Surface quantum dots with pure, coherent, and blinking-free single photon emission

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The surface of semiconductor nanostructures has a major impact on their electronic and optical properties1. Disorder and defects in the surface layer typically cause degradation of charge carrier transport and radiative recombination dynamics. However, surface vicinity is inevitable for many scalable nano-optical applications2,3. Epitaxially grown quantum dots are the best candidate for high-performance single photon emission and show great potential for quantum technologies4. Yet, these emitters only reveal their excellent properties if they are deeply embedded in a semiconductor host5–6. Until today, quantum dots close to surfaces yield weak, broad, and unstable emissions7–10. Here, we show the complete restoration of optical properties from quantum dots grown directly on a semiconductor surface. The vanishing luminescence from the as-grown sample turns into bright, ultra-stable, coherent and blinking-free single photon emission after sulphur passivation. Under quasi-resonant excitation, single photons are generated with 98.8% purity, 77% indistinguishability, linewidths down to 4 µeV and 99.69% persistency across 11 orders of magnitude in time. The emission is stable even after two years and when being subjected to nanomanufacturing processes. Some long-standing stumbling blocks for surface-dominated quantum dots are thereby removed, unveiling new possibilities for hybrid nano-devices11–13 and applications in quantum communication14 or sensing15.

Surface passivation is a key feature of semiconductor technology. The technique was developed at Bell Labs in the 1950s and laid the foundation for today’s silicon-based integrated circuit architectures16. With the advent of optoelectronics, surface treatments have been extended to III–V semiconductors in recent decades17,18. Dangling bonds at the surface typically lead to reduced device performance caused by electron- and hole-trap states within the bandgap1. Surface passivation, either chemical or physical, provides an inert surface layer that saturates dangling bonds, resulting in fewer non-radiative recombination processes and increased photoluminescence yield19.

Novel quantum- and nano-photonic architectures6 commonly feature large surface-to-volume ratios, where surface passivation can play a revolutionary role. Recent works with transition metal dichalcogenide and perovskite nanostructures show that chemical passivation reduces surface states and enhances the optical emission20,21. However, the long-term stability and optical quality of these materials have yet to be significantly improved for use in quantum photonic devices at the single photon level. Today, arguably the most promising semiconductor materials for such applications are epitaxially grown quantum dots (QDs) based on GaAs22,23. They are outstanding sources of indistinguishable single photons and polarization-entangled photon pairs, with a combination of metrics (brightness, purity, coherence, emission rate) that no other source can match24. QDs are typically buried under thick barrier layers to maintain a distance from detrimental surface states. However, to unlock their full potential,
QDs need to be integrated into scalable (hybrid) nanophotonic devices that require a vanishing distance to the surface. For instance, investigating strong coupling to optical cavity fields requires ultra-small mode volumes, and efficient interaction with plasmonic or dielectric nanoparticles can only be achieved within a few nanometers. Other envisioned applications include quantum microscopy and single-photon transistors. Surface proximity of QDs is inevitable in these cases, which is why several groups have been studying single epitaxial QDs very close to the surface for over two decades. Weak and broad emissions were the result, even after surface passivation, rendering advanced quantum optical experiments infeasible.

In this work, we report on the complete restoration of optical properties via chemical passivation of GaAs QDs grown directly on top of an AlGaAs surface, resulting in bright, pure, coherent, indistinguishable, and blinking-free single photon emissions. The high optical quality survives common nanofabrication techniques and is stable for years.

**Growth, passivation and surface composition**

The sample is grown on a GaAs (001) substrate by molecular beam epitaxy via in-situ local droplet etching, as illustrated in Figure 1a: After shortly interrupting the supply of As$_4$ to create a low arsenic environment, Al is deposited and forms droplets, liquefying the Al$_{0.23}$Ga$_{0.77}$As surface beneath. Due to the atomic concentration gradients between the surface layer and the Al droplets, interdiffusion of As occurs. Supply of As$_4$ then ensures the complete formation of symmetric nanoholes on the surface. Ga and As$_4$ are then deposited together, forming GaAs which migrates into the nanoholes. Charge carriers in the GaAs QDs are now energetically confined by the bottom Al$_{0.23}$Ga$_{0.77}$As layer and by vacuum/air at the top (see Fig. 1b).

