Here we implement a photon-scattering approach to realize an entangling quantum gate between a flying photon and a quantum-dot (QD) spin embedded in a planar photonic-crystal waveguide. In this manner, spin-photon entanglement is induced by the sequential scattering of a time-bin encoded photon with the QD heavy-hole spin. This mapping between a photonic qubit and a spin qubit is a key ingredient in, e.g., quantum-repeater protocols [15]. The quantum gate can be fully deterministic, but heralding on the detection of a reflected photon, renders the gate fidelity fully resistant to any residual spectral diffusion intrinsic to the emitter. We note that the coherent scattering preserves the pulse profile of an incoming photon [28], which is advantageous when connecting distant quantum nodes.

The operational principle of the entangling gate is outlined in Fig. 1. A single photon pulse is prepared in a superposition of an early |e⟩ and a late |l⟩ time-bin |ψp⟩ = α |e⟩ + β |l⟩ for α, β ∈ C constituting a flying qubit. The photon is launched into a waveguide where the embedded QD spin is initialized in |ψp⟩ = |ψ⟩. The protocol proceeds by alternating between coherent spin rotations Rŷ and single-photon scattering ˆS, cf. Fig. 1d. A ˆRŷ(π/2) pulse prepares the spin in a superposition of the two spin ground states |ψ⟩ and |ψ⟩, while ˆRŷ(π) serves two purposes: (1) to invert the spin in-between the two scattering events to create entanglement, and; (2) to prolong the spin coherence time by acting as a spin-echo pulse between the two equally long time-bins [29]. ˆS corresponds to the photon being reflected (transmitted) when the QD state is in |ψ⟩ (|ψ⟩) (Fig. 1c). The ideal gate operation results in the output state

\[ |ψ_{\text{out}}⟩ \propto (|α|e⟩ψ⟩ − β|l|ψ⟩)_{r} + (|α|e⟩ψ⟩ − β|l⟩ψ⟩)_{t} \]

which is a superposition of two spatially separated spin-photon Bell states. The subscript \(r\) (\(t\)) indicates that the photon was reflected (transmitted) by the QD. By conditioning on a photon being reflected (transmitted), the Bell state in the first (second) bracket is prepared. We find that the heralded Bell-state fidelity can be near

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unity for our system due to the spectral selectivity of the QD that predominantly reflects photons resonant with the transition $|\uparrow\rangle \rightarrow |\uparrow\downarrow\rangle$, despite residual QD spectral diffusion visible from the broadened transmission dip in Fig. 1c (see Methods; Supplementary Note II).

To calibrate the device, we subdivide the gate protocol into two separate experiments (Fig. 2). The first experiment probes the coherent nature of single-photon scattering (Fig. 2a), whereas the second experiment investigates the spin coherence with the built-in spin-echo sequence (Fig. 2c).

To demonstrate coherent scattering in the single-photon regime, we use a coherent state with a mean photon number per pulse $\bar{n}$ of the last pulse $\gamma \gg \gamma_X$. If the input photon of spectral width $\sigma_c/2\pi$ is much narrower than the QD linewidth $\Gamma/2\pi$, the photon can be fully reflected due to destructive interference in transmission [30]. By interfering the temporal modes of the reflected photon using the same interferometer and projecting on the X-basis $|\pm X\rangle_p = |e\rangle \pm |l\rangle$, the intensities $I_{e,X}$ are measured, which are used to probe the scattering coherence via the photon visibility $V_p \equiv (I_{+X} - I_{-X})/(I_{+X} + I_{-X})$. We observe a maximum value of $V_p = (89.7 \pm 0.4)\%$ that reduces linearly with $\bar{n}$ (Fig. 2b). In the single-photon scattering regime ($\bar{n} \approx 0$), the visibility is given by $V_p = 1 - 2\gamma_d/\Gamma$ (Supplementary Note III), which is only limited by the decoherence from elastic phonon scattering on the excited state with a rate $\gamma_d$. Due to the near-perfect interferometer visibility (99.6\%) [13], the pure dephasing rate can be directly extracted from the y-intercept in Fig. 2b to be $\gamma_d = (0.092 \pm 0.004)$ ns$^{-1}$ (at the operation temperature of $T = 4.2$ K), which is used to estimate the maximum entanglement fidelity, as discussed later.

The second experiment benchmarks the coherence of the internal spin qubit. Specifically we perform a spin-echo sequence by using two $\hat{R}_y(\pi/2)$ pulses separated by a $\hat{R}_y(\pi)$ pulse (Fig. 2c). The spin rotations are implemented using the two-photon Raman scheme demonstrated in Ref. [31] to optically rotate the ground states (see Methods). With the applied echo sequence, the coherence of the QD spin is probed by scanning the phase of the last $\pi/2$ pulse $\phi_r$, which projects the resulting spin
FIG. 2. Coherent single-photon scattering and spin control. (a) Setup for measuring photon visibility. The time-bin encoded qubit is reflected off the QD, which is initialized in the $|⇑⟩$ state. Upon entering the interferometer, the early time-bin is delayed which interferes with the late time-bin, constituting a $|±⟩_X = |⟩_e ± |⟩_l$ basis measurement. PBS, polarizing beam-splitter; BS, 50:50 beam-splitter; QWP (HWP), quarter (half) wave-plate. (b) Visibility of the photonic $|±⟩_X$ basis as a function of the mean photon number per pulse $\bar{n}$. The pure dephasing rate $\gamma_d$ is extracted from the y-intercept where $\bar{n} = 0$. (c) Spin-echo sequence used to probe the spin coherence. The $\pi$-pulse is equally distant from the two $\pi/2$ pulses to eliminate inhomogeneous spin dephasing. The phase of the last $\pi/2$ pulse $\phi_r$ maps the equatorial state $|⇑⟩ + e^{i\phi_r}|⇓⟩$ to the optically bright state $|⇑⟩$ ($\phi_r = \pi$) or dark state $|⇓⟩$ ($\phi_r = 0$). (d) Contrast between the spin $|⇓⟩$ and $|⇑⟩$ populations as a function of $\phi_r$. After characterizing the coherences of both qubits, we are in a position to demonstrate the entangling gate. The spin is first prepared in a superposition state $|+⟩_s \propto |⟩_e + |⟩_l$ by a 3.5 ns $\hat{R}_y(\pi/2)$ pulse (Fig. 3a). The time-bin qubit is attenuated to $\bar{n} \approx 0.07$ before interacting with the QD (Supplementary Note IV). After sequential scattering of each time-bin, the reflected signal is collected and measured by the interferometer. The heralding of the reflected photon component carves out the output state [Eq. (1)] resulting in $|φ^−⟩_r$.

To determine the fidelity of the entangled state, we perform correlation measurements between the photonic modes and spin states. This involves projecting the entangled state on the $\hat{σ}^{(p)}_i \otimes \hat{σ}^{(s)}_i$ bases, where $i \in \{x, y, z\}$ denotes the Pauli operator, and the superscripts s (p) represent the spin and photonic qubits. The state of the reflected photon is detected in different time-bin windows after the interferometer, while the spin readout is performed by applying another rotation pulse $\hat{R}_i$ followed by optical driving of the main transition (See Methods; Fig. 3a). For each experimental setting, we herald on the detection of a reflected photon and the spin readout. The entanglement fidelity is measured using [13]

$$F_{\text{Bell}} = \frac{\langle \hat{P}_z \rangle}{2} + \frac{\langle \hat{M}_y \rangle - \langle \hat{M}_x \rangle}{4},$$

where $\langle \hat{M}_i \rangle = \langle \hat{σ}^{(p)}_i \otimes \hat{σ}^{(s)}_i \rangle$ is the normalized contrast, and $\langle \hat{P}_z \rangle \equiv (1 + \langle \hat{M}_z \rangle)/2$. Figs. 3b-d show the raw (background corrected) coincidence counts in various readout bases. We record $\langle \hat{P}_z \rangle = (90.7 \pm 2.2)\%$, $\langle \hat{M}_x \rangle = (−58.8 \pm 4.5)\%$ and $\langle \hat{M}_y \rangle = (57.3 \pm 6.6)\%$, where resid-
FIG. 3. Generation and verification of Bell states. (a) Experimental sequence consisting of the preparation of spin and photonic qubits, the gate protocol and the readout. The spin state is initialized and read out by optically driving the $|\uparrow\rangle \leftrightarrow |\uparrow\downarrow\rangle$ transition (pale red), and $\hat{R}_i$ controls the spin projection basis. The photonic qubit is prepared and measured by the same interferometer in either the Z-basis (green) or equatorial basis (blue) time window. (b) Raw two-photon coincidences measured during the photonic (p) readout window and spin (s) projections. (c) Measured two-photon coincidences after correcting for laser background from rotation pulses [13]. (d) Visibility fringes of background-corrected two-photon coincidences as a function of the qubit phase $\theta_p$ when the spin state is projected on $|\uparrow\rangle \propto |\uparrow\rangle - |\downarrow\rangle$. Circles (squares/triangles) correspond to projection on the photonic X-basis ($\pm$-Z-basis). Solid curves are fits using $V_s \cos(\theta_p + \theta_{offset})$, and dashed lines are horizontal line fits.

To unravel the physical mechanisms limiting the experimental fidelity, we derive the fidelity (Supplementary Note II) in the perturbative limit of small errors,

$$ F_{\text{theory}}^{\text{Bell}} \approx 1 - \frac{\gamma_d}{\Gamma} - \frac{\Gamma^2}{4\Delta_h^2}, $$

where $\Delta_h$ is the ground-state splitting. Eq. (3) holds in the regime $\gamma_d \ll \Gamma \ll \Delta_h$ and assumes perfect manipulation of the hole spin state. In addition to being resilient to ground-state dephasing due to the built-in spin echo, the protocol is also impervious to errors arising from the spectral mismatch between the incoming optical pulse and the QD transition. This robustness is granted by the QD spectral reflectivity, which sifts out events where the photon interacts with the QD. The off-resonant frequency component of the incident pulse is transmitted without being detected thus having no impact on the gate fidelity. Using Eq. (3) with experimental parameters, the theoretical fidelity is estimated to be $F_{\text{Bell}}^{\text{theory}} = 96.2\%$ (Supplementary Note II). Here the infidelity results from decoherence from elastic phonon scattering $\gamma_d (3.7\%)$ and reflection from the off-resonant spin state $\Gamma/\Delta_h (0.1\%)$. The comparison to the experimental result indicates that several additional error mechanisms influence the experiment. The dominant cause is an incoherent photo-induced spin-flip error leading to non-ideal spin rotations, as is visible on the spin-echo data (Fig. 2d). These rotation errors together amount to a total infidelity of 17% (Supplementary Note II). Additional sources of error originate from driving-induced dephasing due to finite $\bar{n} (6.3\%)$ and imperfect spin readout (2.7%), which are not intrinsic to the protocol. Taking these into account we estimate $F_{\text{total}}^{\text{theory}} \approx 72.3\%$, which agrees with the experimental value within the error margins. Suppressing the photo-induced incoherent spin-flip processes is essential for improving the fidelity further.
Encouragingly spin-rotation fidelities of 98.9\% have been realized in the literature on electron spins [31] and could be combined with nuclear-spin cooling methods to realize $T_2^*$ beyond 100 ns [32]. With these improvements a near-unity entanglement fidelity is within reach.

