Towards quantum-dot arrays of entangled photon emitters

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To make photonic quantum information a reality1,2, a number of extraordinary challenges need to be overcome. One challenge is to achieve large arrays of reproducible ‘entangled’ photon generators, while maintaining compatibility for integration with optical devices and detectors3–5. Semiconductor quantum dots are potentially ideal for this as they allow photons to be generated on demand6–8 without relying on probabilistic processes8,9. Nevertheless, most quantum-dot systems are generated on demand6,7 without relying on probabilistic processes8,9. Nevertheless, most quantum-dot systems are limited by their intrinsic lack of symmetry, which allows only a small number (typically 1 out of 100, or worse) of good dots to be achieved per chip. The recent retraction of Mohan et al.10 seemed to question the very possibility of simultaneously achieving site control and high symmetry. Here, we show that with a new family of (111)-grown pyramidal site-controlled InGaAs quantum dots it is possible to overcome previous hurdles and obtain areas with up to 15% of polarization-entangled photon emitters, with fidelities as high as 0.721 ± 0.043.

The idea underlining the principle of entangled photon emission with quantum dots relies on fundamental quantum physics: particle indistinguishability generates a superposition state when two energetically nearly degenerate quantum levels are populated at the same time. In quantum dots, the entanglement resides in the polarization of two photons emitted during cascaded biexciton–exciton recombination11. This leads to one difficulty: when the two excitonic levels are not perfectly degenerate (that is, there is a fine structure splitting, FSS), entanglement in the emission persists, but a phase term is proportional to both energy and time. This results in a relative rotation of the two photon polarizations (not constant in time), making entanglement virtually impossible to detect in a simple manner12.

All currently reported quantum dot systems allowing entangled photon emission tend to have large FSS, fundamentally allowing only a few (post-growth selected) quantum dots on a semiconductor wafer to be good sources. Furthermore, to date, no entangled photon emission has been demonstrated in any system where accurate quantum dot position control is possible. It is clear from textbook physics that, to observe level degeneracy, one needs symmetric confinement. As discussed in a number of publications, growth along the [111]B crystallographic direction ideally shows C\textsubscript{4v} symmetry13–15, which should allow the realization of large arrays of position-controlled entangled photon emitters. In practice, however, a relatively broad range of FSS can be found on existing (111) systems, as for example in our pyramidal quantum dots. An efficient way to overcome asymmetry-related issues comprised exposing a quantum-dot layer to unsymmetrical dimethylhydrazine (U-DMHy, a standard source of nitrogen during metal–organic vapour phase epitaxy, MOVPE) during the quantum-dot formation process16,17. It was consistently observed that the presence of U-DMHy, within a certain range of growth conditions, helped to improve the symmetry of the quantum dots, enabling reproducible fabrication of nanostructures with FSS consistently below our detection limit of ~4 µeV.

The investigated In\textsubscript{0.25}Ga\textsubscript{0.75}As\textsubscript{1–x}N\textsubscript{x} quantum dots were grown by MOVPE in 7.5-µm-pitch tetrahedral recesses etched in (111)B-oriented GaAs. An atomic force microscopy (AFM) image (Fig. 1a) of a cleaved sample in side view shows the epitaxial layer structure (see Methods), with a quantum dot located at the central axis of the recess, within GaAs barriers. Photoluminescence extraction enhancement was achieved by selectively removing (back-etching) the substrate (Fig. 1b). The typical Lorentzian linewidth of the exciton transition was found to be 80 ± 15 µeV in our first samples. As we discuss in Supplementary section ‘Linewidth’, this broadening is not a fundamental limitation.

Figure 1c presents a typical photoluminescence spectrum of the single quantum dots investigated in this work. The characteristic feature is a biexciton transition (XX) at higher energy (antibinding biexciton) in entangled photon emitters, always accompanied by the presence of a charged exciton (X\textsuperscript{*}) at higher energy. The significance of the spectrum must be stressed, as it acts as a very precise and quick indicator for preselecting quantum dots that emit polarization-entangled photons (see Supplementary section ‘Significance of the excitonic pattern’, for more details).

