On-demand generation of indistinguishable polarization-entangled photon pairs

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An on-demand source of indistinguishable and entangled photon pairs is a fundamental component of various quantum information applications, including optical quantum computing, quantum repeaters, quantum teleportation and quantum communication. Parametric downconversion and four-wave mixing sources of entangled photons have shown high degrees of entanglement and indistinguishability, but the probabilistic nature of their generation process also creates zero or multiple photon pairs following a Poissonian distribution. This limits their use in complex algorithms where many qubits and gate operations are required. Here, we simultaneously show ultrahigh purity (g(2)(0) < 0.004), high entanglement fidelity (0.81 ± 0.02), high two-photon interference non-post selective visibilities (0.86 ± 0.03 and 0.71 ± 0.04) and on-demand generation (efficiency εpam = 0.86 ± 0.08) of polarization-entangled photon pairs from a single semiconductor quantum dot. Through a two-photon resonant excitation scheme, the biexciton population is deterministically prepared by a π-pulse (εbiexciton = 0.98 ± 0.07). Applied on a quantum dot showing no exciton fine-structure splitting, this results in the deterministic generation of indistinguishable entangled photon pairs.

To date, spontaneous parametric downconversion (SPDC) and four-wave mixing sources have been mostly used for the generation of entangled photon pairs to realize quantum communication protocols and to demonstrate basic quantum logic experiments. However, the photon pair statistics of these sources is described by a Poissonian distribution, which also implies the generation of zero and multiple pairs. This leads to errors in the quantum algorithm protocols, effectively limiting their usefulness for deterministic quantum technologies. Radiative cascades in single quantum emitters, such as atoms or quantum dots, can in principle emit, on demand, single pairs of polarization-entangled photons with high generation efficiencies. After optical excitation of two electron–hole pairs (biexciton, termed the |XX⟩ state) in a quantum dot, the biexciton decays through a two-photon cascade (Fig. 1a). If the fine-structure splitting between the intermediate states (excitons, termed |X⟩) is smaller than the radiative linewidth, the two decay paths will be indistinguishable and the two photons will be polarization-entangled, which results in a two-photon Bell state |ψ⟩ = 1/√2(|HXX⟩|HXY⟩ + |VXX⟩|VXY⟩). To ensure the emission of a single pair of entangled photons per excitation pulse, the biexcitonic state has to be pumped into saturation. So far, non-resonant pulsed pumping schemes have been successfully applied for entangled photon generation, but without providing simultaneous information on indistinguishability. In any case, it is well known that non-resonant pumping schemes limit the coherence and indistinguishability of the emitted photons, making them unsuitable for many quantum information applications. In a recent study, Stevenson and co-workers reported the interference and entanglement properties of photons emitted by a quantum dot embedded within a light-emitting diode. However, the measurements were performed in a continuous and sinusoidal a.c. mode without clear pulse separation, and a post-selected visibility of 0.6 was achieved, which was limited by detector jitter. Resonant coherent excitation on quantum dots has proven to be very efficient for achieving a high degree of indistinguishability and coherence.

On-demand single-photon sources with a high degree of indistinguishability of 97% have been realized recently after coherent resonant π-pulse excitation of an excitonic state in a quantum dot.

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Figure 1: Two-photon biexciton excitation scheme for a quantum dot exhibiting no excitonic fine-structure splitting. a, A shaped laser is brought to resonance with the virtual biexciton TPE state, the energy of which lies between the exciton and biexciton emission lines. Two radiative recombination paths are possible to the ground state (|0⟩) via one of two bright exciton states (|X⟩). The polarizations of the two photons are determined by the intermediate exciton state. b, Above-GaAs-bandgap excitation spectrum. The presence of the trion is due to charge capture in the quantum dot. c, Emission spectrum under biexciton direct TPE. Only the exciton and biexciton can be observed, with the same intensities. No trion line is observed. This confirms that excitonic and biexcitonic photons are always emitted as pairs and that the exciton population cannot be transferred to the trion state by capture of charge before the radiative cascade is over. It is worth noting that the generation efficiency of the biexciton increases by a factor of ~4 from non-resonant (in saturation) to resonant excitation (see Supplementary Section 4). The large peak between the exciton and biexciton emission lines is due to some residual scattered laser light.
However, implementation of one-photon resonant excitation of the biexcitonic state is not possible due to the optical selection rules. Here, a resonant two-photon excitation (TPE) scheme, already observed in bulk crystal \cite{15,16} and single quantum dots \cite{17-19}, is applied to coherently populate the biexcitonic state. By setting the pump intensity so that the inversion of the quantum dot from the ground to the biexcitonic state is most probable (so-called $\pi$-pulse), one can deterministically prepare a biexciton after each pulse with near unity fidelity. This gain of efficiency is achieved without any negative impact on multi-photon-reaction probability, as is intrinsically the case for SPDC and is typically reported for incoherent pumping schemes of quantum dots due to the increased contribution of spectrally nearby transitions \cite{20}. Even more importantly, the resonant TPE process also strongly attenuates the contribution to decoherence, which affects solid-state emitters because no phonon relaxation processes are needed for biexciton preparation and no charge carrier is released in the semiconductor matrix, preventing carrier–carrier scattering processes with nearby charge carriers.