**Figure 1: Sample fabrication and morphology.** (a) In-situ Al droplet etching and nanohole infilling are performed on an AlGaAs epi-ready surface, followed by chemical passivation with ODT. (b) Schematic of the sample cross-section (not to scale). (c), (d) AFM images before and after ODT passivation, respectively.

The surface morphology is studied via atomic force microscopy (AFM) and reveals QD containing structures on the surface (Fig. 1c), which are elongated because of the preferential migration of group V atoms along the [1 $\overline{1}$ 0] direction. High growth quality is evident from clear atomic monolayer fluctuations and the extremely low root-mean-square (rms) surface roughness of 0.086 nm. To eliminate surface states, the sample is then passivated with 0.05 mol/L 1-Octadecanethiol (ODT)/ethanol solution for 3 hours at room temperature, forming a protective, self-assembled monolayer of ODT. As can be seen in Fig. 1d, the general surface morphology is preserved and shows only minor changes due to the ODT molecules with their relatively long (18) carbon chains.
An in-depth comparison is now performed between the as-grown (unpassivated) and passivated samples. In contrast to the slight change in surface morphology, drastic changes appear in the optical properties at a temperature of 4 K. The as-grown sample exhibits very weak emissions as shown by the photoluminescence (PL) intensity map displayed in the upper panel of Fig. 2a. However, after surface passivation, the PL yield is enhanced by over one order of magnitude (lower panel of Fig. 2a). Localized intensity maxima are seen throughout the entire sample area, which is a hallmark for the emission of single QDs. Such a drastic change is most plausibly explained by a strong reduction of surface states.

Structural defects in the surface can induce trap states in the band gap as well as band bending by Fermi level pinning (see inset of Fig. 2b), which is detrimental to the QD emission. To understand the influence of ODT passivation on the bonding arrangements at the surface, X-ray photoelectron spectroscopy (XPS) is performed on both samples. Figure 2b shows the XP spectra in the Ga 2p3/2 region, where Ga-O bonds are detected besides Ga-As bonds. This is to be expected, since the surface oxidation of GaAs is an exothermic reaction that proceeds easily under ambient conditions. The XPS spectra after passivation show little difference and feature only a slight reduction of surface oxides. Contributions of newly formed Ga-S bonds (yellow, dashed line) may arise, but are difficult to model due to the strong overlap with the Ga2O3 peak. Since a high PL yield is observed after passivation despite the oxides, most oxide layers can be assumed to exhibit high crystallinity and thus not lead to trap states. However, it is known that such states can form at the surface by point defects, unsaturated dangling bonds of Ga and As atoms, and As-As dimer bonds. The incorporation of sulphur during ODT passivation induces surface reconstruction and the formation of Ga-S to pair the outer-shell electrons, flattening the electronic bands and the QD potential.

**Photoluminescence characteristics**

We now focus on the spectroscopy of single QDs. As shown in Fig. 2c, the emissions from the untreated sample remain faint and broad due to the notoriously high density of surface states, while the passivated sample exhibits bright photoluminescence and narrow transition lines from single QDs. The PL signal compares very well to the fingerprint emission of high quality deeply-buried GaAs QDs, showing strong emissions from neutral exciton (X) and biexciton (XX) as well as several charged excitonic complexes. By performing eight-band $k \cdot p$ theory in combination with the configuration interaction method, we confirm that the exciton energies of QDs at the surface are indeed not expected to change significantly compared to embedded QDs (see supplemental). To enhance the photon extraction efficiency, a glass solid immersion lens (OHARA, S-LAH79) is placed on the passivated sample. The neutral exciton emission is then guided to a single-photon detector, and the raw count rate measured for varying cw excitation laser power (inset of Fig. 2c). A saturated brightness of 1.4 million photon counts per second (cps) is obtained, which compares favourably to that of state-of-the-art buried QDs under a higher refractive index GaP SII. Increased photon extraction is expected for QDs close to the surface of high-refractive index semiconductors, as confirmed by finite-difference time-domain (FDTD) simulations (see supplemental).