Since the present experimental realization of the scattering gate is conditioned on the detection of a reflected photon, the overall gate efficiency is bounded to at most 50\% (Supplementary Note II). By adopting a single-sided waveguide or equivalently coupling the incident light to both reflection and transmission ports via a stabilized interferometer, a fully deterministic spin-photon quantum gate [33] can eventually be realized. For a single-sided device all photons are reflected but with a spin-dependent phase. As such, no heralding of gate operation is required though the gate fidelity will be sensitive to spectral drifts of the QD transition to the second order. This however does not pose a fundamental limit to our platform as near-lifetime-limited QD transitions in photonic structures compatible with the proposed scheme were recently reported [34].

We have demonstrated a photon-scattering quantum gate whereby an incoming photonic qubit becomes entangled with a stationary spin. The system versatility is reflected by the fact that the same QD can also be operated as a source of multi-photon time-bin encoded entanglement generation [13]. Such versatile spin-photon interfaces constitute building blocks of one-way quantum repeaters [15]. Furthermore, a range of new integrated quantum photonics devices and functionalities could potentially be realized, e.g., a deterministic Bell-state analyzer or a photonic quantum non-demolition detector [33, 35] that both rely on faithful coherent quantum state transfer from a flying photon to an emitter. The reflection-based scheme could also be extended to realize non-local quantum entangling gates between distant quantum emitters [8]. Finally, applying the above spin-photon gate interleaved with spin rotations would realize entanglement between two subsequently incoming photons, i.e. a deterministic photon-photon quantum gate [6], which is the most challenging quantum operation in photonics.

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Author Contributions


Competing interests P.L. is founder of the company Sparrow Quantum that commercializes single-photon sources.

Data availability The data are available from the corresponding authors upon reasonable request.

Online Methods

Spin-photon interface. To achieve the highly efficient light-matter interaction required by the protocol, we prepare a QD embedded in a suspended photonic-crystal waveguide (PCW) with two ports (Supplementary Note I). The p-i-n heterostructure contains an intrinsic layer of self-assembled InAs QDs, enabling the electrical control of the QD charge state by applying a forward bias voltage. One experimental challenge is to simultaneously realize optical cycling transitions and spin control. This was recently achieved with a QD in a PCW under an in-plane magnetic field (Voigt geometry) by exploiting the inherent radiative asymmetry of the PCW [36]. We employ a positively charged exciton giving access to a meta-stable hole spin ground state that was characterized in previous work [36]. A spin initialization fidelity of up to 98.6\% is realized together with a spin dephasing time of $T_2^* > 20$ ns, which are essential for the entangling gate. An in-plane external magnetic field ($B_y = 2$ T) Zeeman-splits the QD spin state into four energy levels, see Fig. 1h, where the linearly X- and Y-polarized dipoles form two Λ-systems. Thanks to the optical cyclicity of $C = \gamma_Y / \gamma_X = 14.7 \pm 0.2$ where the radiative decay rate $\gamma_Y$ ($\gamma_X$) is strongly enhanced (suppressed) by the PCW, an effective two-level system $|\uparrow\rangle \leftrightarrow |\uparrow\downarrow\rangle$ resembling a “QD mirror” is realized. This leads to spin-dependent reflection of photons into the same frequency and polarization modes, granting the spectral selectivity necessary for the quantum gate protocol.

Experimental setup. To perform high-fidelity entanglement experiments, the sample chip is cooled to 4.2 K inside a closed-cycle cryostat to suppress phonon scattering. A superconducting vector magnet provides a 2 T in-plane magnetic field enabling Raman transitions between two hole ground states. The sample is imaged with a 0.81 NA objective and brought to focus by translating 3 piezo positioners mounted beneath the sample.
A DC voltage source provides a bias voltage across the sample to populate QD charge states via tunnel coupling to a Fermi reservoir and control the charge environment. The experiment utilizes the same laser setup as in Ref. [13] with a few notable differences: Two continuous wave (CW) lasers (linewidth < 10 kHz) are used for the creation of the photonic qubit, resonant excitation of the QD and spin rotations. One of which is first directed to a double-pass acousto-optic modulator (AOM) setup followed by an electro-optical modulator (EOM); iXBlue NIR-MX800-LN-20) to generate 2 ns (FWHM) pulses for the photonic qubit. The non-diffracted light from the first AOM setup is then sent to a second AOM setup to create spin initialization and readout pulses (200 ns each) of the same laser frequency. The qubit laser pulses and QD emission are focused and collected at the same grating outcoupler using a cross-polarization scheme (Supplementary Note I), while the readout laser is coupled directly on top of the QD (Fig. 1a). Another CW laser is used for coherent spin control. It is sent through a third AOM setup and another EOM which is amplitude-modulated by a microwave source to create a 7 ns π/2-pulse. The phase of the last microwave π/2-pulse is induced by a combination of a phase shifter and switches [13] with a phase offset of ~ 0.3π. The total pulse sequence duration is set to 606 ns. A photonic qubit encoded in the ground state splitting $\Delta$ of $\approx 9\text{GHz}$ is driven by $\Delta_h/2\pi = 7.3\text{GHz}$, thus effectively driving the ground-state spin manifold. The sidebands are red-detuned from the cycling transition by 350 GHz to avoid populating the excited states. The phase $\phi_v$ of the last microwave π/2-pulse is induced by a combination of a phase shifter and switches [13] with a phase offset of ~ 0.3π. The total pulse sequence duration is set to 606 ns. A photonic qubit encoded in time-bins is created by passing the 2 ns pulses through an asymmetric Mach-Zehner interferometer with a time delay of 11.82 ns. Here we chose the FWHM duration for the input pulse to be 2 ns which exceeds the radiative lifetime of the optical transition $\Gamma^{-1} = 0.4\text{ ns}$ for efficient single-photon scattering, but is narrow enough to be fitted within the 11.8 ns time delay when combined with a 7 ns π-rotation pulse and 1 ns rise/fall time. The qubit phase $\theta_p$ can be scanned using a quarter-waveplate (QWP) and a linear polarizer. The reflected signal is then reinserted into the same interferometer and subsequently two narrowband (3 GHz) etalon filters to remove background from the rotation laser as well as QD phonon sidebands. The filtered signal passes through a QWP and an EOM (not shown) which sets a 50/50 splitting ratio on the polarizing beam-splitter (see Fig. 2a). Since both the photonic qubit preparation and readout are performed via the same interferometer, the experiment becomes very robust against any mechanical or thermal drift allowing near-unity interferometric visibility on a week-long timescale [13].

**Spin-photon state projections.** As shown in Fig. 3a, the detection of an early (late) photon traversing through the long (short) path of the interferometer constitutes the $\hat{\sigma}_z^{(p)}$-basis measurement (green). The spin readout in the $\hat{\sigma}_z^{(s)}$-basis is performed by applying another rotation pulse $R_\phi(\theta_p) = R_y(\theta_p) R_y(\pi)$ followed by optical driving of the main transition. Similarly, projection on the $\hat{\sigma}_x^{(p)} \otimes \hat{\sigma}_x^{(s)}$ ($\hat{\sigma}_y^{(p)} \otimes \hat{\sigma}_y^{(s)}$) bases is performed by detecting photons in the middle time window (blue) at $\theta_p \approx 2\pi \equiv \theta_0 + \pi/2$ where the early and late time-bins between the short and long paths [13] interfere, followed by $R_\phi = R_y(\pm\pi/2)$ ($R_z(\pi/2)$) before the spin readout.

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Supplementary Notes - On-chip spin-photon entanglement based on single-photon scattering

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I. CROSS-POLARIZATION SCHEME ON WAVEGUIDES

The waveguide device used in the current experiment is terminated by two high-efficiency grating outcouplers [S1] representing reflection and transmission ports. For implementing the scattering experiment, both the incident laser and reflected signal are coupled to the same grating outcoupler (Fig. S1). To distinguish the signal from the laser background, a cross-polarized scheme is used for excitation and collection. Before entering the waveguide, the polarization of the input light is carefully optimized with a set of waveplates such that it is orthogonal to the polarization of the collected light (set by another set of waveplates on the optical setup). The input is diagonally polarized (which can also be circularly polarized), thus only 50% of the light is coupled to the grating coupler which has a predefined polarization along X [S1]. The X-polarized light is then converted into Y-polarization via the bend in the waveguide and subsequently interacts with the QD. Due to the non-chiral coupling of the waveguide, 50% of the scattered signal thus returns to the same grating coupler, and further passes through the polarization control on the collection path resulting in another 50% loss. Despite the loss in efficiency, the signal-to-noise ratio achieved in this setup can reach $\sim$100-300 depending on the mechanical stability of the optical setup. The transmission port constitutes a second collection path which can be used for both resonant transmission and entangling gate experiments.

![Figure S1](image-url)

**FIG. S1.** Scanning Electron Microscope (SEM) image of the waveguide and polarizations of the input and reflected light. Dark grey arrows denote the predefined polarizations of the grating couplers.

II. SCATTERING THEORY OF SPIN-PHOTON BELL STATE GENERATION

In this section, we discuss the state evolution of the spin-photon system upon applying the entangling gate and develop an analytical expression for the entanglement fidelity. Our strategy is similar to the approach taken in Ref. [S2], which is to first evaluate the fidelity to lowest order in perturbation theory for each of the considered errors. In the end the full fidelity is then found by multiplying the individual fidelities.