We used an epitaxially grown (In,Ga)/GaAs quantum dot for our studies (see Methods). Figure 1b shows a photoluminescence spectrum under above-bandgap excitation. The exciton (1.4212 eV) and biexciton (1.4189 eV) lines are separated by the biexciton binding energy (2.3 meV). The higher energy line corresponds to the trion (indicated by ‘T’), which comprises an exciton and an additional charge. Previous high-resolution polarization-dependent studies have shown that the exciton fine-structure splitting is below 0.3 meV (ref. 11). In the following, we address the quantum dot by tuning a linearly polarized pulsed laser (full-width at half-maximum (FWHM) = 95 meV) in resonance with the virtual state of the biexciton TPE (Fig. 1a). Figure 1c shows a quantum dot spectrum under this TPE. The intensities of the two lines are similar, because only a biexciton can be prepared after each pulse, and the exciton is populated through the radiative cascade after recombination of the biexciton. Because the energy of the laser pulse differs from the exciton and biexciton lines by half the biexciton binding energy, a laser background-free signal of both lines (X and XX) can be collected without any polarization suppression technique, as typically used for resonant excitation experiments on excitons \cite{21,22}. On the one hand, this allows polarization-dependent cross-correlation measurements of the biexciton–exciton cascade (see below), and on the other hand, it allows a perfect single-photon purity ($g^{(2)}(0) < 0.004$) to be achieved for both transitions (Fig. 2).

Figure 3 shows the excitation power dependence of the integrated intensity of the exciton and biexciton lines, respectively. Well-pronounced Rabi oscillations are observed for the biexciton \cite{17,18} emission intensities (Fig. 3a). They reflect the coherent nature of the TPE process together with a high biexciton occupation probability ($\epsilon_{XX} = 0.98 \pm 0.07$). As shown in the Supplementary Section 4, the observed damping is mainly due to phonon-induced decoherence processes and is thus stronger when the quantum dot state rotates faster between $|0\rangle$ and $|XX\rangle$ (that is, when the pulse area is increased) \cite{23}. The two distinctive signatures of the TPE process are the slow increase of the intensity at low power and the change of the Rabi oscillations’ frequency with pump power \cite{27}. The slightly monotonous superimposed on the oscillations can be attributed to the slight chirp \cite{24} of the excitation pulse (see Supplementary Sections 1 and 4) or to some incoherent contribution. Fed through the radiative cascade by the biexciton recombination, the exciton intensity ($\epsilon_X = 0.86 \pm 0.08$) follows a very similar dependence (Fig. 3b).

To evaluate the impact of this quantum dot coherent control on the degree of entanglement of the emitted photon pairs, we performed quantum-state tomography measurements under $\pi$-pulse excitation. By measuring the degrees of polarization correlation between the biexciton and exciton in the linear, diagonal and circular bases, one can obtain a close approximation of the fidelity to the Bell state \cite{25}. Figure 4 presents the six corresponding correlation histograms. In the linear and diagonal bases they show strong bunching when the polarizations of the excitonic and biexcitonic photons are parallel, and antibunching when they are orthogonal. The situation is opposite in the circular basis, where antibunching occurs when the photons are prepared in parallel polarizations. A bunching can be observed when photons are projected in orthogonal circular polarizations. This set of correlation measurements shows a clear signature of entanglement. The
The respective contrasts extracted from them in the linear, diagonal and circular bases are

\[ g_{\text{linear}} = 0.87 \pm 0.02 \]

\[ g_{\text{diagonal}} = 0.67 \pm 0.04 \]

\[ g_{\text{circular}} = -0.69 \pm 0.02 \]