Today, most of the colloidal and epitaxial QDs (especially under resonant excitation) exhibit intermittency in their luminescence and therefore, reduced quantum yield. The PL time traces in Fig. 2c reveal clear blinking and spectral jittering of the emissions from the as-grown sample, confirming the existence of tremendous trap states. After passivation, neither blinking nor spectral jittering is observed on timescales from seconds to minutes, in strong contrast to the common observations in other (near-)surface quantum emitters. The bright, narrow-band and photostable emissions suggest that the typical blinking mechanisms (i.e., QD ionization-neutralization or non-radiative Auger recombination of charged excitons) are suppressed by the passivation. These findings represent the first experimental observation of bright and blinking-free emissions from single epitaxial QDs at the surface.
Figure 2: Properties of the surface QDs before (top) and after passivation (bottom). (a) PL intensity map of the samples at 4K. Faint emissions of the as-grown sample turn into bright and localized emissions after passivation. (b) XP spectra display dominating oxide contributions in the as-grown surface. The passivation slightly reduces Ga-O bonds and adds possible Ga-S bonds. Inset: Schematics of the electronic structure of surface QDs. Passivation removes surface states in the band gap and thereby flattens the electronic bands. (c) High-resolution micro-PL spectra, originally showing weak and broadened emissions. After passivation, high-quality single QD emissions appear, as manifested in various narrow and bright excitonic peaks. Inset: Count rate of the neutral exciton (X) on a single photon detector reaches 1.4 million cps with increased excitation laser power. Below each spectrum, PL intensity time traces over 90s are shown in steps of 1s. Blinking and spectral jitter is observed for the weak emissions of the as-grown sample, whereas passivation leads to highly stable, localized emissions without any blinking, photobleaching, or spectral jittering.

Quantum optical properties

Because of their surface proximity, it is meaningful to examine the sources of noise affecting these QDs. To this purpose, we perform resonant excitation and obtain the noise power spectral density from the intensity time traces of the emitted signal. First, a cw laser is used to resonantly excite the positively charged exciton (X+) in the QD. The resonant laser light and the X+ zero-phonon line are then spectrally blocked by notch filters, and the acoustic phonon sideband signal is collected. Figure 3a shows the detected intensity over the spectral detuning of the excitation laser light. A Lorentzian line shape of the X+ transition is observed, with a narrow linewidth $\Delta \nu = 7.0 \pm 0.1$ µeV. The sources of noise in these QDs are now investigated by obtaining the relative noise power spectral densities at two different laser detuning settings ($\delta = 0$ and $\delta = \Delta \nu / 2$) and subtracting the respective shot noise. As shown in Fig. 3b, low noise powers are observed at $\delta = 0$, slightly decreasing further towards higher frequencies. For $\delta = \Delta \nu / 2$, more obvious noise is visible up to frequencies of several hundred Hz, after which it decreases by over one order of magnitude below the shot-noise limit. This part follows a Lorentzian spectrum, characteristic for an ensemble of two-level fluctuators of local electric fields near the QD. Such charge noise is frequently observed in buried QDs, which is why this type of noise cannot be removed by eliminating the surface states alone. The presence of strong local magnetic field fluctuations (spin noise) would contribute mostly to the noise at $\delta = 0$, which is not observed here. However, the source of the remaining noise at higher frequencies is yet unknown and may be of spin origin, on which future magnetic field dependent measurements may shed light on.
Figure 3: Low noise, blinking free and coherent single photon emission from surface QDs. (a) Acoustic phonon sideband intensity over laser detuning with respect to the $X^+$ resonance, following a Lorentzian line shape of width $\Delta \nu = 7.0 \pm 0.1 \, \mu\text{eV}$. (b) Noise spectra recorded at a detuning of $\delta = 0$ (red) or $\delta = \Delta \nu / 2$ (blue) reveal charge noise at frequencies $< 1 \, \text{kHz}$. The dotted lines correspond to the shot noise limit of the respective measurement. (c) Normalized second-order autocorrelation function for the quasi-resonantly excited $X^+$ in a surface QD. The absence of bunching on time delays from nanoseconds to 10 milliseconds indicates a blinking-free emission, with an extracted on-fraction of $99.69 \pm 0.02\%$. Inset: Second-order autocorrelation on nanosecond timescales reveals pure single photon emission with $g^2(0) = 0.0121 \pm 0.0007$. (d) Hong-Ou-Mandel interference between consecutively emitted $X^+$ photons in the co-polarized (blue) and cross-polarized (black) configurations. The model (red) reveals a single-photon indistinguishability of $I = 0.77 \pm 0.06$. The inset shows a schematic of the optical setup, comprising an unbalanced Mach-Zehnder interferometer, followed by two single photon detectors and a time-correlated single-photon counting unit (TCSPC). (e) Fluorescence lifetime measurement data and theoretical model of a three-level cascade decay results in $T_1(X^+) = 443.1 \pm 1.5 \, \text{ps}$. (f) First order coherence of the $X^+$ emission over the time delay in a Michelson interferometer. The model (red solid curve) reveals a coherence time of $T_2 = 330.0 \pm 9.7 \, \text{ps}$ (corresponds to $\Delta \nu = 4.0 \pm 0.1 \, \mu\text{eV}$). Inset: Histogram of $X^+$ coherence times from 26 representative QDs of the sample.