We start with modeling a right-propagating time-bin photonic qubit $\alpha |e\rangle + \beta |l\rangle$ in a two-sided waveguide where $\alpha, \beta \in \mathbb{C}$, and the QD spin is initially in the ground state $|\downarrow\rangle$. Following from Fig. 1, the gate protocol consists of (1) applying a $\hat{R}_y(\pi/2)$ spin rotation to prepare a superposition spin qubit, (2) scattering of the early photon $\hat{S}_e$, (3) a $\hat{R}_y(\pi)$-rotation, and finally (4) the scattering of the late photon $\hat{S}_l$. Each single-photon scattering process obeys the input-output relations [S3]:

$$
\begin{align*}
|\omega \uparrow\rangle & \rightarrow r_1 |\omega \uparrow\rangle_r + r_1 |\omega \uparrow\rangle_t + r_2 |\omega \downarrow\rangle_r + t_2 |\omega \downarrow\rangle_t ; \\
|\omega \downarrow\rangle & \rightarrow r_1 |\omega \downarrow\rangle_r + t_1 |\omega \downarrow\rangle_t + r_2 |\omega - \Delta \downarrow\rangle_r + t_2 |\omega - \Delta \downarrow\rangle_t ,
\end{align*}
$$

(S1)

where the photon in each time-bin is assumed to center around the resonant frequency of the dominant transition ($|\uparrow\rangle \leftrightarrow |\uparrow\downarrow\rangle$) with a Gaussian spectral profile, and $r_1$ ($t_1$) are the reflection (transmission) operators associated with the QD vertical transition $|\uparrow\rangle \rightarrow |\uparrow\downarrow\rangle$ with a decay rate $\Gamma_1 (\gamma_Y$ in the main text). $r_2$ ($t_2$) corresponds to the diagonal...
transition $|\psi\rangle \rightarrow |\uparrow\psi\rangle$ with decay rate $\Gamma_2 (\equiv \gamma_X)$. $\omega_2 = \omega + \Delta_h$ is the frequency of the Raman photon emitted from the diagonal transition where $\Delta_h$ is the ground-state splitting. The symbol $\circ$ denotes off-resonant scattering when the spin is in $|\psi\rangle$. Using Eq. (S1), the state evolution of the spin-photon system proceeds as

\[
\left( \alpha |e\rangle + \beta |l\rangle \right) \otimes |\psi\rangle \overset{R_0(\pi)}{\longrightarrow} \left( \alpha |e\rangle + \beta |l\rangle \right) \otimes \left( |\uparrow\rangle + |\downarrow\rangle \right) / \sqrt{2}
\]

\[
\overset{\mathcal{S}_r}{\longrightarrow} \left( \alpha \left( r_1^e |e\rangle_r + t_1^e |e\rangle_t + r_2^e |e\rangle_r + t_2^e |e\rangle_t \right) + \alpha \left( r_1^e |e\rangle_r + t_1^e |e\rangle_t - r_2^e |e\rangle_r - t_2^e |e\rangle_t \right) - \beta |l\rangle \right) / \sqrt{2}
\]

\[
\overset{R_0(\pi)}{\longrightarrow} \left( \alpha \left( - r_1^e |e\rangle_r - t_1^e |e\rangle_t + r_2^e |e\rangle_r + t_2^e |e\rangle_t \right) + \alpha \left( r_1^e |e\rangle_r + t_1^e |e\rangle_t - r_2^e |e\rangle_r - t_2^e |e\rangle_t \right) - \beta |l\rangle \right) / \sqrt{2}
\]

In the ideal scenario where the early and late pulses are identical, monochromatic and resonant, and the QD optical cyclicity is infinite with no dephasing and loss, we have: (1) $r_1 \to 1$ (resonant photons are coherently reflected with a $\pi$-phase shift), (2) $t_1 \to 0$ (off-resonant photons are being transmitted instead of reflected), (3) $t_1 \to 0$ (complete destructive interference in the transmission); and (4) $r_2, t_2, t_f \to 0$ (there are no Raman photons in the reflected and transmitted modes due to high cyclicity). As such, the ideal output state of the gate becomes $\alpha |e\rangle - \beta |l\rangle \rangle_r + \alpha |e\rangle - \beta |l\rangle \rangle_t$. Heralding on either the reflection or transmission of a scattered photon projects the system into a different spin-photon Bell state. By varying the phase $\theta_r$ of the photonic qubit where $\beta/\alpha = e^{i\theta_r}$ and $|\alpha|^2 + |\beta|^2 = 1$, all 4 maximally entangled states can be generated by the gate.

### A. Scattering coefficients for a Λ-level emitter in two-sided waveguides

The scattering problem of a weak coherent state on the Λ-level emitter has been solved in Ref. [S4] and its formalism can be easily extended to directly compute the scattering coefficients in Eq. (S1). Specifically, the output field bosonic operator of the waveguide can be expressed in terms of the incident field and dynamical response of the emitter from the non-Hermitian Hamiltonian $\hat{H}_{\text{inh}}$ [S2]. In a two-sided waveguide configuration, we label the field operator in the reflection port by the subscript “r”, and the transmitted port by “t” (Fig. S2). Assuming that a right-propagating light field $a_{\text{in},r}^l$ enters the waveguide, the output field operators on the transmitted (t) and reflected (r) ports are

\[
\begin{align*}
|\omega \rangle \otimes |\uparrow\rangle : \\
\begin{cases} 
\hat{a}_{\text{out},t} = 1 - \frac{2\Gamma_1^t}{\Gamma + 2i\delta_f} \sigma_{11}^{\uparrow} - \frac{2\sqrt{\Gamma_1^t \Gamma_2^t}}{\Gamma + 2i\delta_f} \sigma_{12}^{\uparrow} \hat{a}_{\text{in},t} \\
\hat{a}_{\text{out},r} = - \frac{2\sqrt{\Gamma_1^r \Gamma_2^r}}{\Gamma + 2i\delta_f} \sigma_{11}^{\uparrow} - \frac{2\sqrt{\Gamma_1^r \Gamma_2^r}}{\Gamma + 2i\delta_f} \sigma_{12}^{\uparrow} \hat{a}_{\text{in},r} 
\end{cases}
\end{align*}
\]

\[
\begin{align*}
|\omega \rangle \otimes |\downarrow\rangle : \\
\begin{cases} 
\hat{a}_{\text{out},t} = 1 - \frac{2\Gamma_1^t}{\Gamma + 2i\delta_f} \sigma_{11}^{\downarrow} - \frac{2\sqrt{\Gamma_1^t \Gamma_2^t}}{\Gamma + 2i\delta_f} \sigma_{12}^{\downarrow} \hat{a}_{\text{in},t} \\
\hat{a}_{\text{out},r} = - \frac{2\sqrt{\Gamma_1^r \Gamma_2^r}}{\Gamma + 2i\delta_f} \sigma_{11}^{\downarrow} - \frac{2\sqrt{\Gamma_1^r \Gamma_2^r}}{\Gamma + 2i\delta_f} \sigma_{12}^{\downarrow} \hat{a}_{\text{in},r} 
\end{cases}
\end{align*}
\]

where $\delta_f = \omega_1 - \omega$ is the laser detuning from the transition $|\uparrow\rangle \rightarrow |e\rangle$ for an emitter initialized in $|\uparrow\rangle$. The total decay rate $\Gamma = \Gamma_1 + \Gamma_2 + \gamma_1 + \gamma_2$ where $\Gamma_i (\gamma_i)$ is the radiative decay rate into (out of) the waveguide. $\Gamma_i = \Gamma_i^t + \Gamma_i^r$ includes
both decay rates into the transmitted (‘t”) and reflected (‘r”) waveguide modes. \( \Delta_h \) is the ground-state splitting. The output field operators have different detunings in their denominators because of different initial spin states of the QD: If the spin is initially \(| \uparrow \rangle \), the resonant frequency is \( \omega_1 \); If it is \(| \downarrow \rangle \) then the resonant frequency required to drive the diagonal spin transition is \( \omega_2 = \omega_1 + \Delta_h \). \( \hat{\sigma}_{ij} = |j \rangle \langle i | \) is the atomic operator denoting a spin-flip in the atomic state when \( i \neq j \). Note that when evaluating the probability of a spin-phonon state, i.e., \(| e \rangle \langle \downarrow | \), the corresponding scattering coefficient \( r_i^f(\omega) \) is first convoluted with a Gaussian lineshape \( \Phi_1(\omega) \) and integrated with respect to \( \omega \) [S2]. The individual resonant scattering coefficients in the frequency domain are

\[
\begin{align*}
  t_1(\omega) &= 1 - \frac{2\Gamma_1^t}{\Gamma + 2i\delta_1}, \\
  t_2(\omega) &= -\frac{2\sqrt{\Gamma_1^t\Gamma_2^r}}{\Gamma + 2i\delta_1}, \\
  r_1(\omega) &= -\frac{2\sqrt{\Gamma_1^t\Gamma_1^r}}{\Gamma + 2i\delta_1}, \\
  r_2(\omega) &= -\frac{2\sqrt{\Gamma_1^t\Gamma_2^r}}{\Gamma + 2i\delta_1},
\end{align*}
\]

(S4)

where the off-resonant scattering coefficients are found similarly by replacing \( \delta_1 \to \delta_1 + \Delta_h \).

### B. Projection operators for measuring time-bin encoded photons

At the end of the entangling gate, measurements to read out the state of the photonic qubit are performed by registering detector clicks in three different detection time windows. The detection of a time-bin photon is formulated by projection operators on different photonic readout bases:

\[
| e \rangle \langle e | = \int_{-\infty}^{\infty} \hat{a}_e^\dagger(t)\hat{a}_e(t)dt, \quad | l \rangle \langle l | = \int_{-\infty}^{\infty} \hat{a}_l^\dagger(t+\tau)\hat{a}_l(t+\tau)dt, \quad | e \rangle \langle l | = \int_{-\infty}^{\infty} \hat{a}_e^\dagger(t)\hat{a}_l(t+\tau)dt = (| l \rangle \langle e |)^\dagger,
\]

(S5)

where the bosonic creation operator \( \hat{a}_e^\dagger(t) \) represents the emission of a photon at time \( t \) in the early time-bin, and \( \tau \) is the interferometric delay. The projections \( | e \rangle \langle e | \) (\( | l \rangle \langle l | \)) correspond to detecting photons in the side peak windows (green) (Fig. 3a), whereas \( | e \rangle \langle l | \) refers to projection onto the middle detection window (blue central peak) where the early and late photons interfere. Since we only resolve the time-bin, the creation operator can be expressed in either the time or frequency domain. Using \( a(t) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} a(\omega)e^{i\omega t}d\omega \), one can show

\[
\int_{-\infty}^{\infty} \hat{a}_e^\dagger(t)a_c(t)dt = \int_{-\infty}^{\infty} \hat{a}_e^\dagger(\omega)a_c(\omega)d\omega.
\]

(S6)

This implies we can adopt the same perturbation theory in the frequency domain to evaluate the fidelity as in Ref. [S2].
C. Formula for the operational fidelity

Now, with the time-bin projection operators defined, we can express the entanglement fidelity in terms of the above scattering coefficients. The measure of the quality of generated quantum states is conventionally given by the fidelity, which in our case evaluates the overlap between the output and ideal Bell states:

\[ F_r^{\text{theory}} = \frac{\langle \psi_{\text{ideal}} | \rho_{\text{out}} | \psi_{\text{ideal}} \rangle}{\text{Tr}(\rho_{\text{out}} \rho_{\text{ideal}})} \, . \tag{S7} \]

Here the output reduced density matrix is given by \( \rho_{\text{out}} = \text{Tr}_{\omega}(|\psi_{\text{out}}\rangle \langle \psi_{\text{out}}|) \) which is a partial trace of the output density matrix \( |\psi_{\text{out}}\rangle \langle \psi_{\text{out}}| \) over the transmitted modes and frequency states \( \omega \neq \omega_1 \) not detected in the reflection. The total output density matrix \( \rho_{\text{out}} \) is therefore obtained by effectively tracing out the unwanted modes. For simplicity we assume the use of perfect filters prior to detection which removes photons of frequencies other than \( \omega_1 \). The bandwidth of the etalon filters used in the experiment is \( \sim 3 \) GHz with over 95% transmission. This means the filter bandwidth is much narrower than the ground-state splitting \( \Delta_\hbar/2\pi = 7.3 \) GHz but wider than the QD homogeneous linewidth \( \Gamma/2\pi = 394 \) MHz justifying the assumption. Similarly only the reflected mode is collected. The fidelity in Eq. (S7) is normalized by the success probability or gate efficiency \( P_s = \text{Tr}(|\psi_{\text{out}}\rangle \langle \psi_{\text{out}}|) \equiv \sum_i \langle i_r | (|\psi_{\text{out}}\rangle \langle \psi_{\text{out}}|) |i_r \rangle \), since the gate is heralded by the detection of a photon in the reflection. In such case any event contributing to the loss of the scattered photon (e.g., finite cyclicity, non-zero coupling to leaky modes of the waveguide, and the transmission of a photon, which is effectively treated as loss) does not reduce the gate fidelity.