The fidelity of the state emitted by the quantum dot with respect to the expected state \(|\psi^+\rangle\) is calculated from the three contrasts\(^{25}\):

\[ f = \frac{1 + g_{\text{linear}} + g_{\text{diagonal}} - g_{\text{circular}}}{4} = 0.81 \pm 0.02 \]

This is a clear improvement over the fidelity of 0.72 ± 0.01 extracted from a previous quantum-state tomography performed on the same quantum dot using above-bandgap excitation\(^{11}\), and is related to the improvement in the coherence time \(T_2\), from 229 ps to 357 ps for the X photon and from 114 ps to 192 ps for the XX photon) as a consequence of the XX resonant excitation (see Supplementary Section 5). The still imperfect fidelity might be related to cross-dephasing events that involve polarization-dependent phonon interaction\(^{25}\).

To study the indistinguishability of the photons, two-photon interference measurements were performed. The biexcitonic state of the quantum dot was excited twice every 13 ns by a pair of \(\pi\) pulses separated by a 4 ns delay. With each excitation pulse the quantum dot emitted a pair of photons (X and XX). After spectral selection and projection of their polarization to the horizontal, the photons from the same transition (X and XX) were then fed into an unbalanced Mach–Zehnder interferometer (MZI) with a 4 ns path-length difference. Within the MZI the photons were prepared in orthogonal or parallel polarizations, making them distinguishable or indistinguishable with respect to their polarizations. The two outputs of this interferometer were detected by single-mode fibre-coupled single-photon counters, and a histogram was constructed with the recorded coincidences (Fig. 5). The two photons arrive at the beamsplitter with delays of \(\pm 8, \pm 4, 0, 4\) and 8 ns, building a cluster of five peaks. Because of the 13 ns delay between the clusters, the two outer peaks of a cluster temporally overlap with corresponding peaks from the previous/successive cluster. However, the central peak, which reflects the two-photon interference probability, is well resolved and not affected by any overlap due to the 4 ns time separation between the pulses. This allows a background-free determination of the two-photon interference effect for both transitions (X and XX). Figure 5a,b shows a strong suppression of both coincidence peaks at zero delay when the two incoming photons are prepared in the same polarization (red and blue), in contrast to the orthogonal polarization case (grey). Quantitative evaluation of the raw data results in two-photon interference visibilities of 0.58 ± 0.04 for XX and 0.44 ± 0.03 for X. Taking into account the experimental imperfections such as avalanche photodiode (APDs) dark counts, the reduced mode overlap at the beamsplitter \((1 - \epsilon = 0.95 \pm 0.01)\) and the residual two-photon emission probabilities, we obtain the corrected degrees of indistinguishabilities to be 0.84 ± 0.05 for XX and 0.69 ± 0.04 for X. The degree of

Figure 4 | Biexciton-exciton polarization-dependent cross-correlation histograms under resonant excitation. a–c, Results for the linear basis (a), the diagonal basis (b) and the circular basis (c). An antibunching is observed when the biexcitonic and excitonic photons are projected in orthogonal polarizations in the linear and diagonal bases. A bunching is observed for parallel polarizations. In the circular basis, the opposite is observed. The relative areas of the zero delay peaks from these six histograms are used to derive the cross-polarization contrasts. For clarity, data corresponding to orthogonal polarization configurations are time-shifted.

