Photon-number entanglement generated by sequential excitation of a two-level atom

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Entanglement and spontaneous emission are fundamental quantum phenomena that drive many applications of quantum physics. During the spontaneous emission of light from an excited two-level atom, the atom briefly becomes entangled with the photonic field. Here we show that this natural process can be used to produce photon-number entangled states of light distributed in time. By exciting a quantum dot—an artificial two-level atom—with two sequential π-pulses, we generate a photon-number Bell state. We characterize this state using time-resolved intensity and phase correlation measurements. Furthermore, we theoretically show that applying longer sequences of pulses to a two-level atom can produce a series of multi-temporal mode entangled states with properties intrinsically related to the Fibonacci sequence. Our results on photon-number entanglement can be further exploited to generate new states of quantum light with applications in quantum technologies.
intensity correlation of $g^{(2)} = 1$ and an average photon number of $\mu = 1$ at the source. We confirm this prediction by measuring $g^{(2)} = 0.99 \pm 0.02$ and $\mu/\mu_0 = 1.02 \pm 0.01$ with respect to the average photon number $\mu_0$ produced by a single pulse, which is expected to be near unity at the source. We also verify that producing three or more photons is rare by measuring a small third-order correlation $g^{(3)} = 0.165 \pm 0.007$, which corresponds to a three-photon emission probability of about 3% (a detailed discussion on photon number probabilities and losses is given in the Supplementary Information). The photon statistics thus already suggest a state of the form $|0 \rangle + |2 \rangle$. It remains now to demonstrate a separation of the two photons into early and late time-bins [11], and the presence of coherence with the vacuum part of the state $|00 \rangle$.

To access temporal properties after the application of two pulses, we first measure the temporal profile and find that it matches well with the profile produced after excitation by a single $\pi$-pulse (Fig. 2a), as foreshadowed by Fig. 1. By sweeping $T$ across the wave-packet, we find that the proportion of counts $p_a = \mu/\mu$ detected in each time-bin $a \in \{e, l\}$ cross at the half-life condition (Fig. 2b). This matches the trend given by the ideal $|\phi^+ \rangle$.

We study the two photons composing the total temporal profile by performing time-resolved intensity correlation measurements (Methods). This produces a two-time coincidence map $g^{(2)}(t_1, t_2)$ that can be divided into four time-bin quadrants defined by a chosen $T$, designated ee, el, le and ll as shown in Fig. 2c. The direct inspection of this map reveals that coincident counts between different time-bins (el, le) predominantly occur when $T = \Delta t \approx T_{1/2}$, indicating that the two photons are indeed temporally separated.

To quantify this observation, we analyse each quadrant of the $g^{(2)}$ map individually. The counts in each quadrant are summed and normalized by the product of average photon numbers $\mu, \mu_0$ obtained within each pair of bins $a, b \in \{e, l\}$. This gives the normalized correlation $g^{(2)}_{ab}$ for the pair of time-bins where a coincidence count was detected. The total $g^{(2)}$ is then seen as an average of each $g^{(2)}_{ab}$ weighted by the proportion $p_a p_b$.

The time-bin analysis of $g^{(2)}$ presented in Fig. 2d reveals that anti-bunching occurs for detection within the same bins ($g^{(2)}_{ee, el} < 1$), whereas bunching occurs between different bins ($g^{(2)}_{el, le} = g^{(2)}_{el, le} > 1$). Bunching is maximum when the time-bin threshold is chosen at the half-life; however, the amount of bunching is less than would be expected from an ideal state produced by infinitesimally short pulses and measured with perfect time resolution (dashed curves). This is primarily due to the detection time jitter (Methods), which occasionally detects photons in quadrants ee and ll that would otherwise reside in el or le, and hence decreases $g^{(2)}_{el}$ while increasing $g^{(2)}_{ee}$ and $g^{(2)}_{el}$. From this intensity correlation analysis, we find that 81.5 $\pm$ 0.4% of two-photon measurements occur in different time-bins, evidencing a primary [11] component. We now probe the expected coherent properties of the photonic state using phase correlation measurements.