Figure 1e shows an isotropic linear polarization distribution proving quantum dots to be sources of unpolarized light. The dependence of photoluminescence intensity on excitation power is presented in Fig. 1d. This acts as a preliminary indicator of the excitonic transitions. Despite the deviation from the ideal linear and quadratic power dependence, the type of transition was unambiguously confirmed by photon correlation measurements. Average lifetime values are also consistent with the attributed transition types: 1.8 ± 0.6 ns and 0.9 ± 0.15 ns for exciton and biexciton, respectively. Figure 1f illustrates exciton (X) and biexciton (XX) transitions measured/filtered at perpendicular linear polarization angles. No difference can be identified visually between the spectra. In the presence of low symmetry (that is, with FSS), both peaks should be composed of two energetically distinguishable linearly polarized components (typically referred to as H and V). Here, no particular crystallographic direction can be consistently associated with H and V components, as the origins of FSS are related to random effects, and not to a shape elongation along certain directions as in self-assembled quantum dots. In our work, H and V only indicate 0 and 90° angles with respect to the linear polarizer. To obtain the exact value of FSS we apply a well-known FSS measuring procedure that involves taking a set of polarized spectra at smaller polarization angle steps (Fig. 1g)\textsuperscript{18}.

If the intermediate exciton level is degenerate, during the recombination cascade the emitted pair has a polarization
entanglement expressed by the Bell state \( |\psi\rangle = \frac{1}{\sqrt{2}}( |XX\rangle + |RR\rangle) \). Using Jones vectors, the state can be rewritten as \( |\psi\rangle = \frac{1}{\sqrt{2}}( |HH\rangle + |VV\rangle) \) or \( |\psi\rangle = \frac{1}{\sqrt{2}}( |HH\rangle + |DD\rangle ) + |AA\rangle \), where \( L(R) \) are circularly left (right) hand, \( H(V) \) are horizontally (vertically) and \( D(A) \) are diagonally (antidiagonally) polarized photons, respectively, and \( XX \) and \( X \) indicate biexciton and exciton, respectively. As a consequence, an ideal source of polarization-entangled photons should display a perfect correlation measured in the linear and diagonal bases and a perfect anticorrelation in a circular basis. Figure 2a presents polarization-resolved second-order correlation functions taken in the abovementioned polarizations on a representative dot. Clear bunching is observed in co-polarized linear and diagonal and counter-polarized circular photon correlation curves (these non-classical correlations can be lost as soon as the FSS is measurable, for example, \( \sim 3 \) μeV; see Supplementary Information). The degree of correlation of an unpolarized source can be obtained directly from the experimental data according to \( C_{\text{basis}}(x) = |\langle\phi|x\rangle|^2 = |\langle\psi|x\rangle|^2 = \frac{g_{xx,x}^2}{g_{xx,x}^2 + g_{xx,x}^2} \), where \( xx(x) \) indicates the polarization of a biexciton (exciton) and \( x \) the orthogonal polarization of an exciton. Figure 2b plots degrees of correlation at different excitation delays for the same quantum dot. This can be used to calculate the fidelity of the entangled state \( \rho = \frac{1}{2}( |XX\rangle \langle XX| + |RR\rangle \langle RR|) \) (see Methods). In the statistical distribution of quantum dots described in the following, we use the fidelity as a figure of merit to evaluate entanglement, as the experimental procedure is significantly less time-consuming than reconstructing a full density matrix. This is accepted by the scientific community as a proof of entanglement (indeed the method gives an equivalent result to a full quantum state tomography). The calculated fidelity value of 0.670 ± 0.035 in the given example (Fig. 2a,b) exceeds the maximum limit of classically correlated light (0.5) by nearly five standard deviations, indicating the entangled nature of the emitted photons. The highest obtained value over the sample was 0.721 ± 0.043. As discussed elsewhere, a variety of effects, such as spin scattering, background light and dephasing, are responsible for producing reduced fidelities.