The superior emission characteristics allow for quantum optical experiments at the single-photon level. On-demand single photon generation from a different surface QD is now achieved by pulsed (80 MHz) quasi-resonant p-shell excitation of the positively charged $X^+$ state. To assess the single-photon purity and possible blinking mechanisms, second-order autocorrelation measurements are performed using a Hanbury Brown and Twiss setup. The absence of coincidences at zero-time delay (see inset of Fig. 3c) indicates pure single-photon emission, with a raw value of $g^2(0) = 0.0121 \pm 0.0007$, corresponding to a single-photon purity of over 98.8\%.

Unlike colloidally synthesized and unpassivated surface emitters, epitaxial QDs exhibit blinking on much faster time scales. Charge fluctuations in the QD are the most common cause for blinking in (quasi-)resonant excitation schemes, revealed by a characteristic bunching in the second-order autocorrelation ($g^2(0) > 1$)\(^3\). Deterministic QD charging is achievable with charge-tunable diode structures, which recently led to the demonstration of the first blinking-free single-photon emission from GaAs QDs under resonance fluorescence conditions\(^2\). Since the surface QDs in this work are not embedded in such a structure, strong PL blinking is intuitively expected. As shown in Fig. 3c, the coincidence...
histogram is now calculated with logarithmically spaced bins, on the timescale from nanoseconds to milliseonds. No obvious bunching is observed on any time scale. By fitting the auto-correlation data\textsuperscript{43} (see supplemental) an emission on-fraction $\beta = 99.69 \pm 0.02\%$ is obtained, suggesting that the electronic ground is dynamically occupied by exactly one single hole in a very efficient manner. This blinking-free emission of surface QDs is striking, as it is even untypical for state-of-the-art epitaxial QDs with thick barriers.

Now the single-photon indistinguishability is determined, a crucial parameter for the manifold applications involving photonic quantum gate operations\textsuperscript{44}. We first compare photons emitted with a time separation of $\Delta t = 3$ ns, and thereby bypassing the low-frequency ($<1$ kHz) charge noise in the surface QDs (Fig. 3b). The single-photon stream is sent to a Hong-Ou-Mandel interferometer (inset of Fig. 3d) comprising a half-wave plate (HWP) to define the relative polarization state of the interfering photons. As expected, co-polarized photons result in anti-bunching, whereas cross-polarized photons are distinguishable and therefore give rise to coincidences at zero-time delay. An indistinguishability of $I = 0.77 \pm 0.06$ is extracted, which is mostly limited by the excitation mechanism: At quasi-resonant excitation, a higher energetic form of the charged exciton is first populated. This “hot” charged exciton $X^+_G$ relaxes to the charged exciton state $X^+$ and subsequently decays radiatively to the single positively charged ground state of the QD (inset of Fig. 3e). A theoretical model of a three-level cascade decay\textsuperscript{43} convoluted with the detector response function is therefore used to fit the fluorescence lifetime data in Fig. 3e. The $X^+_G$ exhibits a lifetime of $T_1(X^+_G) = 88.1 \pm 0.7$ ps and for the radiative lifetime of $X^+$ we extract $T_1(X^+) = 443.1 \pm 1.5$ ps. It is known that in cascade decays the photon indistinguishability is intrinsically limited by the entangled nature of the emission\textsuperscript{45,46}. The upper limit of the indistinguishability can therefore be estimated to $I_{\text{limit}} = \Gamma(X^+_e)/(\Gamma(X^+_h) + \Gamma(X^+_+)) = 83 \%$, with the radiative decay rates $\Gamma(X^+_h) = 1/T_1(X^+_h)$ and $\Gamma(X^+_+) = 1/T_1(X^+_+)$.

