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Entangling a Hole Spin with a Time-Bin Photon: A Waveguide Approach for Quantum Dot Sources of Multiphoton Entanglement

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Deterministic sources of multiphoton entanglement are highly attractive for quantum information processing but are challenging to realize experimentally. In this Letter, we demonstrate a route toward a scalable source of time-bin encoded Greenberger-Horne-Zeilinger and linear cluster states from a solid-state quantum dot embedded in a nanophotonic crystal waveguide. By utilizing a self-stabilizing double-pass interferometer, we measure a spin-photon Bell state with (67.8 ± 0.4)% fidelity and devise steps for significant further improvements. By employing strict resonant excitation, we demonstrate a photon indistinguishability of (95.7 ± 0.8)% which is conducive to fusion of multiple cluster states for scaling up the technology and producing more general graph states.

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A near-deterministic source of multiphoton entanglement will enable significant advancements in quantum information processing such as one-way quantum computing consuming few-photon Greenberger-Horne-Zeilinger (GHZ) states [1,2] or one-way photonic repeaters relying on large photonic graph states [3,4]. Until now, spontaneous parametric down-conversion has been the leading source of entangled photons and has enabled demonstrations of 12 entangled photons [5]. However, this approach is based on probabilistic photon sources and is thus challenging to scale up. An alternative approach proposed by Lindner and Rudolph exploits a single solid-state quantum emitter as an on-demand entanglement source [6]. In their proposal, a semiconductor quantum dot (QD) containing a single spin is repeatedly excited to create a linear cluster state of polarization encoded photons. Despite its elegance, this approach has several experimental drawbacks. Specifically, natural spin precession is employed in a weak external magnetic field for which the QD spin is poorly decoupled from the nuclear spin bath [7–9]. Additionally, the need to collect two orthogonally polarized modes is often experimentally incompatible with the resonant excitation required for high photon indistinguishability [10]. These challenges were partly combated in the landmark experiment of Ref. [11], where the use of dark-exciton states and nonresonant excitation enabled the explicit demonstration of three-qubit entanglement and extrapolation to five qubits. Nonetheless, the further scalability of multiphoton entanglement sources has been an open question so far only addressed theoretically [12,13].

A key feature is the generation of high-fidelity multiphoton entanglement while preserving photon indistinguishability—a crucial requirement for fusion-based photonic quantum computing [1,14].

In this Letter, we perform the first demonstration of QD spin-photon entanglement using a time-bin protocol capable of generating multiphoton GHZ and linear cluster states. This protocol has been used to demonstrate spin-photon entanglement from nitrogen-vacancy (NV) centers [15–17]; the first steps using a micropillar QD source were implemented in Ref. [18] without proving entanglement, and a detailed theoretical treatment was recently published [12,13]. Contrary to the Lindner–Rudolph protocol, we apply strong magnetic fields which permit the use of spin-echo pulses and render the protocol insensitive to spin dephasing [12]. Additionally, a single optical transition is excited resonantly and emits highly indistinguishable photons into a single-mode photonic crystal waveguide (PCW) with near-unity internal collection efficiency [19,20]. Here, we focus on measuring spin-photon Bell states using a novel double-pass interferometer and determine a fidelity of 67.8%. This fidelity is predominantly limited by the quality of the spin rotations, which can be substantially improved in future experiments [21].

