Enhanced telecom emission from single group-IV quantum dots by precise CMOS-compatible positioning in photonic crystal cavities

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I Simulation of photonic crystal cavity modes

Supplementary Fig. S1 presents the electric-field energy density profiles for five L3 photonic crystal resonator (PCR) modes as calculated by the finite-difference time-domain (FDTD) solver MEEP. The cavity design is optimized for a high Q factor of the ground mode M0 as proposed in Ref.¹. In agreement with earlier simulations²³, each mode shows a unique spatial field energy pattern, and requires a specific dot position for an optimized coupling, in agreement with the experimental photoluminescence (PL) data presented in our work. Note that the well-known polarization states of the L3 modes indicated in the plot enable an unambiguous identification of the observed spectral emission peaks with the simulated cavity modes.

II Fabrication process

In the first processing step (Supplementary Fig. S2a), alignment marks are etched deep into the buried oxide (BOX) of the custom silicon-on-insulator (SOI) substrate. They are defined by electron beam lithography (EBL) in a Raith eLine Plus facility equipped with a laser stage that allows for an alignment precision in the 10 nm range. Anisotropic etching into the BOX is performed at cryogenic temperatures in an inductively coupled (ICP) Oxford 100 reactive ion etcher (RIE) employing an SF₆/O₂ process⁴.

In the second step (Supplementary Fig. S2b), a set of pits for the preferential nucleation of QDs⁵⁷ is fabricated by EBL and a shallow (35±5 nm deep) ICP-RIE process. Complete arrays of pits with twice the periodicity of the air hole patterns for the PCRs are realized in this lithography level. Only the one pit that defines the QD position in each of the subsequently processed cavities is slightly moved out of registry in a series of ten adjacent PCR structures. In this way, the cavity QD is shifted systematically between the center and the terminating air hole at one edge of the L3 cavity. The alignment marks are used for positioning of the pit-pattern mask, thus defining a reference frame that will be employed again for the later PCR fabrication step. Writing complete pit arrays with comparably small periods is essential for tight size control and perfect ordering of the QDs⁶.

In the third processing step (Supplementary Fig. S2c) the epitaxial layers are deposited. The samples are prepared for epitaxy by resist removal in organic solvents and an oxygen-plasma followed by an RCA cleaning procedure⁸. A final treatment in 1% hydrofluoric acid (HF) removes the natural oxide and provides a hydrogen-terminated surface that stabilizes the pit pattern during subsequent heat treatments⁹. The templates are then immediately transferred into the load-lock chamber of a Riber SIVA 45 molecular beam epitaxy (MBE) system and subsequently degassed in the ultra-high vacuum environment of the growth chamber at 750°C for 45 min. The epitaxial layer sequence consists of a 40 nm thick Si buffer, 0.66 nm of Ge and a 110 nm thick Si cap. During buffer growth the substrate temperature is ramped from 450 to 550°C and is then kept constant at 700°C for Ge deposition. Ge growth leads to
Fig. S 1. Simulated cavity modes of an ideal L3 resonator. The electric-field energy density distributions of cavity modes M0 to M4 are shown in a normalized contour plot, and the respective calculated mode resonance wavelength, $Q$-factor and dominant polarization state are indicated.

an approximately three monolayer (ML) thick, Ge-rich WL that covers the entire sample\textsuperscript{10}, and arrays of perfectly ordered Ge QDs which nucleate at the centers of the predefined pits\textsuperscript{6}.

Two versions of the Si capping layer were grown on a pair of samples with otherwise identical growth and layout parameters. For the first one, the substrate temperature was ramped from 350 to 450°C to achieve conformal covering of the Ge QDs\textsuperscript{11}. The second cap version commenced again at 350°C to preserve the QD shape, but was then ramped to 700°C to partly planarize the epilayer surface\textsuperscript{12}. The conformally capped samples were mainly used for visualizing the QD positions by SEM. These samples allowed us to validate an overall alignment precision of about 20 nm for the complete process. The partly planarized samples were used for optical characterization because their more homogeneous slab thickness leads to better defined cavities.

In the last processing step (Supplementary Fig. S2d) the initially defined alignment marks are exploited again to align the air-hole mask of the PCRs in such a way that the QDs become centered in the air holes. In this way, all QDs are removed during air hole etching except the one remaining in the cavity. The air holes are ICP-RIE etched with perpendicular sidewalls down to the BOX in an SF\textsubscript{6}/O\textsubscript{2} cryo-process. In a last step, the BOX beneath the PCR structures is selectively removed with 40% HF to achieve free-standing PCR slabs.