The application of different surface QDs in interference-based quantum gate operations requires a coherent single photon emission on long timescales (e.g., seconds). The respective indistinguishability $I_{\text{long}} = T_2/T_1$ is now sensitive to low-frequency charge noise which affects the coherence time $T_2$.

Theoretical modelling of the single-photon interference visibility (Fig. 3f) in a Michelson interferometer yields a coherence time of $T_2 = 330.0 \pm 9.7$ ps, which corresponds to a Lorentzian line shape of width $\Delta \nu = 4.0 \pm 0.1$ $\mu$eV. The indistinguishability on long timescales is therefore $I_{\text{long}} = 0.37 \pm 0.03$, which allows for interfering single photons from different QDs. This value is comparable to deeply-buried GaAs QDs, and could be improved further by employing a strictly resonant excitation scheme or using a n-i-p diode to reduce charge noise. We confirm that the photon coherence is representative for the whole sample by measuring 26 QDs, resulting in an average of $T_2 = 276 \pm 175$ ps ($\Delta \nu = 5 \pm 3$ $\mu$eV).

**Device stability**

Surface QDs have potential advantages for application in nanophotonic hybrid systems\textsuperscript{12}. However, one may worry if they are robust enough during the device fabrication because of their immediate proximity to the environment. Therefore, we use electron beam lithography and gold deposition to define markers (central sketch of Fig. 4), which is the first step for reproducibly fabricating hybrid devices. By detecting the markers, the same pre-selected QDs can be readily identified in AFM measurements (Fig. 4a), photoluminescence intensity maps (Fig. 4b), or other microscopy techniques for further processing. Before fabrication, the surface QDs were passivated with ODT, showing narrow and strong optical emissions (Fig. 4c). Subsequent spin-coating, exposure, developing, and deposition steps expose the surface to a harsh environment, introducing trap states. Therefore, the PL intensity degrades by over 50% and the exciton peaks are significantly broadened (e.g., a linewidth of 145.2 $\pm$ 5.0 $\mu$eV in the

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second panel of Fig. 4c). After isopropanol cleaning, which can dissolve the ODT monolayer on the surface, no QD emission is detected at all. However, the PL can be fully restored by another passivation step, making the entire process robust and highly reproducible.

Figure 4: Roadmap for the application of surface QDs. (a) AFM image and (b) PL intensity map of passivated surface QDs, showing the same area on a device with fabricated gold markers (sketch in the center). Inset: light microscopy image showing four markers (adjacent markers separated by 20 μm) together with QD emissions. The red square denotes the area displayed in (a) and (b). (c) PL spectra of representative QDs that were collected after each fabrication step. The high-quality signals before the fabrication can be retrieved by passivating the sample a second time with ODT after fabrication. (d) Comparison of PL spectra from representative QDs of the same passivated sample before and after two years (vertically shifted for clarity), showing no visible degradation of the emission. Inset: comparison of X linewidths of the corresponding samples. After two years, the average linewidth (resolution limited by setup) shows only a slight increase from 19.6 ± 4.7 µeV (40 QDs measured) to 29.2 ± 4.0 µeV (20 QDs measured).

The long carbon chain of ODT with heavy molecular weight enables the formation of an inert layer over the QDs, making the surface very stable even after long exposure to air. Figure 4d shows a PL spectrum measured with a passivated sample in April 2020. After two years in ambient conditions (May 2022) the sample shows nearly the same spectra. The X linewidths are statistically evaluated (inset of Fig. 4d) using different QDs that are excited via above-band excitation. Only a slight broadening is observed after two years, from 19.6 ± 4.7 µeV to 29.2 ± 4.0 µeV. Here, the spectrometer resolution in the range of 10-15 µeV and possible X fine structures impose a lower bound to the measured linewidth. The surface QDs therefore do not suffer severely from aging, showing remarkable long-term stability that is on par with the deeply buried QDs.