The entanglement protocol builds upon our recent work on uniting optical cycling transitions with optical spin control [22] as summarized in Fig. 1(a). A positively charged InAs QD in an in-plane (Voigt geometry) magnetic field \( B_z = 2 \, \text{T} \) gives rise to a pair of Zeeman-split heavy-hole ground states \(|\uparrow\rangle \text{ and } |\downarrow\rangle \), which are used as a spin...
qubit [23], and a pair of Zeeman-split trions $|↑↑↓\rangle$ and $|↓↑↓\rangle$ enabling photon generation. The four linear dipoles are driven by a red-detuned Raman laser to rotate the spin qubit [21,22] while the PCW [Fig. 1(b)] selectively enhances the optical transitions resulting in the optical cyclicity $C = \gamma_c/\gamma_s = 14.7 \pm 0.2$ [22], which is otherwise unity in a homogeneous environment or a cylindrically symmetric cavity. Spin-photon entanglement is then generated according to the protocol in Fig. 1(c). Optical pumping initializes $|\downarrow\rangle$, and a $\hat{R}_y(\pi/2)$ rotation around the y axis prepares the superposition state $(|\downarrow\rangle + |\uparrow\rangle)/\sqrt{2}$. The QD is then subjected to a y-polarized optical $\pi$ pulse resonant with $|\uparrow\rangle \leftrightarrow |\uparrow\downarrow\rangle$. As the other y-polarized transition is detuned by $\Delta_0 = 2\pi \times 17$ GHz, trion excitation and thus photon emission is conditioned on $|\uparrow\rangle$. The enhanced cyclicity ensures photon emission via the spin-preserving $|↑↑↓\rangle \rightarrow |↑\rangle$ transition with probability $C/(C+1) \approx 94\%$. Thus, an early excitation entangles $|\uparrow\rangle$ with an early photon $|e\rangle$. The spin states are then swapped by a $\pi$ rotation before applying a late excitation pulse resulting in the emission of a late photon $|l\rangle$. The resulting state is the spin-photon Bell state

$$|\psi_{\text{Bell}}\rangle = (e^{i\phi_e}|\uparrow, l\rangle - |\downarrow, e\rangle)/\sqrt{2}, \quad (1)$$

where the emission time of the single photon is maximally entangled with the hole spin, and $\phi_e$ is the phase difference between the two excitation pulses. By working in the rotating frame of the Raman laser, the spin does not precess and only rotates when we actively apply a Raman pulse in contrast to Refs. [6,11].

The entangled state is generated and analyzed using the setup in Fig. 1(d). The QD is held at 4 K in a closed cycle cryostat and is subjected to lasers propagating from free space. The early and late excitation pulses are generated by injecting a single pulse into the excitation pass [dashed line in Fig. 1(d)] of a time-bin interferometer (TBI) which sets the phase $\phi_e$ and the time delay $T_{\text{inf}} = 11.8$ ns between the early and late pulses. $T_{\text{inf}}$ is sufficient for performing photon emission (400 ps lifetime) and a $\pi$ rotation (7 ns). The QD emits photons into the guided PCW mode, which is coupled to a single-mode fiber via a grating coupler [24]. From here the photon stream enters the detection pass of the TBI [orange line in Fig. 1(d)] where a pair of 3 GHz FWHM etalon filters are used to reject the Raman laser scatter and the QD phonon sideband [25]. Next, a polarizing beamsplitter PBS$_1$ transmits or reflects with equal probability. This passive routing leads to three detection windows [see Fig. 1(e)] which herald the photonic measurement basis. An early (late) detection corresponds to an early (late) photon propagating through the short (long) TBI arm and thus constitutes a Z-basis measurement. By contrast, the middle window represents the time-bin photon interfering with itself on the 50/50 beamsplitter BS$_1$.  