We want to emphasize that EBL is not essential to our approach and that the required position accuracy is routinely achieved in CMOS processes on an industrial scale. The technological approach of this work can thus be considered as CMOS compatible.

III Additional photoluminescence data and control experiments

In Supplementary Fig. S3a, PL data demonstrating the identification of observed single dot emission peaks with specific PCR modes are shown. For a cavity with parameters $a = 378$ nm, $r/a = 0.31$, the broad time-averaged emission spectra of our QDs allow coupling to five cavity modes M0 to M4. As is the case for the PL data presented in the main text, the dot emission intensity into specific modes is strongly dependent on the dot position. The dotted vertical lines in the plot indicate the wavelength position of the ideal cavity modes as calculated in
Fig. S 2. Fabrication process for photonic crystal resonators with a single Ge quantum dot at defined positions within the cavity. a, Alignment markers defined by electron beam lithography are etched deep into the buried oxide. b, Definition of the preferential nucleation sites for Ge QDs. c, Molecular beam epitaxial growth of the dots and the Si cap layer. d, Fabrication of the PCR by dry etching and subsequent buried oxide removal. In this step all QDs in registry with the air holes of the PCR are removed.

our simulations. Although there is a consistent, slight wavelength shift between calculated and experimentally observed modes, each emission peak can be clearly identified with a specific resonator mode. Such small shifts are indicative for minor deviations between the simulated and the implemented geometry parameters of the PCR. In particular, the (effective) hole radii may be affected by the fabrication process due to, e.g., surface roughness or slight tapering of the air holes. The pronounced influence of small changes of \( r/a \) is confirmed by the spectral shifts in Supplementary Fig. S3b, where \( r/a \) varies between 0.305 and 0.325 at \( a = 378 \) nm, with the QD being in the center position for all four spectra. Supplementary Fig. S3c illustrates the well-known dependence of the L3 cavity modes on the photonic crystal (PC) period, which was exploited in our studies to align the resonator modes with the broad QD spectra. As expected for PCRs with constant slab thickness \( h \), the mode wavelengths shift with \( a \) linearly\(^{13}\).

Note that according to Supplementary Figs. S3b,c the highest-\( Q \) mode M0 emits much weaker than modes M2 and M3, even if M0 is spectrally aligned with the maximum of QD emission. This is due to a pronounced difference in outcoupling efficiency between the modes inherent to the employed resonator design, which was originally developed for a high \( Q \) value of M0 and weak out-coupling perpendicular to the PC slab\(^1\). Supplementary Figs. S3b,c also contain a reference signal from an array of bare QDs which was measured on an area outside the PCR structures, where the original QD array is preserved after air-hole etching. The reference spectra are multiplied by a factor of 15, highlighting the enhancement of QD emission by the cavities.

As discussed in the main text, no prominent saturation effects of the PL intensity as a function of the QD position within the PCR are observed, indicating that the radiative decay is not dominating the decay dynamics of the optically excited QD in the PCR. As a consequence, time resolved PL experiments monitoring the excited state lifetime of the QD are not sensitive to the radiative recombination time, and thus, to the Purcell factor \( F_P \). Under these experimental conditions a very rough but reliable estimate of the lower limit for \( F_P \) of the observed PCR modes can be given based on the data in Fig. S3c. In the following, \( F_P \) of the M3 mode is considered, as this low-\( Q \) mode produces the most intense detector signal due to a high collection efficiency in the experimental setup. The spectral peak emission of the M3 mode is by a factor 60 higher than the ensemble reference maximum. The dot reference ensemble within the diameter of the excitation laser spot on average counts 12 dots. Next, we estimate the
**Fig. S 3.** Additional photoluminescence data. **a**, spectra of an L3 cavity with \( a = 378 \) nm, \( r/a = 0.31 \) with a single QD emitting into cavity modes M0 to M4. Spectra for three different locations of the Ge dot are displayed: Center position (black line), edge position (red line) and intermediate position (green line). The vertical lines and the arrows mark the simulated mode maxima (labeled M0*–M4*), which coincide to within 15 nm with the associated PL peaks. **b**, Shift of the PL spectra with filling factor \( r/a \) at fixed \( a = 378 \) nm, and, **c**, with period \( a \) at fixed \( a/r = 0.31 \). For **b** and **c** the QD is at the cavity center. As a reference, PL spectra (amplified by a factor of 15) of ensembles of ordered QDs outside the photonic structures are shown in gray. **d**, Comparison of PL spectra from a cavity \((a = 399 \) nm, \( r/a = 0.31)\) with the QD in its center (red line) and an identical cavity that contains just the wetting layer but no dot (black line, scaled up \( \times 5 \)). As a reference, a PL spectrum from an all-Si photonic crystal cavity is shown, where the latter was subjected to all processing steps with exception of the Ge epilayer growth (blue line, scaled up \( \times 200 \)).