In general, a two-photon polarization state can be fully described by a density matrix. Figure 2c plots real and imaginary parts of the density matrix reconstructed from the experimental data (for the representative analysed quantum dot) using a quantum state tomography procedure. The density matrix is represented using a quantum state tomography. The calculated fidelity value of 0.670 ± 0.035 in the given example (Fig. 2a,b) exceeds the maximum limit of classically correlated light (0.5) by nearly five standard deviations, indicating the entangled nature of the emitted photons. The highest obtained value over the sample was 0.721 ± 0.043. As discussed elsewhere, a variety of effects, such as spin scattering, background light and dephasing, are responsible for producing reduced fidelities.
Figure 2 | Polarization-entangled photons. a, Representative second-order correlation functions taken for co-polarized and cross-polarized biexciton–exciton photons in three different polarization bases for a specific dot as discussed in the main text. Strong bunching of co-polarized linear and diagonal and cross-polarized circular $g^{(2)}$ functions clearly identifies non-classical correlations. As a rule of thumb, when curves are normalized with respect to the side peaks, entanglement can be identified by analysing each pair of $g^{(2)}$ curves. The area under each bunching peak (around zero delay) has to be more than twice the area under the corresponding peak (ideally antibunched) from the other curve; $g^{(2)}$ bunched $> 2g^{(2)}$ antibunched (see Methods). b, Degrees of correlation in different polarization bases for the same measurement/dot. c, Representative density matrix reconstructed by a quantum state tomography procedure (note that a different quantum dot was used in these measurements than in a and b).

Figure 3 | High density of quantum dots emitting polarization-entangled photons. a, Distribution of fidelity values of all measured quantum dots. The limit of 0.5 for classically correlated light is passed in the majority of cases. b, Distribution of emission energy of the quantum dots in a (standard deviation of 4.7 meV). Locally (for example, areas A and B), the standard deviation can be reduced to $\sim 2.5$ meV. c, Images of two randomly selected areas (A and B in the previous graphs) taken with the sample surface visualization system installed in the set-up. The green and red circles identify all preselected quantum dots for polarization-entanglement characterization. The green spots identify quantum dots with an entangled state fidelity (values are above the spots) of >0.5, whereas red circles indicate fidelities of <0.5.
To demonstrate the high density of entangled photon emitters, a number of randomly chosen quantum dots, preselected mostly according to their photoluminescence spectrum (Fig. 1c) (as discussed above), were characterized in terms of fidelity. Figure 3a presents a distribution of measured fidelity values, which shows that 75% of the preselected quantum dots passed the limit of 0.5 for classically correlated light. Figure 3c shows two images of two randomly selected areas (A and B) obtained by the sample surface imaging system. Green circles indicate quantum dots that have a fidelity of the entangled state that is >0.5, and the red circles indicate quantum dots with fidelities ≤0.5. In both cases, the percentage of quantum dots with fidelity >0.5 is at least 15% of the overall quantum-dot field. This high concentration of quantum dots emitting entangled photons corresponds mainly to those sample areas where the substrate was not fully removed during back-etching. The density of quantum dots emitting polarization-entangled photons drops in the central areas (where the full substrate removal procedure was more effective), probably due to the prolonged etching, which might have degraded quantum-dot quality. This assumption is supported by the better statistics of the distribution of entangled photon emitters in the central area achieved in a successful reproduction of the sample.

Finally, another remarkable feature of our site-controlled quantum-dot family also worthy of mention is the uniformity of the emission energy. Figure 3b presents the emission energy distribution of the dots selected for the fidelity measurements. The total area of interest was 4×3 mm². Although the total standard deviation is 4.7 meV, over a short spatial range its distribution is reduced to ±2.5 meV (areas A and B).

In conclusion, our important finding is the high density of quantum dots that emit polarization-entangled photons without external manipulation of the electronic states. Areas containing at least 15% entangled photon emitters could be found easily. This is a major improvement over other types of epitaxial quantum dots, where only a small number of all the quantum dots can be direct emitters of entangled photons. This may enable large arrays of entangled photon-emitting units to be built that could be integrated into future quantum computation devices. Moreover, the control of pyramidal site-controlled quantum dots could, in the future, be enhanced by local metallic gates, which could be used to inject carriers and/or electronically manipulate excitonic states. The demonstrated apex-up geometry and micrometre-pitch pyramids are in fact beneficial for such a design. In addition, local strain manipulation could be utilized. Together, these techniques (and further epitaxial optimization) should allow the development of ‘perfect’ arrays of entangled photon emitters, where all devices would act as ‘good’ emitters, effectively contributing to the construction of real-world quantum computation with flying qubits.