The intensity correlation $g^{(2)}_{\text{HOM}}$, at the output of a path-unbalanced Mach–Zehnder interferometer—commonly used to measure HOM bunching—can oscillate as the interferometer phase $\phi$ evolves. This occurs when the input contains coherence between any states that differ by two photons (second-order coherence). We recently used this technique to measure the amount of coherence generated between the vacuum and two photons when exciting a two-level atom with a single $2\pi$-pulse [4]. We use this same concept to characterize the number coherence generated after applying sequential $\pi$-pulses by interfering two of the generated photonic states (Fig. 3a).

The correlation $g^{(2)}_{\text{HOM}}(\phi)$ depends on both $g^{(2)}$ and the mean wave-packet overlap $M$ (ref. [3]). By considering photon-number coherence, a phase-dependent term arises [26]:

$$2g^{(2)}_{\text{HOM}}(\phi) = 1 - M + g^{(2)} - c^{(2)} \cos(2\phi),$$

(1)
Fig. 2 | Characterization of intensity. a, The temporal profile measured after applying two π-pulses separated in time by the half-life $\Delta t \simeq T_{\pi/2}$. The profile is divided into early and late bins defined by the $T$. The black dashed line shows the single-photon profile obtained for the same measurement duration after applying a single π-pulse. b, The normalized difference in average photon number $c_{\ell e} = -c_{ee}$, by analytically each quadrant of the time-resolved correlation map $G^{(2)}(t, t')$ divided by $T$ into four quadrants corresponding to each pair of time-bins where a coincidence detection occurs. The intensity correlation for each quadrant $g_{ab}^{(2)}$ normalized by the square average photon number $\mu_\ell$ detected in bins $a, b \in \{e, l\}$, computed from $c$ as $T$ is swept over the wavepacket. We obtain $g_{el}^{(2)} = g_{le}^{(2)}$ by averaging the counts in the off-diagonal quadrants. The dashed curves in b and d show the values expected for an ideal $|\phi^+\rangle$. The solid curves show the measured values where the standard uncertainty is smaller than the thickness of the line.

where $c_{\ell e}^{(2)}$ is an intensity-normalized value quantifying the second-order coherence, as described in the Supplementary Information. In our setup, $\phi$ freely evolves on a slow timescale (Methods). To accurately extract $c_{\ell e}^{(2)}$, we simultaneously monitor the self-homodyne signal $I_{el} = (\mu^+ - \mu^-) / \mu \propto \cos(\phi)$, which is the normalized difference in average photon number $\mu^\ell$ detected at each output. As $g_{el}^{(2)}$ and $I_{el}$ depend on the phase through $-\cos(2\phi)$ and $\cos(\phi)$, respectively, we expect a quadratic phase-correlated parametric relationship $g_{el}^{(2)} \approx -I_{el}$, with an amplitude of $c_{\ell e}^{(2)}$.

Although an ideal $|\phi^+\rangle$ should give $I_{el} = 0$, as it does not have first-order coherence, the finite temporal width of pulses applied to the atom inevitably causes a small signal $I_{el} \ll 1$. We believe this signal is produced by the atom directly when a photon is occasionally emitted during the excitation pulse, which allows for the remainder of the pulse to prepare the atom in a superposition state; however, it could also arise from over/underestimating the π-pulse conditions or from imperfect polarization filtering of the excitation pulses. By monitoring this remnant self-homodyne signal, we observe the expected quadratic signature and use it to measure $c_{\ell e}^{(2)}$ for three different pulse separations $\Delta t$ (Fig. 2b). The amplitude of oscillation increases with decreasing $\Delta t$ due to normalizing by intensity, hence illustrates a convergence toward the vacuum. A full analysis and discussion of measurements when varying $\Delta t$ is available in the Supplementary Information. The total time-integrated value $c_{\ell e}^{(2)}$ indicates substantial second-order coherence, but it does not distinguish states of the form $|00\rangle + |22\rangle$ from $|00\rangle + |11\rangle$. For this, we resolve the measurement in time.