Conclusion and Outlook

Today, it is a common understanding that high-quality quantum emitters must be capped\(^1\), to protect them from surface states. In this work, we show that high-quality material growth in combination with a suitable passivation step can effectively eliminate surface states, resulting in a single-photon emission with unprecedented brightness, coherence and absence of blinking from nanoseconds to tens of seconds. This represents an important breakthrough for both colloidal\(^8\) and epitaxial\(^22\) QD communities, since understanding and controlling coherence and blinking has been a daunting task for over 20 years\(^18\). Deeper understanding of this system can be gained by scrutinizing the structural properties of the surface and the mechanisms behind the fast charging of the QD ground state under quasi-resonant excitation. The surface QDs are stable in air and withstand demanding device fabrication. Many exciting
experiments, which require the closest interaction with a high-quality quantum emitter, are therefore unlocked, e.g., coupling to Mie-resonant dielectric particles or realizing strong light-matter interaction within an extremely small plasmonic nanocavity.

Methods

Sample fabrication. After growing 238 nm Al_{0.23}Ga_{0.77}As, the sample is annealed under As_4 flux for 10 seconds. Next, the As_4 is stopped for 15 seconds, followed by 0.22 nm Al deposition to etch nanoholes on the surface. After infilling the nanoholes with 2 nm GaAs on the AlGaAs surface, the sample is annealed for another 2 minutes under As_4 flux. Then the sample was cooled down from 620 °C to 350 °C with rate of 220 °C/min and transferred out of the growth chamber of the molecular beam epitaxy machine. ODT solution was prepared by dissolving 146 mg of 98% ODT (Sigma-Aldrich) in 10 ml of 99.8% ethanol (Roth). For passivation, the as-grown sample was directly immersed in ODT solution for 3 hours at room temperature and dried naturally.

Optical excitation. The optical measurements were performed at T = 4 K with the sample in a closed-cycle helium cryostat. The PL spectra were obtained under cw excitation with 675 nm laser light, which has been focused by an objective of NA = 0.7 to irradiate a single QD and collect its emission. For obtaining the PL intensity maps, the sample was excited using a light emitting diode emitting at 455 nm, and the QD emissions were collected with a CCD camera. The optimum quasi-resonant excitation conditions were found by performing photoluminescence excitation experiments with a step size of 0.002 nm and using a cw Ti:sapphire laser. Second-order autocorrelation, lifetime and indistinguishability measurements were carried out by exciting the QD using a pulsed Ti:sapphire laser. Hereby, the laser signal with 80 MHz repetition rate and pulse duration of 140 femtoseconds was pulse-shaped to a spectral width of 37 GHz. The QD signal was collected and coupled into a polarization-maintaining single-mode fibre and sent to a superconducting nanowire single photon detector.

Indistinguishability measurements. The QD signal was guided to a free space, unbalanced Mach-Zehnder Interferometer comprising a time delay of 3 ns. The time difference between the two consecutively emitted photons was matched to the interferometer delay by carefully tuning the time differences in the excitation pulses.

Noise measurements. The resonant cw laser light and the X⁺ zero-phonon line are spectrally blocked by notch filters with a spectral width of <0.6nm. The acoustic phonon sideband signal is collected and coupled to a polarization-maintaining single-mode fibre and sent to an avalanche photodiode. The relative noise power spectral densities are obtained at two different laser detuning settings (δ=0 and δ=Δν/2) and subtracted by the respective shot noise limit. The count rates from the phonon sideband signal are 96.4 kHz and 49.2 kHz on the single photon detector, respectively. The distinct peaks in Fig. 3b between 80 Hz and 350 Hz are attributed to noise in the setup and electronics.

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Author contributions

F.D. conceived the project. M.Z. and F.D. supervised the whole research. X. C., E. R. and R. K. were responsible for sample growth. X. C., P. L. performed surface passivation. X. C., B. B. and R. H. carried out the AFM measurements. P. L. was responsible for XPS characterization and data analysis. J. Y., X. C. and M. Z. performed most of the optical measurements and data interpretation. T. F. was responsible for noise spectra analysis with support from M.O. C. M fabricated gold markers. X. C., C. M performed optical imaging. Y. Z. supported the optical measurements and Z. A. the RF measurement. V. K. simulated the single-particle and exciton energies. M. Z. designed the sample fabrication scheme. F. B. worked on theoretical models for second-order correlation and noise spectra. C. S. performed FDTD simulations. X. C., M. Z. and F. D. wrote the manuscript with input from all other authors.