Using Eq. (S2), the normalized output reduced density matrix is found to be

\[ \rho_{\text{out}} \equiv \frac{1}{2P_s} \left( |\alpha|^2 |r_1|^2 |e \downarrow \rangle \langle e \downarrow | - |\alpha^* r_1^*|^2 |e \downarrow \rangle \langle e \downarrow | - |\alpha^* r_1^*|^2 |l \uparrow \rangle \langle l \uparrow | + |\alpha|^2 |r_1|^2 |e \uparrow \rangle \langle e \uparrow | + \ldots \right) \]

For instance we write out two of the matrix elements in \( \rho_{\text{out}} \) using the results from Sec. II A and Eq. (S6):

\[ \frac{1}{2} |\alpha|^2 |r_1^*|^2 |e \downarrow \rangle \langle e \downarrow | = \frac{1}{2} |\alpha|^2 \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} d\omega d\omega' r_1(\omega) r_1^*(\omega') \Phi_1(\omega') \Phi_1(\omega) \hat{a}_r(\omega) \hat{a}_r(\omega') ; \]

\[ \frac{1}{2} |\alpha|^2 |r_1^*|^2 |e \downarrow \rangle \langle l \uparrow | = \frac{1}{2} |\alpha|^2 \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} d\omega d\omega' r_1(\omega) r_1^*(\omega') \Phi_1(\omega') \Phi_1(\omega) \hat{a}_r(\omega) \hat{a}_r(\omega') ; \]

where \(|\theta\rangle\) is the vacuum state. For simplicity we now say that the early and late scattering events are identical for any given input frequency thus \( r_1^* = r_1 \). The reasoning behind this is further discussed in Sec. IID. Therefore, for an ideal Bell state in the reflected mode: \(|\psi_{\text{ideal}}\rangle = |e \downarrow \rangle - |l \uparrow \rangle\), the overlap of one of the density matrix elements with the ideal state becomes

\[ \langle \psi_{\text{ideal}} | \frac{1}{2} |\alpha|^2 |r_1^*|^2 |e \downarrow \rangle \langle e \downarrow | \psi_{\text{ideal}} \rangle \]

\[ = \frac{1}{2} \int_{-\infty}^{\infty} d\omega d\omega' d\omega'' (|e \downarrow \rangle \langle e \downarrow | - |l \uparrow \rangle \langle l \uparrow |) \left[ r_1(\omega) r_1^*(\omega') \Phi_1(\omega') \Phi_1(\omega) \hat{a}_r(\omega') \hat{a}_r(\omega) \right] ; \]

\[ = \frac{1}{2} \int_{-\infty}^{\infty} d\omega d\omega' d\omega'' r_1(\omega) r_1^*(\omega') \Phi_1(\omega') \Phi_1(\omega) \delta(\omega - \omega') \delta(\omega' - \omega'') \]

\[ = \frac{1}{2} |\alpha|^2 \int_{-\infty}^{\infty} \Phi_1(\omega)^2 r_1(\omega)^2 d\omega . \]

Including all terms, the conditional fidelity is found to be

\[ F_r^{\text{theory}} = \frac{1}{2P_s} \int_{-\infty}^{\infty} |\Phi_1(\omega)|^2 r_1(\omega)^2 d\omega , \tag{S8} \]

where the success probability \( P_s \) is the trace of the output density matrix over the four basis states \(|i\rangle = (|e \downarrow \rangle_r , |e \downarrow \rangle_r , |l \uparrow \rangle_r , |l \uparrow \rangle_r)\) in the Hilbert space of the spin-photon system. It is given by

\[ P_s = \text{Tr}(|\psi_{\text{out}}\rangle \langle \psi_{\text{out}}|) = \sum_i \langle i_r | (|\psi_{\text{out}}\rangle \langle \psi_{\text{out}}|) |i_r \rangle = \frac{1}{2} \int_{-\infty}^{\infty} |\Phi_1(\omega)|^2 r_1(\omega)^2 d\omega + \int_{-\infty}^{\infty} |\Phi_1(\omega)|^2 r_1(\omega)^2 d\omega . \tag{S9} \]

Combining Eqs. (S8) and (S9) results in the formula for the gate fidelity conditioned on reflected photons

\[ F_r^{\text{theory}} = \frac{\int_{-\infty}^{\infty} |\Phi_1(\omega)|^2 r_1(\omega)^2 d\omega}{\int_{-\infty}^{\infty} |\Phi_1(\omega)|^2 r_1(\omega)^2 d\omega + \int_{-\infty}^{\infty} |\Phi_1(\omega)|^2 r_1(\omega)^2 d\omega} . \tag{S10} \]
D. Perturbative form of the entanglement fidelity

The two integrals in Eq. (S10) are the probabilities of detecting a photon of frequency \( \omega_1 \) originated from the resonant scattering of the spin state \( |↑⟩ \) and the off-resonant reflection from \( |↓⟩ \) respectively. In particular, using Eq. (S4) we find

\[
\int_{-\infty}^{\infty} |\Phi_1(\omega)|^2 |r_1(\omega)|^2 d\omega = \int_{-\infty}^{\infty} e^{-\frac{(\omega - \omega_1)^2}{2\sigma^2}} \left| 2\sqrt{\Gamma_1 \Gamma_2} \right|^2 d\omega \approx 1 - \frac{4\sigma^2}{\Gamma^2} - \frac{\Gamma^2 - \Gamma_1^2}{\Gamma^2};
\]

\[
\int_{-\infty}^{\infty} |\Phi_1(\omega)|^2 |r_1(\omega)|^2 d\omega \approx \frac{\Gamma_1^2}{\Gamma^2 + 4\Delta_h^2},
\]

where we assume that the scattered photon is equally coupled to the reflected and transmitted modes, i.e., \( \Gamma^r = \Gamma^t = \Gamma_i/2 \). \( \sigma \) is the standard deviation of the spectral width of the incident Gaussian pulse. In evaluating Eq. (S11) perturbatively we assume the frequency detuning \( \delta_1 \) to be small compared to the QD total decay rate \( \Gamma \) and the ground-state splitting \( \Delta_h \) for efficient light-matter interaction.

1. Spectral mode mismatch

By heralding on the detection of a reflected photon of frequency \( \omega_1 \) within the time-bin window, the entanglement fidelity becomes immune to the spectral error due to the nonzero bandwidth \( \sigma_o \) of the incident pulse to lowest order in perturbation theory. Using Eqs. (S10) and (S11), the resultant fidelity is

\[
F^\text{theory}_r \approx 1.
\]

Simply stated, photons which are not resonant with the QD transition will be transmitted instead of reflected. Since the gate is conditioned on the reflection of either an early or a late photon, the transmission of the photon only reduces the success probability. The gate will thus have unity fidelity as long as the dynamics of the early and late scattering events are identical. The same argument can be made for the broadening of the QD optical transition due to slow spectral diffusion compared to the QD lifetime. The spectral jittering on the QD resonance is modelled by taking \( \delta_1 \rightarrow \delta_1 + \delta_e \) where \( \delta_e \) follows a Gaussian spectral diffusion profile \( N(0, \sigma_e) \) [S2].

If the entangling gate is heralded on the presence of transmitted photon; however, the fidelity becomes susceptible to the spectral mismatch error. A similar fidelity analysis shows

\[
F^\text{theory}_t \approx 1 - \frac{4\sigma^2}{\Gamma^2} - \frac{\Gamma^2 - \Gamma_1^2}{\Gamma^2},
\]

as the spectral infidelity arises from incomplete destructive interference between the incident field and the resonantly scattered photon \( (t_1 \neq 0) \). Any spectral effects reducing this interference would stain the quality of the entangled state. It is important to note that despite the QD spectral reflectivity, there is still a small probability of detecting undesired Raman photons of frequency \( \omega_2 = \omega_1 + \Delta_h \) in the reflection due to the finite optical cyclicity. These photons result from the imperfect QD two-level system and are filtered out.

2. Finite cyclicity and coupling loss

On the reflection port, photons could either originate from (i) resonant reflection on the spin-preserving transition (indicated by \( r_1 \)), (ii) resonant Raman spin-flip process to \( |\uparrow⟩ \) (\( r_2 \)), or (iii) off-resonant reflection from \( |\downarrow⟩ \) (\( r_1 \)). A high cyclicity reduces the probability of resonant spin-flip process but strengthens off-resonant reflection. The undesired events (ii) and (iii) can be reduced by having a larger ground-state splitting \( \Delta_h \gg \Gamma \). Coupling to lossy modes of the waveguide implies that the reflected photons are lost without being detected; as a result these events do not affect the fidelity. Effectively we find

\[
F^\text{theory}_r \approx 1 - \frac{\Gamma^2}{4\Delta_h^2} \left( \frac{C}{C+1} - \frac{\gamma_1}{\Gamma} \right)^2 \left[ 2 - \left( \frac{C}{C+1} - \frac{\gamma_1}{\Gamma} \right)^2 \right] \approx 1 - \frac{\Gamma^2}{4\Delta_h^2}.
\]

Here \( \gamma_1 \) is the radiative rate from the main transition \( |\uparrow⟩ \rightarrow |\uparrow\uparrow⟩ \) which couples to lossy modes. In deriving Eq. (S14) we define the optical cyclicity \( C \equiv (\Gamma_1 + \gamma_1)/(\Gamma_2 + \gamma_2) \) [S5] and the total decay rate \( \Gamma = \Gamma_1 + \Gamma_2 + \gamma_1 + \gamma_2 \) where \( \Gamma_2 \) \( (\gamma_2) \) is the radiative decay rate into (outside) the waveguide.
3. Phonon-induced pure dephasing

The interaction of the QD with a phononic environment results in the broadening of the zero-phonon line and a broad phonon sideband \([S6–S9]\). The latter can be filtered out while the former contributes to the reflection of incoherent photons which scramble the phase coherence of the spin-photon Bell state. The incoherent photons are only slightly broadened and thus cannot easily be removed by filters.