Consider the interference of two ideal Bell states (Fig. 3a). This gives rise to four cases. First, the vacuum inputs $|00\rangle$ will give a trivial output. Second, two $|11\rangle$ states will cause HOM bunching (Fig. 3c). As opposed to the ideal single-photon case in which $M = 1$, an ideal $|\phi^+\rangle$ should give $M = 1/2$. This is as both early and late photons bunch with their pair in the same time-bin ($M_e = M_l = 1$), but each pair can still exit the beam splitter independently ($M_{el} = M_{le} = 0$). Third, the cases combining $|00\rangle$ and $|11\rangle$ can occur in two ways (Fig. 3d). If cases two and three produce indistinguishable outputs, then a quantum interference occurs due to the erasure of the information about which path the two photons took through the interferometer. This two-photon interference evidences the presence of a path-entangled Bell state between the upper $U$ and lower $L$ paths of the interferometer: $(a|U\rangle \langle 0|U_\ell + e^{i\theta}\langle L_e|L_\ell|0\rangle) / \sqrt{2}$, and it causes an oscillation of coincident counts depending on $\phi$ that contributes to the $c_{\ell e}^{(2)}$ term of $g_{el}^{(2)}$. Thus, monitoring the oscillation of coincident counts constitutes a Bell-state measurement of this path-entangled state produced by the $|\phi^+\rangle$ input; however, to distinguish $|\phi^+\rangle$ from arbitrary states of the form $|00\rangle + |22\rangle$, we must measure the component of $c_{\ell e}^{(2)}$ that arises from coincidences between photons arriving in different time-bins and show that it exceeds any contribution from $c_{ee}^{(2)}$.

To measure $M_{ab}$ and $c_{ab}^{(2)}$, we use the same approach used to obtain $g_{el}^{(2)}$, by analysing each quadrant of the time-resolved correlation map $G_{HOM}^{(2)}(t_1, t_2, \phi)$ for a given $T$. We subdivide each map $G_{HOM}^{(2)}(t_1, t_2, \phi)$ corresponding to each $I_{el}(\phi)$, which produces four quadratic signatures similar to those presented in Fig. 3b, with one
corresponding to each quadrant. We then fit these four sets of data to extract the quantities $c_\delta^{(1)}$ and $M_\delta$.

From the time-bin analysis of phase correlations, we see that the mean wavepacket overlaps of photons in the same bins $M_e$ and $M_\delta$ both remain relatively high and intersect at the half-life, whereas the overlap between bins $M_\delta$ dips nearly to zero (Fig. 3c). This indicates that the photons composing $|11\rangle$ are mostly indistinguishable, yet almost fully distinguishable from each other. Interestingly, $M_e$ exceeds the mean wavepacket overlap measured after a single x-pulse ($M = 0.77$) when $T < T_{5%}$. We attribute this to the sharp temporal truncation of photons in the early bin, which causes a spectral broadening that partially overcomes dephasing. This truncation does not modify the temporal shape of photons in the late bin, which remain as exponentially decaying profiles. Hence, $M_e$ converges to the single-photon case when $T > T_{5%}$. The observed crossing and dip follows that predicted by the ideal state and verifies the scenario described by Fig. 3c when $T = T_{5%}$.

We find that the trend for $c_\delta^{(1)}$ mimics that of $g_\delta^{(2)}$ as predicted by the ideal state, with $c_\delta^{(1)}$ peaking when $ce_\delta^{(2)}$ and $c_\delta^{(2)}$ intersect at the half-life (Fig. 3f); however, the magnitudes are further suppressed relative to the ideal case as the coherence is susceptible to dephasing in addition to errors caused by imperfect pulses and detection jitter. That said, we find that $ce_\delta^{(2)}$ is much greater than $c_\delta^{(2)}$ and $c_\delta^{(2)}$ at the half-life, indicating that the majority of the oscillation observed in $g_\delta^{(2)}$ arises from a coherence between the vacuum $|00\rangle$ and two photons arriving in orthogonal time-bins $|11\rangle$.