FIG. 1. Time-bin entanglement protocol. (a) Energy level diagram of a positively charged QD in a Voigt magnetic field. The $|\uparrow\rangle \leftrightarrow |\uparrow \downarrow \rangle$ transition is used to emit photonic qubits and to perform spin initialization and readout via optical pumping. The trion decay paths have rates $\gamma_c$ and $\gamma_s$. $\Omega_c$ is the spin Rabi frequency, and $\Delta_0$ is the sum of the Zeeman splittings. (b) Scanning electron micrograph of the PCW with QD dipoles overlaid. (c) Experimental pulse sequence consisting of pumping pulses (red squares), rotation pulses (purple squares), and fast optical $\pi$ pulses (red pulses). The spin-photon state is indicated on top with $|\varnothing\rangle$ denoting a photon vacuum and $|e\rangle(|l\rangle)$ denoting an early (late) photon. (d) Experimental setup. The PCW embedded QD is subjected to lasers propagating from free space. A double-pass TBI defines the excitation pulses (dashed red line) and interferes the emitted photons (solid orange line) resulting in mutual phase stability. A polarizer with angle $\theta_{\text{pol}}$ determines the photonic readout basis. $\lambda/2(\lambda/4)$ denotes half (quarter)-wave plates. (e) Fluorescence histogram resulting from Bell state generation summed over both detectors including a 50 ns spin readout detection window (shaded area). The inset shows a magnified view of the early (e), middle (m), and late (l) detection windows (2 ns each) which herald the measurement basis of the photonic qubit. The peaks at 120 ns are optical reflections inside the TBI.
A photon click on detector $D_1$ or $D_2$ thus projects the photon onto $|\phi_\ell\rangle = (|e\rangle \pm e^{i\phi_e}|l\rangle)/\sqrt{2}$ where $\phi_d$ is the phase of the detection pass. Since excitation and detection passes use the same interferometer the phases $\phi_d$ and $\phi_e$ are mutually stable [26] giving a stable detection pattern without the need for active stabilization [16,27].

We now proceed to quantify the spin-photon entanglement using the approach of Ref. [28] which is scalable to future, large $N$-qubit GHZ states by only requiring $N+1$ measurement settings for an exact fidelity estimate. Adapting the technique to a Bell state gives

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

where $\hat{P}_z = |\uparrow, l\rangle \langle \uparrow, l| + |\downarrow, e\rangle \langle \downarrow, e|$ measures the classical correlations and $\hat{M}_x = \hat{\sigma}^{(s)} \otimes \hat{\sigma}^{(p)}$ and $\hat{M}_y = \hat{\sigma}^{(p)} \otimes \hat{\sigma}^{(s)}$ measure both qubits along $X$ and $Y$, respectively. $\hat{\sigma}$ denotes single qubit Pauli matrices, $s(p)$ superscripts denote a spin(photonic) qubit, and we designate the logical qubits $|0\rangle = |\uparrow\rangle, |l\rangle$ and $|1\rangle = |\downarrow\rangle, |e\rangle$. We post-select on measuring photons in both the photonic and spin readout windows and achieve a 124 Hz coincidence rate when repeating the experiment at a 1.65 MHz repetition rate. As the spin readout can only detect $|\uparrow\rangle$ we apply a $\hat{R}_i$ rotation prior to readout [Fig. 1(c)] fulfilling $\hat{R}_i |s\rangle = |\uparrow\rangle$ to realize the desired spin projector $|s\rangle\langle s|$. Figure 2(a) shows the results of a ZZ-basis measurement. Projection on $|e\rangle$ and $|l\rangle$ is given by the photon detection time (both detectors are treated equally), and the $\hat{R}_i$ pulse is toggled between a 0 and a $\pi$ rotation to realize projections onto $|\uparrow\rangle$ and $|\downarrow\rangle$, respectively. Normalizing across all four projections yields $\langle \hat{P}_z \rangle = (89.3 \pm 0.4)\%$. The imbalance of $|\uparrow\rangle |l\rangle$ and $|\downarrow\rangle |e\rangle$ is primarily a consequence of the imperfect $\hat{R}_i$ spin rotation (discussed later) which reduces the probability of measuring $|\downarrow\rangle$.