We follow the approach in Ref. \([S2]\) and model this incoherent process as Markovian decoherence given by a dephasing rate \(\gamma_d\) with the Lindblad operator \(\sqrt{2\gamma_d}\sigma_{ee}\) where \(\sigma_{ee} \equiv |↑\downarrow\rangle\langle \downarrow|\) is the atomic excited state. The dephasing leads to a quantum jump to the excited state (with a dephasing probability \(P_{\gamma_d}^{\omega_2}\)) followed by the decay to either of the two hole ground states with probabilities set by the transition rates \(\Gamma_i/\Gamma\). The emitted photon into the waveguide is represented by a normalized photon density matrix \(\rho_{\gamma_d}^{\omega_2}\). This is described by the density matrix

\[
\rho' = \rho + P_{\gamma_d}^{\omega_2} \rho_{\gamma_d}^{\omega_2} \otimes |\downarrow\rangle\langle \downarrow|,
\]

where \(\rho\) is the density matrix without a dephasing quantum jump. Initially there are also incoherent photons of frequency \(\omega_2\) due to finite optical cyclicity but these are subsequently filtered out together with phonon sidebands. \(\rho_{\gamma_d}^{\omega_2} \otimes |\downarrow\rangle\langle \downarrow|\) is the photon density matrix resulting from the incoherent dephasing with a probability given by

\[
P_{\gamma_d}^{\omega_2} = \frac{\Gamma'}{\Gamma} P_{\gamma_d} \approx \frac{\Gamma'}{\Gamma} \left[-2\sqrt{\frac{2\gamma_d}{\Gamma}}\right]^2 = 2\gamma_d \left(1 - \frac{1}{C+1} - \frac{\gamma_1}{\Gamma}\right) \left(1 - \frac{1}{C+1} - \frac{2\gamma_d}{\Gamma} - \frac{\gamma_1}{\Gamma}\right].
\]

Here we take \(\Gamma = \Gamma_1 + \Gamma_2 + 2\gamma_d + \gamma_1 + \gamma_2\) with the optical cyclicity \(C \equiv (\Gamma_1 + \Gamma_2)/(\gamma_1 + \gamma_2)\) and \(\Gamma_i/2 = \Gamma'_i\) to find \(\Gamma'_i = |C(\Gamma - 2\gamma_d)/(C + 1) - \gamma_1|/2\) since the dephasing effectively becomes an additional decay channel; however, the probability for the incoherent excited state to decay is governed only by the branching ratio thus \(\Gamma_i/\Gamma = |C/(C+1) - \gamma_1/\Gamma|/2\).

To evaluate the effect of pure dephasing in the gate protocol, it is instructive to consider the propagation of the error as there are two separate scattering events which will both lead to incoherent decay. Since Eq. (S15) depends on whether there is a quantum jump to the excited state, we can assume that pure dephasing occurs primarily when the incident photon is resonant with the QD state since the excited state is unlikely to be populated via off-resonant scattering. As such, using Eq. (S15) there are two additional incoherent density matrices in the normalized output reduced density matrix

\[
\rho_{\text{out}}' = \frac{P_s \rho_{\text{out}} + \frac{1}{2}|\alpha|^2 P_{\gamma_d}^{\omega_2} \rho_{\gamma_d}^{\omega_2} \otimes \hat{R}_y(\pi) |\downarrow\rangle\langle \downarrow| \hat{R}_y(\pi) + \frac{1}{2}|\beta|^2 P_{\gamma_d}^{\omega_2} \rho_{\gamma_d}^{\omega_2} \otimes |\downarrow\rangle\langle \downarrow|}{P_s + \text{Tr} \left(\frac{1}{2}|\alpha|^2 P_{\gamma_d}^{\omega_2} \rho_{\gamma_d}^{\omega_2} \otimes \hat{R}_y(\pi) |\downarrow\rangle\langle \downarrow| \hat{R}_y(\pi) + \text{Tr} \left(\frac{1}{2}|\beta|^2 P_{\gamma_d}^{\omega_2} \rho_{\gamma_d}^{\omega_2} \otimes |\downarrow\rangle\langle \downarrow|\right)\right)}.
\]

Using Eq. (S16) with \(|\alpha| = |\beta| = 1/\sqrt{2}\), the entanglement fidelity under pure dephasing is

\[
F_{\text{theory}} = \langle \psi_{\text{ideal}} | \rho_{\text{out}}' | \psi_{\text{ideal}} \rangle = \frac{\int_{-\infty}^{\infty} |\Phi(\omega)|^2 |r_1(\omega)|^2 d\omega + \int_{-\infty}^{\infty} |\Phi(\omega)|^2 |\hat{r}_1(\omega)|^2 d\omega + \int_{-\infty}^{\infty} |\Phi(\omega)|^2 |\hat{r}_1(\omega)|^2 d\omega + \int_{-\infty}^{\infty} |\Phi(\omega)|^2 |\hat{r}_1(\omega)|^2 d\omega + P_{\gamma_d}^{\omega_2}}{\int_{-\infty}^{\infty} |\Phi(\omega)|^2 |r_1(\omega)|^2 d\omega + \int_{-\infty}^{\infty} |\Phi(\omega)|^2 |\hat{r}_1(\omega)|^2 d\omega + P_{\gamma_d}^{\omega_2}} \approx 1 - \gamma_d/\Gamma.
\]

4. Spin dephasing

In this section, we investigate how the decoherence of the spin states affects the entanglement fidelity. Specifically we consider the dephasing of the QD spin ground states, due to the presence of an external Overhauser field effectively formed by a neighboring nuclear ensemble. This effect causes a superposition spin qubit to precess on the equatorial plane at a random frequency \(\delta_g\) slower than the QD decay rate, which is modelled by applying a time evolution operator \(\hat{T}(\Delta t) = \exp\left(-i\hat{\delta}_g\hat{S}_z\Delta t\right)\) on the superposition spin state, where \(\hat{S}_z = \hat{\sigma}_z/2\) \([S2]\). In the course of the entangling gate, a \(\pi\)-pulse is applied between two scattering events to ensure the precession of the spin is reversed and thus the spin is eventually refocused. In theory, the superposition qubit starts to precess at \(t_0\) and the \(\pi\)-rotation pulse is applied at \(t_r\). The spin is then refocused and read out at \(t_f\) when \(t_r - t_s = t_s - t_0 = \Delta t\) must be satisfied for the perfect echo condition. In the experiment, a rotation pulse \(\hat{R}_z = \hat{R}_{y,\phi_0}(\pi/2)\) is applied at \(t_r\) to project the spin state onto one of its poles thus preventing further precession.
To understand how spin echo works for the gate, we introduce the spin-echo operator \( \hat{U}_{\text{echo}} = \hat{T}(t_r - t_\pi) \hat{R}_y(\pi) \hat{T}(t_\pi - t_0) \) which transforms the spin states into

\[
\begin{align*}
\hat{U}_{\text{echo}} |\uparrow\rangle &= -e^{-i\delta_y(2t_r - t_\pi - t_0)} |\downarrow\rangle = \lambda_\theta |\downarrow\rangle; \\
\hat{U}_{\text{echo}} |\downarrow\rangle &= e^{i\delta_y(2t_r - t_\pi - t_0)} |\uparrow\rangle = \lambda_\theta |\uparrow\rangle.
\end{align*}
\] (S19)

With Eq. (S19), the output state in Eq. (S2) becomes

\[
|\psi_{\text{out}}\rangle = \left( -\alpha \lambda_\theta r'_1|e\rangle_r + \alpha \lambda_\theta \hat{r}'_1 |e\rangle_r + \beta \lambda_\theta r'_1 |l\rangle_r - \beta \lambda_\theta |l\rangle_r + \ldots \right) / \sqrt{2}.
\] (S20)

Eq. (S20) implies that the phase coherence between \(|e\rangle_r\) and \(|l\rangle_r\) depends on (i) the accumulated phase from spin precession, and (ii) the phase acquired from the early and late single-photon scattering events which is determined by the exact time of scattering occurred within the optical pulse. The former is effectively removed by the echo sequence as \(2t_\pi - t_r - t_0 = 0\), whereas the latter is made equal by interfering the time-bins with a matching time delay \(\tau = 11.8\) ns on the detection path. Since the time-bin qubit is created and measured using the same interferometer setup, by having an equal time delay \(\tau_e = \tau_d = \tau\) for the excitation and detection paths, the interferometer temporally picks out events in which the exact time of scattering is in the same position of the pulse, i.e., \(r_1'(t') = r_1'(t')\) for some time \(t' \in \Phi_1(t)\) within the optical pulse. Therefore, the coherence of the spin-photon Bell state is well-preserved.

5. Incoherent spin-flip error and finite \(T_2^*\)

The next error concerns spin decoherence induced by the red-detuned spin rotation laser and due to finite spin coherence time \(T_2^*\). The former effect has been observed in Refs. [S10, S11] which results in power-dependent spin-flips, thereby destroying the coherence of the spin qubit during spin rotations. Despite its exact origin being not fully resolved, its effect on the spin coherence and the fidelity can be approximated by modelling the spin-flip error by a depolarizing channel \(E_{\text{depol}}\), with the probability of undergoing a random spin-flip \(p\) dependent on the incoherent spin-flip rate \(\kappa\) and the duration of the respective rotation pulse \(T_\pi\). The action of the depolarizing channel on a density matrix \(\rho\) is denoted by \(E_{\text{depol}}(\rho) = (1 - p)\rho + pI/2\), where \(I\) is the identity matrix. As an example, after applying a \(\hat{R}_y(\pi/2)\) pulse on a spin state initialized in \(|\uparrow\rangle\), the spin density matrix transforms according to

\[
E_{\text{depol}}(\hat{R}_y(\pi/2)\rho\hat{R}_y(\pi/2)) = E_{\text{depol}}(\frac{1}{2}(\hat{F}_z\hat{R}_y(\pi/2)\rho\hat{R}_y(\pi/2) + (1 - \hat{F}_z)\rho_-))
\]

\[
= (1 - p_{\pi/2})\left( \frac{1}{2}(\hat{F}_z\hat{R}_y(\pi/2)\rho\hat{R}_y(\pi/2) + (1 - \hat{F}_z)\rho_-) + \frac{p_{\pi/2}}{2}I \right)
\]

\[
= \left[ (1 - p_{\pi/2})(\hat{F}_z \approx \frac{1}{2}) + \frac{1}{2} \right] (1 - p_{\pi/2}) = E_{\pi/2},
\] (S21)

where \(\rho_-\) is the initial spin density matrix and \(\rho_- \equiv |\downarrow\rangle_s \langle \downarrow|_s\). \(E_{\pi/2}\) is the output density matrix. In addition to the incoherent spin flip with a probability \(p_{\pi/2}\) we here include known imperfections of the rotation pulse \(\hat{R}_y(\pi/2)\), which has a fidelity of \(\hat{F}_z\) to coherently rotate the spin to the superposition state \(|\up\rangle_s\) and a probability of \(1 - \hat{F}_z\) to project onto \(|\down\rangle_s\). The fidelity of coherent spin rotation is determined by the limitations of the two-photon Raman scheme, which is dominated by the spin coherence time [S12]:

\[
\hat{F}_z \approx 1 - \frac{2}{\pi^2} \left( \frac{T_{\tau,\pi/2}}{T_2^*} \right)^2,
\] (S22)

for a pulse duration of \(T_{\tau,\pi/2}\).