The three intensity-normalized quantities $c_\delta^{(2)}$, $M_e$, and $M_\delta$ can be used to estimate the magnitude of some density matrix elements of the photonic state at the source, before losses from collection. From this, we estimate that the emitted state has an entanglement concurrence of $C = 0.70 \pm 0.05$. Note that a positive value $C > 0$ unambiguously indicates the presence of quantum entanglement. We also estimate a fidelity of $\mathcal{F} = 0.79 \pm 0.03$ with respect to $|\psi^+\rangle$.

**Discussion**

Our fidelity and concurrence estimates are limited by the detector jitter time. The measurements of $c_\delta^{(2)}$ and $M$ suggest that a fidelity up to 0.86 is possible with this device using detectors with a time jitter well below\(^\text{17}\) the pulse timescale ($t_p = 20 \text{ ps}$) used in our experiments, which would reduce the proportion of two-photon events occurring within the same time-bins (see or I) down to $3t_p/8T \approx 5\%$ (Supplementary Information). Using shorter pulses ($t_p \ll T_e$) in combination with low time-jitter detectors would bring the fidelity up to at most $\sqrt{M} \approx 0.91$, which is limited by the dephasing of this device. Note that $M_e \geq 0.975$ has been achieved with quantum dot devices\(^\text{4,30}\), which could provide a fidelity of up to 0.987.

In our experiments we do not perform a full quantum state tomography on the photonic state to retrieve its density matrix as it is difficult to spatially separate and independently analyse the time-bin modes. We instead characterize the photon-number entanglement of the state via intensity and phase correlation measurements, which, under some reasonable assumptions, allows for a partial reconstruction of the density matrix and for estimates of fidelity and concurrence (Supplementary Information). One approach to separate the time-bins would be to use a ultrastable optical switch; this would in turn allow for single-qubit gates, quantum teleportation and Bell tests. For our system, the short lifetime dictates an optical switching time on the picosecond timescale, which is achievable using lithium niobate integrated photonic circuits\(^\text{42}\); however, our approach for generating photonic entanglement can be applied to any coherently controlled source of indistinguishable photons modelled by a two-level system.
Our entangling protocol also has a simple extension to multi-mode entanglement by applying a longer sequence of π-pulses. As detailed in the Supplementary Information, the photonic state $|\psi_i\rangle$ produced by $N$ pulses has a recursive nature that becomes transparent when labelling the time-bins in reverse chronological order. In this case, by applying the matrix product state formalism, we find that the final state can be determined from the Fibonacci-like relation

$$|\psi_N\rangle = \alpha_N |\psi_{N-2}\rangle + \beta_N |\psi_{N-1}\rangle,$$

where $\alpha_m = e^{-\Delta t_m/2T_L}$, $\beta_m = \sqrt{1 - \alpha_m^2}$, $t_m = |0\rangle = |1\rangle$, and where $m$ labels the $m$th time-bin from the end of the sequence. If $\Delta t_m > T_L$, then the atom relaxes to the ground state at the end of the sequence, then $N = 1$ pulse produces a single photon $|\psi_1\rangle = |1\rangle$ and $N = 2$ pulses produces the entangled state: $|\psi_2\rangle = \alpha_2 |0\rangle |0\rangle + \beta_2 |1\rangle |1\rangle$. By choosing the pulse separations $\Delta t_2 = T_1 \ln(2)$ and $\Delta t_1 = T_1 \ln(3)$, we obtain the maximally entangled W-class state produced by $N = 3$ pulses: $|\psi_3\rangle = (|001\rangle + |100\rangle + |111\rangle)/\sqrt{3}$. In general, the entangled states produced by this sequence belong to the class of matrix product states with two-dimensional bonds, and they are not equivalent to N-qubit W states for $N \geq 4$. Further studies are needed to identify the type and amount of entanglement provided by multi-pulse sequences applied to two-level atoms.