Competing financial interests

The authors declare no competing financial interests.

References:

1. Boles, M. A., Ling, D., Hyeon, T. & Talapin, D. V. The surface science of nanocrystals. Nat. Mater. 15, 141–153 (2016).
2. García de Arquer, F. P. et al. Semiconductor quantum dots: Technological progress and future challenges. Science (80-. ). 373, eaaz8541 (2022).
3. Lodahl, P., Mahmoodian, S. & Stobbe, S. Interfacing single photons and single quantum dots with photonic nanostructures. Rev. Mod. Phys. 87, 347–400 (2015).
4. Lu, C.-Y. & Pan, J.-W. Quantum-dot single-photon sources for the quantum internet. Nat. Nanotechnol. 16, 1294–1296 (2021).
5. Tomm, N. et al. A bright and fast source of coherent single photons. Nat. Nanotechnol. 16, 399–403 (2021).
6. Senellart, P., Solomon, G. & White, A. High-performance semiconductor quantum-dot single-photon sources. Nat. Nanotechnol. 12, 1026–1039 (2017).
7. Lee, S. F. & Osborne, M. A. Brightening, Blinking, Bluing and Bleaching in the Life of a Quantum Dot: Friend or Foe? ChemPhysChem 10, 2174–2191 (2009).
8. Efros, A. L. & Nesbitt, D. J. Origin and control of blinking in quantum dots. Nat. Nanotechnol. 11, 661–671 (2016).
9. Heyn, C. et al. Droplet etched GaAs quantum dots close to surfaces and metallic interfaces. J. Appl. Phys. 121, 044306 (2017).
10. Wang, C. F. et al. Optical properties of single InAs quantum dots in close proximity to surfaces. Appl. Phys. Lett. 85, 3423–3425 (2004).
11. Kurizki, G. et al. Quantum technologies with hybrid systems. Proc. Natl. Acad. Sci. 112, 3866–3873 (2015).
12. Chang, D. E., Sørensen, A. S., Demler, E. A. & Lukin, M. D. A single-photon transistor using nanoscale surface plasmons. Nat. Phys. 3, 807–812 (2007).
13. Yeo, I. et al. Strain-mediated coupling in a quantum dot–mechanical oscillator hybrid system. Nat. Nanotechnol. 9, 106–110 (2014).
14. Gisin, N. & Thew, R. Quantum communication. Nat. Photon. 1, 165–171 (2007).
15. Marciniak, C. D. et al. Optimal metrology with programmable quantum sensors. Nature 603, 604–609 (2022).
16. Frosch, C. J. & Derick, L. Surface Protection and Selective Masking during Diffusion in Silicon. J. Electrochem. Soc. 104, 547–552 (1957).
17. Sandroff, C. J., Hegde, M. S., Farrow, L. A., Chang, C. C. & Harbison, J. P. Electronic passivation of GaAs surfaces through the formation of arsenic - sulfur bonds. Appl. Phys. Lett. 54, 362–364 (1989).
18. Hasegawa, H. & Akazawa, M. Surface passivation technology for III–V semiconductor nanoelectronics. Appl. Surf. Sci. 255, 628–632 (2008).
19. Lunt, S. R., Ryba, G. N., Santangelo, P. G. & Lewis, N. S. Chemical studies of the passivation of GaAs surface recombination using sulfides and thiols. J. Appl. Phys. 70, 7449–7467 (1991).
20. Amani, M. et al. Near-unity photoluminescence quantum yield in MoS2. Science (80- ). 350, 1065–1068 (2015).
21. Utzat, H. et al. Coherent single-photon emission from colloidal lead halide perovskite quantum dots. Science (80- ). 363, 1068–1072 (2019).
22. Zopf, M. et al. Entanglement Swapping with Semiconductor-Generated Photons Violates Bell’s Inequality. Phys. Rev. Lett. 123, 160502 (2019).
23. Huber, D. et al. Strain-Tunable {GaAs} Quantum Dot: A Nearly Dephasing-Free Source of Entangled Photon Pairs on Demand. Phys. Rev. Lett. 121, 33902 (2018).
24. Zhai, L. et al. Low-noise GaAs quantum dots for quantum photonics. Nat. Commun. 11, 4745 (2020).
25. Reithmaier, J. P. et al. Strong coupling in a single quantum dot–semiconductor microcavity system. Nature 432, 197–200 (2004).
26. Pfeiffer, M. et al. Eleven nanometer alignment precision of a plasmonic nanoantenna with a self-assembled GaAs quantum dot. Nano Lett. 14, 197–201 (2014).
27. Balasubramanian, G. et al. Nanoscale imaging magnetometry with diamond spins under ambient conditions. Nature 455, 648–651 (2008).
28. Manna, S. et al. Surface passivation and oxide encapsulation to improve optical properties of a single GaAs quantum dot close to the surface. Appl. Surf. Sci. 532, 147360 (2020).
29. Chellu, A. et al. GaAs surface passivation for InAs/GaAs quantum dot based nanophotonic devices. Nanotechnology 32, 130001 (2021).
30. Keil, R. et al. Solid-state ensemble of highly entangled photon sources at rubidium atomic transitions. Nat. Commun. 8, 15501 (2017).
31. Budz, H. A. & LaPierre, R. R. Properties of octadecanethiol self-assembled monolayers deposited on GaAs from liquid and vapor phases. J. Vac. Sci. Technol. A 26, 1425–1431 (2008).
32. Komsa, H.-P. & Pasquarello, A. Dangling bond charge transition levels in AlAs, GaAs, and InAs. Appl. Phys. Lett. 97, 191901 (2010).
33. Zhang, Z. & Jr. Yates, J. T. Band bending in semiconductors: Chemical and physical consequences at surfaces and interfaces. Chem. Rev. 112, 5520–5551 (2012).
34. Laukkanen, P. et al. Passivation of III–V surfaces with crystalline oxidation. Appl. Phys. Rev.
35. Cuypers, D. et al. Sacrificial Self-Assembled Monolayers for the Passivation of GaAs (100) Surfaces and Interfaces. Chem. Mater. 28, 5689–5701 (2016).