The probability of introducing a depolarizing error \(p_{\pi/2}\) during a \(\hat{R}_y(\pi/2)\) rotation is estimated by integrating the exponential distribution over the pulse duration for a given incoherent spin-flip rate \(\kappa\):

\[
p_{\pi/2} = \int_0^{T_{\tau,\pi/2}} \kappa e^{-\kappa t} dt = 1 - e^{-\kappa T_{\tau,\pi/2}}.
\] (S23)

The exponential distribution describes the probability of a random spin-flip occurring in a certain time period, where the spin-flip event is assumed not to depend on how much time has passed in the protocol (i.e. it is memory-less).
Similarly, for a $\hat{R}_y(\pi)$ pulse applied on an arbitrary spin state $\rho_s$,

$$E_{\text{depol}}(\hat{R}_y(\pi)\rho_s\hat{R}_y(\pi)) = (1 - p_\pi)\left(\mathcal{F}_\pi\hat{R}_y(\pi)\rho_s\hat{R}_y(\pi) + (1 - \mathcal{F}_\pi)\rho_s\right) + \frac{p_\pi T}{2} \mathcal{I} = \left[\begin{array}{c|c}
(1 - p_\pi)[\mathcal{F}_\pi\rho_1 + (1 - \mathcal{F}_\pi)\rho_3] + \frac{p_\pi T}{2} & (1 - p_\pi)[\mathcal{F}_\pi\rho_3 + (1 - \mathcal{F}_\pi)\rho_2] \\
(1 - p_\pi)[\mathcal{F}_\pi\rho_2 + (1 - \mathcal{F}_\pi)\rho_3] & (1 - p_\pi)[\mathcal{F}_\pi\rho_1 + (1 - \mathcal{F}_\pi)\rho_4] + \frac{p_\pi T}{2}
\end{array}\right] \equiv E_{\pi}, \quad (S24)$$

where the initial spin density matrix is

$$\rho_s \equiv \begin{bmatrix} \rho_1 & \rho_2 \\ \rho_3 & \rho_4 \end{bmatrix}, \quad (S25)$$

and $p_\pi$ is the probability of introducing the depolarizing error during a $\hat{R}_y(\pi)$ rotation found similarly as in Eq. (S23).

Using Eq. (S24) and $\rho_1 = \rho_2 = \rho_3 = 0$, $\rho_4 = 1$, the total $\pi$-rotation pulse fidelity which includes the contribution from both coherent and incoherent spin-flip processes can be estimated to be

$$\mathcal{F}_{\pi,\text{total}} = (1 - p_\pi)\mathcal{F}_\pi + \frac{p_\pi}{2} = 1 + e^{-\kappa T_{\pi,\tau}} \left[\frac{1}{2} - \frac{2}{\pi^2} \left(\frac{T_{\tau,\pi}}{T_2}\right)^2\right] \approx 1 - \frac{1 - \frac{T_{\tau,\pi}}{T_2}}{\frac{2}{\pi^2} \left(\frac{T_{\tau,\pi}}{T_2}\right)^2}. \quad (S26)$$

Using experimental values for the incoherent spin-flip rate $\kappa = 0.021\, \text{ns}^{-1}$ and the spin coherence time $T_2 = 23.2$ ns which are extracted in separate experiments [S12], we then estimate $\mathcal{F}_{\pi,\text{total}} \approx 91.6\%$ for $T_{\tau,\pi} = 7$ ns. For $T_{\tau,\pi} = 21.4$ ns estimated in Ref. [S5], the corresponding $\pi$-rotation fidelity is 91.2% which agrees well with the experiment (91%).

Now we consider the evolution of the spin-photon system during the entangling gate. The protocol begins by preparing a time-bin photonic qubit $|\rho_p\rangle$ and a spin state in $\rho_s$:

$$|\rho_p\otimes\rho_s\rangle = \begin{bmatrix} \alpha^2 & \alpha^* \beta \beta^* \\ \alpha^* \beta & |\beta|^2 \end{bmatrix} \otimes \begin{bmatrix} 0 & 0 \\ 0 & 1 \end{bmatrix}$$

$$\hat{R}_y(\pi/2) \rightarrow (\mathcal{I} \otimes E_{\text{depol}}^s)(|\rho_p\otimes\rho_s\rangle) = \begin{bmatrix} \alpha^2 |E_{\pi/2}^1\rangle & \alpha^* \beta \beta^* |E_{\pi/2}^2\rangle \\ \alpha^* \beta^* |E_{\pi/2}^3\rangle & |\beta|^2 |E_{\pi/2}^4\rangle \end{bmatrix} \frac{1}{\sqrt{2}} \begin{bmatrix} \rho_1 \rho_4 \\ \rho_3 \\ \rho_2 \\ \rho_4 \end{bmatrix}$$

$$\hat{R}_y(\pi) \rightarrow \mathcal{F}_\pi \rightarrow \frac{1}{\sqrt{2}} \begin{bmatrix} \alpha^2 |E_{\pi}^1\rangle & \alpha^* \beta \beta^* |E_{\pi}^2\rangle \\ \alpha^* \beta^* |E_{\pi}^3\rangle & |\beta|^2 |E_{\pi}^4\rangle \end{bmatrix} \frac{1}{\sqrt{2}} \begin{bmatrix} \rho_1 \rho_4 \\ \rho_3 \\ \rho_2 \\ \rho_4 \end{bmatrix}$$

where the basis states that span the spin-photon density matrix are $\{|\uparrow\rangle_r, |\downarrow\rangle_r, |\uparrow\downarrow\rangle_r, |\downarrow\uparrow\rangle_r\}$ conditioned on the detection of a reflected photon, as the events in which photons are transmitted do not contribute to the fidelity. The terms which are of interest are highlighted in red. As an example we evaluate one of the matrix elements $|e \uparrow\rangle_r |e \uparrow\rangle_r$:

$$|\alpha|^2 E_{\pi}^1(\rho_1^1, \rho_1^1) = |\alpha|^2 (1 - p_\pi)[\mathcal{F}_\pi\rho_4^1 + (1 - \mathcal{F}_\pi)\rho_1^1] + \frac{p_\pi}{2} \right) = |\alpha|^2 \left(\frac{1 - p_\pi}{2} |\mathcal{F}_\pi| \beta|^2 + (1 - \mathcal{F}_\pi) |\beta|^2 \right) + \frac{p_\pi}{2}. \quad (S28)$$
For an ideal state of $|\psi_{\text{ideal}}\rangle = (\alpha |e \rangle + \beta |i \rangle)_{r}$ where $|\alpha| = |\beta| = 1/\sqrt{2}$, the entanglement fidelity is given by

$$F_{\text{theory}}^r = \left(\frac{\langle \psi_{\text{ideal}} | \rho_{\text{out}} | \psi_{\text{ideal}} \rangle}{\text{Tr}(\langle \psi_{\text{ideal}} | \psi_{\text{ideal}} \rangle)}\right) = \left|\frac{\alpha^4 E_2^4(\rho_1, \rho_1) + |\beta|^4 E_2^4(\rho_1, \rho_1) - |\alpha|^2 |\beta|^2 r_1^4 E_2^4(\rho_1, \rho_1)}{|\alpha|^4 E_2^4(\rho_1, \rho_1) + |\beta|^4 E_2^4(\rho_1, \rho_1) + |\alpha|^2 |\beta|^2 r_1^4 E_2^4(\rho_1, \rho_1) - |\alpha|^2 |\beta|^2 r_1^4 E_2^4(\rho_1, \rho_1)}\right| \approx 1 - \frac{5\pi \kappa}{4 \Omega - 3\frac{1}{\Omega^2 T_r^2}}.$$ (S29)

for $\kappa \ll \Omega$ where $\Omega$ is the spin-rotation Rabi frequency and $\Omega T_r = \pi$ for a $\pi$-pulse. Using the relevant parameters: $T_r, \pi = 7$ ns, $T_r, \pi/2 = 3.5$ ns, $\kappa = 0.021$ ns$^{-1}$ and $T_r^* = 23.2$ ns, we find $F_{\text{theory}}^r = 82.94\%$ from the analytical form in Eq. (S29) taking $r_1 = 1$ and $\hat{r}_1 = 0$.

6. Spin readout error

The non-ideal spin readout by optical pumping is also considered to be one of the dominant sources of imperfections as it directly influences the spin readout basis. Due to finite optical cyclicity, optically pumping of the main transition can unfavourably result in an opposite outcome by flipping the spin state:

$$\rho_{\text{out}} \xrightarrow{\text{Spin readout}} F_R \rho_{\text{out}} + (1 - F_R) \sigma_x (\pi) \rho_{\text{out}} \sigma_x (\pi).$$ (S30)

where the readout fidelity is estimated to be $F_R = 96.6\%$ [S12]. Using Eqs. (S27) and (S30), the resulting entanglement fidelity under both rotation error and imperfect spin readout is $F_{\kappa, R}^{\text{theory}} = 80.24\%$. From here it is apparent that the dominant infidelity results from incoherent spin flips (17%). To further investigate the influence of this error we plot the entanglement and $\pi$-rotation fidelities as a function of the spin-flip rate $\kappa$ (Fig. S3), which indicates a linear dependence in the perturbative regime where $\kappa \ll \Omega$. Dashed black line shows the corresponding fidelities when $\kappa = 0.021$ ns$^{-1}$. An order of magnitude reduction in $\kappa$ would lead to an improved entanglement fidelity of $F_{\kappa, R}^{\text{theory}} = 93.16\%$.