**Conclusion**

We have shown that the light–matter entanglement occurring during spontaneous emission from a two-level atom is a fundamental resource for generating entangled light. By probing the temporal domain of pulsed light emitted by an artificial atom after a double π-pulse excitation, our measurements demonstrate the generation of a photon-number Bell state. By adding more consecutive π-pulses, we herald that this protocol can produce multipartite temporal entanglement, and it is a step closer to the generation of high-order Fock states and cat states, which require dynamic control of the light–matter coupling strength. Such a new class of photonic states could serve as building blocks for distributing entanglement, quantum state teleportation, and may allow new ways to implement quantum random walks, quantum sensing and photonic networks. We also believe that the sequential coherent driving of multi-level atomic systems during spontaneous emission offers promising perspectives for generating high-dimensional entanglement; for example, using the biexciton-exciton cascade in semiconductor quantum dots or in combination with spin–photon entanglement protocols.

**Online content**

Any methods, additional references, Nature Research reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s41566-022-00979-z.

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Methods
More details about the quantum-dot–micropillar source used in our experiments can be found in ref. 46, source number 3, following the numbering of this reference. This source was chosen over others for its very high emission efficiency, which allows for fast collection of time-resolved maps and third-order intensity correlations. The experiments were performed in a standard resonant cross-polarization set-up\(^4\)\(^6\). We prepare the two-π pulse sequence in a compact Michelson interferometer. This provides passive phase-stabilization for the delayed output pulses and independent intensity tuning. One of the mirrors is mounted on a nanometric translation stage allowing for delay tuning of up to 175 ps. The laser pulses have a temporal full-width at half-maximum of ~20 ps. The coincidence maps are retrieved via time-tagging of the photon events with respect to the laser clock\(^3\). The phase correlation measurements are implemented in a path unbalanced Mach–Zehnder interferometer, with a delay of 12.3 ns in one of the arms, matching the laser repetition rate\(^4\). The phase of the interferometer evolves slowly, performing a π shift on the ~5 s timescale. The relatively fast 100 ms time to acquire a single time-resolved correlation map \(G_{\text{HOM}}^{(2)}(t_1, t_2, \phi)\) allows us to consider the ϕ constant for each map.

The device and set-up losses are detailed in refs. 46,47. The probability of having a single photon per pulse excitation in the collection single-mode fibre is ~10%. All measurements are performed using superconducting nanowire single-photon detectors with ~70% quantum efficiency and ~50 ps full-width at half-maximum Gaussian jitter. The \(g^2\) HOM and \(g_{\text{HOM}}^{(2)}\) measurements are intensity-normalized quantities that are insensitive to photon losses, thus allowing characterization of the photonic state at the source level before any photon collection, transmission and detection losses. Under a single π pulse excitation, the count rate per detector in the intensity correlation measurements \(g^2\) Hanbury–Brown–Twiss set-up, with three detectors) is 1.256 ± 0.007 MHz per detector, and the count rate per detector for the \(g_{\text{HOM}}^{(2)}\) (output of the Mach–Zehnder interferometer) is 0.899 ± 0.005 MHz per detector. The photon time-tags are processed in a HydraHarp 400 autocorrelator, with a temporal discretisation of 8 ps.

Data availability
The experimental data that support the findings of this study are available in figshare at https://doi.org/10.6084/m9.figshare.16838248.

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Author contributions
The experiments were conducted by J.C.L. and C.A.-S. Data analysis was carried out by S.C.W. and C.A.-S. with help from J.C.L. and P.H. Theoretical modelling was performed by S.C.W., M.M., C.S. and A.A., with help from J.C.L. and C.A.-S. Cavity devices were fabricated by A.H. and N.S. from samples grown by A.L. based on a design of L.L. Etching was performed by I.S. The manuscript was written by S.C.W. and C.A.-S. with assistance from C.S. and P.S. and input from all authors. The project was supervised by C.A.-S. with the collaboration of C.S. and P.S.

Competing interests
N.S. is a co-founder—and P.S. is a scientific advisor and co-founder—of the single-photon-source company Quanda. The other authors declare no competing interests.

Additional information
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