In order to measure time-bin encoded photons in the equatorial plane of the Bloch sphere, we add a controllable phase difference between the early and late excitation pulses: The pulses are combined on PBS$_1$ [Fig. 1(d)], converted to opposite circular polarizations with a $\lambda/4$ plate, and projected onto the transmission axis of a rotatable polarizer, which adds $2\theta_{\text{pol}}$ to the phase $\phi_e$. The effect of $\theta_{\text{pol}}$ is evident from the gray data in Fig. 2(b) where classical excitation pulses are reintercepted into the detection pass. This reveals a full oscillation in the detector contrast with near-perfect visibility. The angle $\theta_{\text{pol}} = \theta_0$ gives bunching on detector $D_1$ and corresponds to the condition $\phi_d = \phi_e$, $\theta_0$ depends on the specific TBI alignment but is stable on a week-long timescale. We then run the entanglement protocol, project the QD spin onto $|\pm X\rangle_s = (|\uparrow\rangle \pm |\downarrow\rangle)/\sqrt{2}$ for $\hat{R}_i = \hat{R}_i (\pm \pi/2)$, and measure the spin state dependent contrast; see Fig. 2(b). Crucially, the two fringes are perfectly in-phase and out-of-phase with the classical TBI response. In summary, measuring the photon in the $X$ basis corresponds to setting $\theta_{\text{pol}} = \theta_0$ and assigning the photon states $|+X\rangle_p$ and $|-X\rangle_p$ to a middle window detection on $D_1$ and $D_2$, respectively. Figure 2(c) shows the outcomes of an XX-basis measurement. The outcomes are normalized as before, and the contrast between the positive and negative eigenstates of $\hat{M}_x$ results in $\langle \hat{M}_x \rangle = (-42.3 \pm 1.1)\%$. The $\hat{M}_y$ measurement is similarly realized by setting $\theta_{\text{pol}} = \theta_0 + \pi/4$ and $\hat{R}_i = \hat{R}_i (\pm \pi/4)$ leading to $\langle \hat{M}_y \rangle = (42.1 \pm 1.1)\%$. By applying Eq. (2) we arrive at the final estimate $\mathcal{F}_{\text{Bell}} = (65.7 \pm 0.4)\%$ which exceeds the classical threshold of 50% by 39 standard deviations using only 6 min of acquisition and no corrections. We note that some of the recorded spin-photon coincidences are due to uncorrelated laser leakage.
However, this contribution is minor, and correcting for the background (as in Ref. [17]) leads to a corrected fidelity of \( F_{\text{corr}} = (67.8 \pm 0.4)\% \). NV centers [16,17] have produced similar quality Bell states but with greater reliance on background subtraction. In the interest of understanding the relevant error mechanisms we perform a Monte Carlo simulation of the experiment including all errors and backgrounds (Supplemental Material [29]). This yields the fidelity \( F_{\text{Bell}} = 67.8\% \) (to be compared against \( F_{\text{Bell}} \)) and the detection pattern in Fig. 2 which is in good agreement with the experimental values and supports our error analysis.

We now highlight two of the errors limiting \( F_{\text{Bell}} \). The dominant error is the quality of Raman pulses used for \( X \) and \( Y \) rotations of the hole spin. By measuring the dampening of Rabi oscillations between \( |\uparrow\rangle \) and \( |\downarrow\rangle \) we extract a \( \pi \)-pulse fidelity of \( F_x = 88.5\% \) which is predominantly limited by incoherent spin-flips between the two spin states (Supplemental Material [29]). Naturally, the time-bin protocol relies on highly coherent spin control, and \( F_x = 88.5\% \) alone limits the Bell state fidelity to 77% according to Monte Carlo simulations. Thus, we may attribute \( \sim 70\% \) of the measured infidelity to this mechanism. Fortunately, a recent scheme using electron spins and nuclear spin cooling [21] demonstrated \( F_x = 98.8\% \) and could readily be implemented in the experimental protocol. This would increase the achievable fidelity to \( F_{\text{Bell}} = 97.3\% \) (neglecting other errors).

A second relevant error is the single-photon purity and indistinguishability of the emitted photons. The latter error reduces the measurement contrast in the \( XX \) and \( YY \) bases [12] and is additionally relevant for combining multiple smaller cluster states through entanglement fusion [2]. To accurately characterize the time-bin encoded photon, we retain the magnetic field and minimally modify the pulse sequence to emit two separable single photons [Fig. 3(a)].