36. Huber, D. et al. Single-particle-picture breakdown in laterally weakly confining GaAs quantum dots. Phys. Rev. B 100, 235425 (2019).

37. Nie, W. et al. Experimental optimization of the fiber coupling efficiency of GaAs quantum dot-based photon sources. Appl. Phys. Lett. 119, 244003 (2021).

38. Yang, J. et al. Photoneutralization of charges in GaAs quantum dot based entangled photon emitters. Phys. Rev. B 105, 115301 (2022).

39. Curto, A. G. et al. Unidirectional Emission of a Quantum Dot Coupled to a Nanoantenna. Science (80-. ). 329, 930–933 (2010).

40. Hansom, J., Schulte, C. H. H., Matthiesen, C., Stanley, M. J. & Atatüre, M. Frequency stabilization of the zero-phonon line of a quantum dot via phonon-assisted active feedback. Appl. Phys. Lett. 105, 172107 (2014).

41. Simon, C. & Poizat, J. P. Creating single time-bin-entangled photon pairs. Phys. Rev. Lett. 94, 030502 (2005).

42. Schöll, E. et al. Crux of Using the Cascaded Emission of a Three-Level Quantum Ladder System to Generate Indistinguishable Photons. Phys. Rev. Lett. 125, 233605 (2020).

43. Reindl, M. et al. Highly indistinguishable single photons from incoherently excited quantum dots. Phys. Rev. B 100, 155420 (2019).

44. Niroomand, M. et al. Fluorescence intermittency in single cadmium selenide nanocrystals. Nature 383, 802–804 (1996).

45. Baranov, D. G. et al. All-dielectric nanophotonics: the quest for better materials and fabrication techniques. Optica 4, 814–825 (2017).

46. Chikkaraddy, R. et al. Single-molecule strong coupling at room temperature in plasmonic nanocavities. Nature 535, 127–130 (2016).