7. Driving-induced dephasing due to multi-photon scattering

Another source of error originates from the finite multi-photon component of the input pulse, which destroys the QD ground-state spin coherence through successions of photon-scattering events within the pulse. The driving-induced dephasing probability $p_d$ is related to the success probability of scattering $P_{\omega_1} + P_{\omega_2}$ and the mean photon number in the driving pulse $\bar{n}$ via $p_d = 1 - \exp(-\bar{n}(P_{\omega_1} + P_{\omega_2}))$ [S13]. This can be understood as the probability of $\bar{n}$ disjoint successful scattering events. To describe the effect of this error, we adopt a phase-damping model $\mathcal{E}_d$ where

$$\mathcal{E}_d \left( \text{Tr}_p (\hat{S}(\rho_p \otimes \rho_s)) \right) \equiv \left(1 - \frac{p_d}{2}\right) \text{Tr}_p (\hat{S}(\rho_p \otimes \rho_s)) + \frac{p_d}{2} \hat{\sigma}_z \text{Tr}_p (\hat{S}(\rho_p \otimes \rho_s)) \hat{\sigma}_z = \left[ \begin{array}{cc} s_1 & (1 - p_d)s_3 \\ (1 - p_d)s_3 & s_4 \end{array} \right].$$ (S31)
Here $\hat S$ is the scattering matrix acting on the spin-photon density matrix and $s_i$ is the scattering amplitude obtained from $\hat S$. $E_d$ introduces dephasing only to the QD spin state thus the photonic component is traced out before applying the phase-damping channel. Now we follow the same approach in Sec. II D 5 and consider propagation of the dephasing error in the protocol:

$$\rho_p \otimes \rho_s \xrightarrow{R_y(\pi/2)} \frac{1}{2} \begin{bmatrix} 1 & 1 \\ 1 & 1 \end{bmatrix} \alpha^* \beta' \rho$$

$$\frac{1}{2} \begin{bmatrix} |\alpha|^2 \beta E_d \left( \begin{array}{c} r_1 E_d \left( \begin{array}{c} r_1^* \r_1^* \\ r_1^* \r_1^* \end{array} \right) \alpha^* \beta E_d \left( \begin{array}{c} r_1^* \r_1^* \\ r_1^* \r_1^* \end{array} \right) \end{array} \right) \\
\alpha^* \beta E_d \left( \begin{array}{c} r_1 E_d \left( \begin{array}{c} r_1^* \r_1^* \\ r_1^* \r_1^* \end{array} \right) \end{array} \right) \end{bmatrix} = \frac{1}{2} \begin{bmatrix} \alpha^* \beta E_d \left( \begin{array}{c} r_1 E_d \left( \begin{array}{c} r_1^* \r_1^* \\ r_1^* \r_1^* \end{array} \right) \end{array} \right) \\
\alpha^* \beta E_d \left( \begin{array}{c} r_1 E_d \left( \begin{array}{c} r_1^* \r_1^* \\ r_1^* \r_1^* \end{array} \right) \end{array} \right) \end{bmatrix}.$$

Similarly, the entanglement fidelity under the driving-induced dephasing is found to be

$$F_{\text{theory}} = \frac{1}{2} |r_1|^2 \left[ 1 + e^{-2\tilde{n}\left(P_{\omega_1} + P_{\omega_2}\right)} \right] \frac{1}{2} \left[ 1 + e^{-2\tilde{n}\left(P_{\omega_1} + P_{\omega_2}\right)} \right] \approx 1 - \tilde{n}\left(P_{\omega_1} + P_{\omega_2}\right),$$

where the probability of successful scattering at zero detuning is given by Eq. (S4):

$$P_{\omega_1} + P_{\omega_2} = |r_1|^2 + |t_1|^2 + |r_2|^2 + |t_2|^2 \approx 1 - 2 \left( \frac{\gamma_1 + \gamma_2}{\Gamma} \right) \left( 1 - \frac{1}{C + 1} - \frac{\gamma_1}{\Gamma} \right).$$

To estimate the infidelity in the experiment, we first extract the average number of photons in the pulse $\tilde{n} = S_n \cdot T_p \cdot \Gamma = 0.0496 \times 0.2976 \times 2 \text{ ns} \times 2.48 \text{ ns}^{-1} \approx 0.0732$ where the relevant parameters are obtained from saturation measurements (Sec. IV). Given that optically cyclicity $C = 14.7$ and radiative loss rate $\gamma_1 = \gamma_2 = 0.05 \text{ ns}^{-1}$, the experimental infidelity is estimated using the exact form in Eq. (S33) to be $1 - F_{\tilde{n}}^\text{theory} = 6.34\%$.

### E. Overall fidelity and gate efficiency

Assuming perfect manipulation of the hole spin state, the fidelity of the entangling gate is expressed by:

$$F_{\text{theory}} = 1 - \left( \frac{\gamma_d}{\Gamma} + \frac{\Gamma^2}{4\Delta_h^2} \right),$$

which is estimated to be 96.2% with $\Gamma = 2.48 \text{ ns}^{-1}$, $\Delta_h = 2\pi \times 7.3 \text{ GHz}$ [S5] and $\gamma_d = 0.092 \text{ ns}^{-1}$ (fitted in Sec. III). This predominantly reflects the infidelity from phonon-induced pure dephasing $1 - F_{\gamma_d}^\text{theory}$ as the off-resonant reflection error $\Gamma^2/\Delta_h^2$ is comparably small. Together with the incoherent spin-flip and the readout errors discussed in Sec. II D, we estimate the overall entanglement fidelity $F_{\text{total}}^\text{theory}$ using

$$F_{\text{total}}^\text{theory} \approx F_{\gamma_d}^\text{theory} \times F_{\alpha,R}^\text{theory} \times F_{\tilde{n}}^\text{theory} = 72.3\%,$$

which agrees with the experimentally achieved value $(74.3 \pm 2.3)\%$ including error margins. The remaining infidelity is likely a combination of imperfect spin initialization $(1.4\%)$ [S11], and non-Gaussian pulse shaping of the input photon which are not included in the theory. Using $C = 14.7$, optical pulse duration $T_p = 1/2\sigma_o = 2 \text{ ns}$, $\gamma_1 = 0.05 \text{ ns}^{-1}$
(estimated), $\sigma_c = 0.3$ ns$^{-1}$ (estimated), the entangling gate efficiency is represented by the success probability where

$$P_s = \frac{1}{2} \left[ \int_{-\infty}^{\infty} |\Phi(\omega)|^2 |r_1(\omega)|^2 d\omega + \int_{-\infty}^{\infty} |\Phi(\omega)|^2 |\tilde{r}_1(\omega)|^2 d\omega + P_{\text{inc}}^2 \right]$$

$$\approx \frac{1}{2} \left[ 1 - \frac{4\sigma_c^2}{\Gamma^2} - \frac{4\sigma_c^2}{\Gamma^2} - \frac{2}{C+1} \frac{4\gamma_d}{\Gamma} \left( 1 - \frac{1}{C+1} \right) - \frac{2\gamma_1}{\Gamma} + \left( \frac{2\gamma_d}{\Gamma} + \frac{\Gamma^2}{4\Delta_h^2} \right) \left( 1 - \frac{1}{C+1} - \gamma_1 \frac{2}{\Gamma} \right) \right]$$

$$\approx \frac{1}{2} \left[ 1 - \frac{4\sigma_c^2}{\Gamma^2} - \frac{4\sigma_c^2}{\Gamma^2} - \frac{2}{C+1} \frac{4\gamma_d}{\Gamma} \left( 1 - \frac{1}{C+1} \right) - \frac{2\gamma_1}{\Gamma} + \frac{\Gamma^2}{4\Delta_h^2} \left( 1 - \frac{1}{C+1} - \gamma_1 \frac{2}{\Gamma} \right) \right], \quad (S37)$$

and is estimated to be 33.3%.

### III. PHOTON VISIBILITY

Here we derive an analytical form of the visibility as a function of the QD pure dephasing rate. In the experiment, a time-bin encoded qubit (a weak coherent state) is scattered by a QD spin embedded in a two-sided photonic-crystal waveguide, and is subsequently measured by an asymmetric Mach-Zehnder interferometer with equal time delay as the qubit. The visibility is therefore a measure of the temporal overlap between the time-bins of the scattered pulses. To model this, we consider the scattering of the time-bin photon with the QD and project the output state onto the phasor X-bases. The initial state of the system is expressed as

$$|\text{in}\rangle = |e\rangle + |l\rangle/\sqrt{2} \otimes |\theta\rangle.$$  

Here we have neglected the multi-photon components from the coherent state since we are interested in the effect of pure dephasing. For a complete modelling of the photon visibility, however, one should include the effect of multi-photon scattering and inelastic contributions [S14]. With Eq. (S4) the output state becomes

$$|\text{out}\rangle = \frac{1}{\sqrt{2}} \left[ r_1 |e\rangle_{r} + |l\rangle_{r} \right] + \frac{1}{\sqrt{2}} \left[ t_1 |e\rangle_t + |l\rangle_t \right] \otimes |\phi\rangle + \frac{1}{\sqrt{2}} \left[ r_2 |e\rangle_r + |l\rangle_r \right] + t_2 |e\rangle_t + |l\rangle_t \right] \otimes |\phi\rangle, \quad (S38)$$

where the superscript prime (‘’) represents a scattered photon of frequency $\omega_2 \neq \omega_1$ and the subscript “r” (“t”) indicates a reflected (transmitted) photon. We then seek the photonic density matrix by tracing out the spin degree of freedom, the transmitted photons as well as the wrong frequency state $\omega_2$. For ease of computation the scattering coefficients are replaced by $C_i$ where $i$ refers to the time-bin, thus

$$|\text{out}\rangle_p = \frac{\text{Tr}_{s,t,w}(|\text{out}\rangle \langle \text{out}|)}{\text{Tr}(\langle \text{out}| \langle \text{out}|)} = |C_e|^2 |e\rangle \langle e| + |C_l|^2 |l\rangle \langle l| + C_e C_l^* |e\rangle \langle l| + C_l C_e^* |l\rangle \langle e|. \quad (S39)$$