**Methods**

**Quantum dot growth.** Quantum dots were grown by MOVPE in 7.5-μm-pitch tetrahedron recesses etched by a wet chemical etching procedure in a (111)B-oriented GaAs substrate. Standard MOVPE precursors, namely trimethylgallium, -indium, -aluminium), U-DMHY and arsine (AsH3), were used, in a reactor where tetrahedron recesses etched by a wet chemical etching procedure in a (111)B-

**Substrate removal.** A relevant issue for as-grown samples is the low photoluminescence extraction efficiency, which can lead to unfavourable experimental conditions. To overcome these problems, the substrate was selectively etched away, leaving an array of apex-up pyramids standing on a supporting substrate. The sample was attached by thermocompression gold-bonding to a (100)GaAs sample before the ‘all chemical’ substrate removal procedure was performed. The enhancement in photoluminescence extraction was due to the pyramidal shape, which acts as a lens, and also, in a minor way, to the reflecting gold layer under the base of the pyramid. In most cases, substrate removal is an essential procedure to obtain a significant photoluminescence signal from the quantum dots.

**Measurement and set-up details.** Photoluminescence data were taken using a conventional micro-photoluminescence set-up, which enabled access to individual quantum dots. The sample was cooled to 7 K by a closed-cycle, low-vibration microphotoluminescence helium cryostat (ARS cryo). Quantum dots were excited non-resonantly with a semiconductor laser diode emitting at 635 nm at a repetition rate of 40 MHz. Backscattered light was collected through a 30° objective with a numerical aperture of 0.55. Exciton and biexciton transitions were filtered for correlation measurements by two monochromators using spectral resolutions of 0.5 meV and 0.2 meV, respectively. Each filtered transition was divided by a polarizing beamsplitter and sent to silicon avalanche photodiodes (APDs). Typical numbers of counts per second during the measurements of entangled photons were 3,000 and 700 for exciton and biexciton transitions, respectively, yielding the detection of ~810 entangled photon pairs per hour. Four synchronized sequences of APD signals were fed to a photon-counting module and analysed to build four correlation curves. Such a four-detector set-up allows a complete polarization basis to be measured at once, increasing the precision of the quantum state tomography procedure by reducing statistical errors (for example, errors present due to sample drifting or excitation intensity fluctuations), which are harder to avoid during the long experimental procedure necessary if only a single polarization state intensity is considered at a time. The intensity of each of the states directly involved in correlation analysis (18 basis) was obtained by correlating second-order correlation function values to probabilities. Polarization bases were chosen by using an appropriate combination of half- or quarter-waveplates and polarizers.

The FSS was measured by placing a polarizer in front of the spectrometer entrance and rotating the half-waveplate by fixed steps. The corresponding excitation and birefringence transitions were fitted by Lorentzian and Lorentz functions. Two peaks were plotted as a function of polarization angle. In the presence of small FSS, the curves typically followed a sinusoidal function from which the FSS value could be obtained directly. Such a fitting procedure, combined with the 18 μeV experimental resolution, improved our total measurement resolution. The second-order correlation function value was obtained by dividing all events at zero delay peak (from ~12.5 ns to 12.5 ns) by the mean value of events at the side peaks. For the calculations, raw data were used without background subtraction. The fidelity $F$ of the entangled state $|\psi\rangle = \frac{1}{\sqrt{2}}(|H\psi\rangle + |V\psi\rangle)$ was calculated from the degree of correlation $C_{\text{XX}}$ obtained in the $(|H\rangle, \text{diagonal})$ and $(|H\rangle, |V\rangle)$ polarization bases$^{27}$; $C_{\text{XX}} = \langle g_{\text{XX}} \rangle - g_{\text{XX}}^0$, where $g_{\text{XX}} = \langle |XX\rangle \rangle$ and $g_{\text{XX}}^0 = \langle |XX\rangle \rangle$. Entanglement was confirmed when $F > 0.5$ (if $|C_{\text{XX}}^0| < |C_{\text{XX}}|$, as a rule of thumb, $g_{\text{XX}}^0 > 2g_{\text{XX}}^0$, for the bases where bunching is expected to co-polarized photons).

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Author contributions
G.J. carried out optical characterization of the samples and data analysis, and wrote the manuscript with E.P. L.O.M. assisted in optical characterization and data analysis. V.D. and A.G. participated in the production of the samples, processing and microscopy characterization. E.P. conceived the study, participated in its design and coordination, and contributed to writing the manuscript. All authors commented on the final manuscript.

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