Figure 5 | Two-photon interference histograms, obtained under resonant excitation. a,b, Results for excitonic photons (a) and biexcitonic photons (b). On both graphs, the coloured plots correspond to the two-photon interference pattern of the parallel-polarized single photons and the grey graphs correspond to the two-photon interference of cross-polarized single photons. When photons arrive at the beamsplitter in parallel polarization (that is, they are indistinguishable), the central peak is strongly suppressed. The grey histograms are used to quantify the central peak suppression and to extract the visibilities of the two-photon interference. Side peaks were also used for cross-checking the extracted values. They were evaluated to 0.86 ± 0.03 for XX and 0.71 ± 0.04 for X after dark count and set-up imperfection corrections.
indistinguishability can be independently checked using the relative intensities of the side peaks and their known peak area ratios, which essentially produces the same results: 0.86 ± 0.03 for XX and 0.71 ± 0.04 for X (for more details see Supplementary Section 7). The difference between the exciton and the biexciton visibilities arises from the fact that the biexciton state lifetime introduces a time jitter to the exciton population, which results in a degradation of the excitonic photon indistinguishability9–28. We anticipate that this issue could be overcome by bringing a cavity mode in resonance with the biexciton transition. The enhancement of the biexciton radiative rate by a Purcell effect would efficiently reduce the undesired time jitter.

All the features presented here show that the resonant biexciton excitation is particularly suited to quantum computation or communication schemes29, which require efficiently generated photons linked by a high degree of entanglement and able to interfere correctly on a beamsplitter. Ultimately, more sophisticated coherent control of the quantum dot state could upgrade the performances of the source even further. The use of the pulse echo technique9,30, suppressing the precession of the Bloch vector during excitation, would be another step towards very pure state generation. Application of the TPE technique on symmetric quantum dots (C_{3v})31 with nearly zero fine-structure splittings embedded in resonator9 or nanowire structures32, which allow ultrahigh collection efficiency, would open the way to very competitive semiconductor entangled photon sources.

Methods

The investigated sample consisted of a single layer of self-assembled In(Ga)As quantum dots on a semi-insulating GaAs (001) substrate, and was grown by molecular beam epitaxy. The quantum dots were confined within a 260-nm-thick GaAs layer, which acted as a cavity. A bottom distributed Bragg reflector (DBR) made up of 15 pairs of AlAs/GaAs (78.4 nm/65 nm) and one single pair of GaAs/AlAs on top was grown to enhance the collection efficiency. To carry out micro-photoluminescence measurements, the sample was mounted in a helium-flow cryostat at 4 K. Adopting an orthogonal geometry for excitation and detection improved the rejection of scattered laser light. For side excitation with shaped pulses from a Ti: Sapphire laser system with a repetition rate of 76 MHz (see Supplementary Section 1), the DBR structure acted as a waveguide. Quantum dot emission was collected from the top (NA = 0.4) and analysed with a spectrometer/charge-coupled device combination. This could be used to record the emission light spectrum with a resolution of 50 μeV or as a monochromator for spectral filtering. For autocorrelation, as well as for indistinguishability measurements, fibre-coupled set-ups were used. To perform the quantum state tomography experiment, an assembly of waveplates (λ/2, λ/4) and polarizing beam splitters projected the quantum dot luminescence in the particular polarization basis before the spectral filtering of each individual emission line took place. Three polarization-independent interferential filters (ΔA = 0.8 meV), adjusted for laser wavelength, were mounted in the detection path for laser suppression. Single-mode fibres, connecting the APDs, provided spatial filtering of the scattered laser light. The external efficiency, that is, the collection efficiency of the set-up, was estimated by using the count rate per APD in the correlation measurements (2,500–3,000 counts s^{-1}) and reversely adding up the optical losses and efficiencies of each element of the set-up. This led to a count rate of ~200–300 kilocounts s^{-1} after the first lens and hence a collection efficiency of ~0.4%.

To determine the biexciton preparation fidelity we carried out calculations using a numerically exact real-time path-integral approach. Within this microscopic treatment we modelled the quantum dot as an electronic four-level system (Fig. 1a) driven by an external laser field and coupled to a continuum of acoustic bulk phonons, where both the carrier light as well as the carrier–phonon interaction were fully taken into account. Details of the method and the model, including material parameters, can be found in refs 33 and 24, respectively, while details of our analysis are provided in the Supplementary Section 4.

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Author contributions
M.M., S.B., K.D.J. and P.M. conceived and designed the experiments. M.M., S.B. and K.D.J. performed the experiments. M.M. and S.B. analysed the data. M.G. carried out the theoretical calculations. S.B. and P.M. wrote the manuscript with input from the other authors.

Additional information
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Competing financial interests
The authors declare no competing financial interests.