The effect of the redistribution of light due to the differences in the far-field radiation pattern of a dielectric slab and a PCR: For QD emitters in a dielectric slab we assume a homogeneous ensemble emission over a solid angle of \( 4\pi \) sr. For an experimental numerical aperture \( NA \) of 0.7, as rough estimate of the collection efficiency 2% results for the ensemble emission. Further assuming a collection efficiency of 50% for the M3 mode (only the surface emission into one half-space is collected in our experiment) leads to an absolute lower limit of \( F_P > 30 \) for the M3 cavity mode. Note that an accurate estimation of the actual collection efficiency for the cavity modes and the ensemble in the Si slab without cavity is extremely difficult, and we therefore limit our attempts to this crude but reliable lower-limit evaluation. The emission of a single QD in central position within the L3 PCR is therefore enhanced by at least a factor of 30 as compared to a single dot in a Si slab, where the actual \( F_P \) value can be easily one order of magnitude higher and has an upper limit in the simulated value for an ideal cavity (without Ge material) of 2500.

Supplementary Figure S3d presents control experiments confirming beyond doubt the identification of the observed PL peaks as single dot emission into resonator modes. The plot compares PL spectra from a cavity with a centered QD and \( a = 399 \) nm, \( r/a = 0.31 \) (red line) with a reference cavity of identical geometry that contains just the Ge wetting layer, but no dot (black line). The latter is scaled up by a factor of five for better visibility. The Ge
WL behaves essentially as a quantum well for holes\textsuperscript{10} that is evenly distributed over the whole cavity. The latter evidently contributes to the PL signal of the cavity modes, but the signal is by a factor of 5 - 20 weaker than the signal caused by a single QD in the cavity. In addition, any features in the resonator spectrum that depend on the dot position have to be related to the QD, i.e. they cannot be induced by the delocalized wetting layer. Supplementary Fig. S3d further shows the PL spectrum measured for a PCR that contains no Ge at all (blue broken line, amplified 200 times) but was otherwise processed identically to the other samples. Evidently, the emission of residual defects is negligibly small in our PCRs, and the emission studied in our work clearly originates from single QDs. It has to be mentioned that control measurements on a dot-free PCR are absent from the work reported in\textsuperscript{14}, making the exclusion of defect-related emission difficult. Furthermore, no clear emission from a QD ensemble in a Si slab without a PCR was observed in\textsuperscript{14} even at 7 K, which is in strong contrast to both the room-temperature emission from the PC samples in this reference and the clear emission from our comparable QD ensembles.

IV Discussion on the origin of broad Ge single-dot spectra

We know from power-dependent PL measurements on single Ge QDs without resonator structure that Ge QDs on Si(001) have rather broad PL signals, even in the limit of very low excitation densities\textsuperscript{15}. These broad peaks were attributed to the interplay of several mechanisms that can be related to the type-II band offset and the indirect band gaps of Si and Ge\textsuperscript{15}. The type-II band offset leads to strong hole confinement in the Ge QD, whereas the small intrinsic conduction band offset allows only for weak electron confinement in local strain pockets of the Si matrix\textsuperscript{16,17}. Moreover, the strain fields lift the sixfold conduction band degeneracy of Si, leading to a dense and complex system of energy levels\textsuperscript{16,17}. As a consequence, electron fluctuations are to be expected at the high excitation densities employed here leading to spectral diffusion and significantly broadened emission spectra in time averaged experiments\textsuperscript{15,18}. While this temporal fluctuation of an actually narrow QD transition is not ideal for many applications, it proves advantageous in terms of spectral matching between cavity and emitter: The on a small time scale narrow transitions still allow system operation in a bad cavity regime, whereas a random ‘scanning’ of the transition energy on a larger time scale due to environmental fluctuations enables a statistical alignment between dot and cavity mode. The conditions for spectral matching are therefore tremendously relaxed, and only the spatial matching between QD position and mode profile has to be optimized as demonstrated in our work.

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