Now (S39) is used to evaluate the middle-bin intensity in detector D2(D1):

$$I_{D2/D1} = \int \text{Tr} \left[ \left( \hat{a}_e \pm e^{i\theta_p} \hat{a}_l \right) \langle \text{out}\rangle_p \langle \text{out}| \right] \left( \hat{a}_e \pm e^{-i\theta_p} \hat{a}_l \right) / \sqrt{2} dt, \quad (S40)$$

where the output photon state is projected onto the superposition state $\hat{a}_e(t) \pm e^{i\theta_p} \hat{a}_l(t)$ which is equivalent to adding a phase shifter on the long path of the excitation interferometer and interfering both bins. Setting $\theta_p = 0$ implies projecting the output state into the $p_{\pm} = |\pm X\rangle_p \langle \pm X|$ bases as described in the main text. The projected state is then traced out in both the early and late time bases. The photon visibility is defined as the normalized contrast of the middle-bin intensity when $\theta_p = 0$:

$$V_p = \frac{I_{D2} - I_{D1}}{I_{D2} + I_{D1}} = \frac{\int \text{Tr} \left[ \hat{a}_e(|\text{out}\rangle_p \langle \text{out}| \langle \text{out}| \right) \hat{a}_l + \hat{a}_l(|\text{out}\rangle_p \langle \text{out}| \langle \text{out}| \right) \hat{a}_e^\dagger \right] dt}{\int \text{Tr} \left[ \hat{a}_e(|\text{out}\rangle_p \langle \text{out}| \langle \text{out}| \right) \hat{a}_l + \hat{a}_l(|\text{out}\rangle_p \langle \text{out}| \langle \text{out}| \right) \hat{a}_e^\dagger \right] dt}. \quad (S41)$$

To further simplify the above expression, we consider the scattering events of the early and late bins to be identical, i.e., with the same scattering coefficient $C_e = C_l = r_1$, as justified in Sec. II D 4. Therefore, under this assumption the photon visibility becomes unity in the single-photon regime.
A. Visibility expression including pure dephasing

Following from the discussion in Sec. II D 3, we can now take into account the effect of phonon-induced pure dephasing. In essence, the resulting spin-photon density matrix is the sum of coherent and incoherent parts as described by Eqs. (S15) and (S16). The advantage of the formalism in (S15) is that its effect can be straightforwardly included in Eq. (S39). Accordingly, the new photonic density matrix becomes

$$\langle \text{out}' \rangle_p \langle \text{out}' |_p \approx \langle \text{out} \rangle_p \langle \text{out} |_p + \frac{1}{2} P_{\omega_1}^{\text{d}} \rho_{\text{d},e}^{\text{c}} |0 \rangle_l \langle 0 |_l + \frac{1}{2} P_{\omega_1}^{\text{d}} \rho_{\text{d},l}^{\text{c}} |0 \rangle_e \langle 0 |_e, \quad (S42)$$

where the last two terms correspond to dephasing occurring during the single-photon scattering of either the early or late time-bin. The effect of pure dephasing on the multi-photon component is not considered due to its polynomial dependence on the mean photon number per pulse $\bar{n}$, which is negligible as $\bar{n} \ll 1$. Note that the incoherent photon does not interfere with other photons since $\text{Tr} \left( \hat{a}_e \rho_{\text{d},e}^{\text{c}} |l \rangle \langle l |_l \right) \rho_{\text{d},e}^{\text{c}} = \text{Tr} \left( \hat{a}_e \rho_{\text{d},e}^{\text{c}} \right) \times \text{Tr} (|l \rangle \langle l |_l ) = 0$. This means only the total intensity is affected and Eq. (S41) can be simplified as

$$V_p = \int_{-\infty}^{\infty} |\Phi_1(\omega)|^2 |r_1(\omega)|^2 d\omega \int_{-\infty}^{\infty} |\Phi_1(\omega)|^2 |r_1(\omega)|^2 d\omega + P_{\omega_1}^{\text{d}} \approx 1 - \frac{2 \gamma_d}{\Gamma}. \quad (S43)$$

This indicates that in the single-photon scattering limit where $\bar{n} \approx 0$, the y-intercept of the visibility curve in Fig. 2b is given by the pure dephasing rate per QD total decay rate $\Gamma$. Here $\Gamma = 2.48 \text{ ns}^{-1}$ is measured in Ref. [S5]. A linear fit of the data gives a y-intercept of $V_p(\bar{n} = 0) = 0.926 \pm 0.003$ implying $\gamma_d \approx (0.092 \pm 0.004) \text{ ns}^{-1}$.

IV. MEAN PHOTON NUMBER PER PULSE

FIG. S4. Saturation measurement to calibrate the mean photon flux. (a) Time-resolved histogram of the measurement sequence. A 2 ns pulse gets reflected from a QD prepared in $|\uparrow\rangle$ via optical spin pumping followed by a $\pi$-rotation pulse. The reflected signal is time-gated (green shaded region) and recorded for each input power. Peaks at around 100 and 215 ns are laser scatter from the time-bin interferometer and the optical breadboard respectively. The spin readout at 300 ns maintains the same duty cycle as the entangling gate experiment and does not affect the gated counts. (b) Gated fluorescence in the reflection as a function of the input pulse power. Blue (red) circles are summed counts over a time window of 3 ns, when the QD spin is prepared in $|\uparrow\rangle$ ($|\downarrow\rangle$). Fitted (black solid line) using Eq. (S44). Around 0.075 nW is used for a single pulse in the entangling gate experiment.

Apart from measuring the photon visibility, another approach to probe the single-photon nature of the scattering process is through QD saturation measurement, in which the QD response is observed by scanning the power of the input qubit laser. From fitting the scattered signal, the mean photon number per pulse $\bar{n}$ can be extracted, where $\bar{n} \ll 1$ indicates the scattering occurs in the single-photon regime. To mimic the entangling gate experiment, we prepare a single pulse of 2 ns duration and scatter on a QD spin initialized in either $|\uparrow\rangle$ or $|\downarrow\rangle$. Due to the QD spin-dependent reflectivity, the input photon which is resonant with the QD transition $|\uparrow\rangle \rightarrow |\downarrow\uparrow\rangle$ is coherently reflected. By time-gating on the reflected signal (Fig. S4a; green shaded region) and increasing the input power, the
QD prepared in $|⇑\rangle$ becomes saturated (Fig. S4b). The intensity in the reflected signal is fitted assuming a two-level system between $|⇑\rangle \rightarrow |↑⇓⇑\rangle$:

$$I_r \propto \frac{(1 + 2\gamma_d/\Gamma)\Omega_1^2}{(\Gamma + 2\gamma_d)^2/4 + \delta^2 + 2(1 + 2\gamma_d/\Gamma)\Omega_1^2} \equiv \frac{b_1b_1P}{1 + b_2b_1P},$$

(S44)

where $\Omega_1$ is the Rabi frequency driving the transition $|⇑\rangle \rightarrow |↑⇓⇑\rangle$, $\delta$ is the probe laser detuning, $b_1, b_2$ and $b_3$ are free fit parameters. In particular, $b_1$ is the calibrating parameter that associates the Rabi frequency to the input power $P$ via $\Omega_1 = \sqrt{b_1P}$ which includes optical losses on the excitation path.

Eq. (S44) holds when $\Gamma \gg \kappa_g$ and $T_p \gg \Gamma^{-1}$ where $\kappa_g$ is the effective spin-flip rate between the hole ground states and $T_p$ is the qubit pulse duration. The first condition implies that the main transition is eventually saturated as the QD decays faster than the spin can recycle, thus $|⇓\rangle$ effectively becomes dark. This is generally true since $\kappa_g$ is typically on the order of $10^{-7}$ ns$^{-1}$ at the plateau center voltage [S15], which is lower than $\Gamma = 2.48$ ns$^{-1}$. The second condition ensures that the QD decays back to $|⇑\rangle$ before the next scattering event within the pulse. When the driving pulse is sufficiently long, i.e., $T_p = 2$ ns $> \Gamma^{-1} = 0.4$ ns with increasing power, the QD saturates similarly as when being driven by a weak continuous-wave laser. In addition, a finite optical cyclicity leads to a resonant spin-flip into the dark state $|⇓\rangle$ reflecting a photon of frequency $\omega_2 \neq \omega_1$ which is filtered out, thus only reducing the total intensity included in $b_2$ and not affecting the scaling of the relevant parameters in Eq. (S44).

Fitting the data in Fig. S4b, we extract $b_1 = 0.64$ ns$^{-2}$/nW, $b_2 = 1.03$ and $b_3 = 7.7 \times 10^4$. The mean photon number per QD lifetime or mean photon flux is defined to be $\bar{n}_F = S n_c$ where $n_c \equiv (1 + 2\gamma_d/\Gamma)/\beta^2$ is the critical photon flux leading to an excited state population of 1/4 [S16]. Therefore, with $\gamma_d = 0.092$ ns$^{-1}$ (see Sec. III), a beta-factor of $\beta = 0.95$ (estimated), and $S = b_1 \times b_2 \times \bar{F} \approx 0.0496$ is the saturation parameter for an input power of $P = 0.075$ nW used for a single pulse in the entangling experiment, we estimate $n_F \approx 0.0148$ and the average number of photons in the scattering pulse $\bar{n} = n_F T_p \Gamma = 0.073 \ll 1$. A scaling factor between $\bar{n}$ and the input power $\bar{S}_n = 10^{-2} b_1 b_2 n_c T_p \Gamma / 2 \approx 0.005$/nW (assuming transmission of $10^{-2}$ with a neutral-density filter) is also obtained and subsequently adopted in Fig. 2b.

V. CONCURRENCE ESTIMATE

Another measure frequently used in the literature to quantify entanglement is the concurrence $C$, which for a non-separate bipartite system $C > 0$ and reaches unity for a maximally entangled state. Here we estimate a lower bound for the concurrence using the raw and background-corrected coincidences recorded in the experiment (shown in Figs. 3b-c). The concurrence is given by

$$C = \max(0, \sqrt{\lambda_0} - \sum_{i=1}^{N} \sqrt{\lambda_i}),$$

(S45)

where $\lambda_i$ are eigenvalues of the matrix $\rho_{\text{meas}}(\hat{\sigma}_y^{(p)} \otimes \hat{\sigma}_y^{(s)})\rho_{\text{meas}}(\hat{\sigma}_y^{(p)} \otimes \hat{\sigma}_y^{(s)})^\dagger$, and $\rho_{\text{meas}}$ is the normalized measured density matrix of the spin-photon state assuming negligible off-diagonal entries [S17]

$$\rho_{\text{meas}} = \begin{bmatrix}
\rho_{↑⇑,↑⇑} & 0 & 0 & 0 \\
0 & \rho_{⇑⇓,⇑⇓} & \rho_{⇑⇑,⇑⇓} & 0 \\
0 & \rho_{⇑⇓,⇑⇑} & \rho_{⇑⇑,⇑⇑} & 0 \\
0 & 0 & 0 & \rho_{⇓⇓,⇓⇓}
\end{bmatrix},$$

(S46)

which shares the same form as Eq. (S27). Using the coincidence counts in Figs. 3b-c and Eq. (S45), we find a raw (background-corrected) concurrence of $C \geq 0.345 \pm 0.012$ (0.495 $\pm 0.105$) with error bounds obtained by Monte Carlo simulations.