By using the TBI, we simultaneously measure the \( g^{(2)} \) intensity autocorrelation and Hong–Ou–Mandel (HOM) visibility by letting the detection time herald the experiment. Figure 3(b) shows the delays between photons recorded in either the early or late detection windows and constitutes two sets of \( g^{(2)} \) measurements. A slight bunching is observed for short delays owing to nondeterministic initialization of the hole charge state. Normalizing \( g^{(2)} \) at long delays and averaging over early and late gives \( g^{(2)}(0) = (4.7 \pm 0.6)\% \) from which 1.1% may be attributed to excitation laser scatter. The remaining contribution is likely a result of multiphoton emission owing to the fast, Purcell enhanced decay rate \( \gamma_0 = (\gamma_x + \gamma_y) = 2.54 \text{ ns}^{-1} \) and the FWHM duration of the \( \pi \) pulse \( T_{\text{opt}} = 35 \text{ ps} \). \( T_{\text{opt}} \) represents a trade-off [12,13] between multiphoton emission (minimized for \( T_{\text{opt}} \ll \gamma_0^{-1} \) and unwanted excitation of \( |\downarrow\rangle \leftrightarrow |\uparrow\rangle \) (minimized for \( T_{\text{opt}} \gg \Delta_0^{-1} \)). A larger magnetic field will increase \( \Delta_0 \) and permit a shorter \( T_{\text{opt}} \) and thus reduced \( g^{(2)}(0) \). Figure 3(c) shows the delay between two photons recorded within the same experimental repetition when at least one photon was measured in the middle window. Following Ref. [43], this estimates a raw indistinguishability of \( \Phi_{\text{raw}} = 1 – 2N_2/(N_1 + N_3) = (86.5 \pm 0.6)\% \) [integration windows given in Fig. 3(c)]. \( \Phi_{\text{raw}} \) is primarily limited by the finite \( g^{(2)}(0) \) according to \( \Phi_{\text{raw}} \approx \Phi/\sqrt{1 + 2g^{(2)}(0)} \), which assumes the multiphoton contribution to consist of distinguishable photons [43,44].

Correcting for \( g^{(2)}(0) \) and the slight imperfection of the TBI yields a corrected HOM visibility of \( \Phi = (95.7 \pm 0.8)\% \) which is compatible with the QD state of the art [45–47].

In summary, we have used a PCW embedded QD to implement a scalable protocol for the generation of time-bin entangled photonic states. This is facilitated by the PCW platform which offers a compelling marriage of spin control and photonic enhancement. Operating at high magnetic fields allows spin initialization without projective measurements, and the photon indistinguishability is independent of the magnetic field strength as a single optical transition is used for emission. Our insights from theory and simulation show a clear path toward improving the fidelity with the quality of spin rotations requiring the most
attention. Indeed, given realistic PCW parameters and perfect Raman pulses we expect to reach an error level of 2.1% per emitted photon [13]. The generalization to more photons is straightforward and only requires additional rotation and excitation pulses to create a multiphoton GHZ or 1D-cluster state with photonic qubits emitted every 28 ns. We have attempted a three-qubit GHZ state (Supplemental Material [29]) but only measured a $F_{\text{GHZ}} = (42.3 \pm 1.4\%)$ fidelity due to the imperfect Raman pulses.

Another promising aspect of our approach is the entanglement generation rate. Our 124 Hz Bell-state detection rate is already favorable against similar protocols based on NV centers (7 mHz in Ref. [17]) despite our limited and non-optimized total detection efficiency $\eta_{\text{total}} = 0.3\%$. Indeed, a QD-to-fiber collection efficiency of $\eta = 7\%$ was recently demonstrated in a similar PCW structure, and further realistic improvements may facilitate collection efficiencies as high as $\eta = 78\%$ [47]. Finally, we note that the high magnetic field regime can give access to nuclear magnon modes [48], which may be used as a long-lived quantum memory for repeater applications [49] or as an ancillary qubit for use in photonic graph state generation [50,51].

The supporting data for this Letter are openly available from [52].

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