Single-photon emission from electrically driven InP quantum dots epitaxially grown on CMOS-compatible Si(001)

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Abstract
The heteroepitaxy of III–V semiconductors on silicon is a promising approach for making silicon a photonic platform. Mismatches in material properties, however, present a major challenge, leading to high defect densities in the epitaxial layers and adversely affecting radiative recombination processes. However, nanostructures, such as quantum dots, have been found to grow defect-free even in a suboptimal environment. Here we present the first realization of indium phosphide quantum dots on exactly oriented Si(001), grown by metal–organic vapour-phase epitaxy. We report electrically driven single-photon emission in the red spectral region, meeting the wavelength range of silicon avalanche photodiodes’ highest detection efficiency.

(Some figures may appear in colour only in the online journal)

1. Introduction
Making silicon a photonic platform represents a long-winded challenge for the semiconductor community. Direct heteroepitaxy of III–V semiconductors on silicon (Si) would be highly desirable, enabling on-chip optical interconnects [1–3] and quantum optical applications [4, 5]. Monolithic integration of both material systems is hampered by different material properties, leading to high defect densities in the epitaxial layers. To date, numerous realizations of light emitting diodes (LEDs) or even lasers on Si have been reported [6–9], mostly, however, using thick buffer layers (up to 10 µm) or misoriented substrates. The latter are used in order to prevent the formation of antiphase domains, a problem related to the growth of polar on nonpolar material. Using offcut substrates, however, precludes a direct integration in mainstream Si technology, as complementary metal oxide semiconductor (CMOS) processes require exactly oriented Si(001) substrates. Thick buffer layers are undesirable as they cause large height differences and thus negatively impact high resolution lithography. Alternatively, III–V emitters have been realized on Si by hybrid technologies [10, 11]; this, however, is a very complex method, making CMOS integration time-consuming and expensive. The methods mentioned above are indispensable for providing the optimal growth conditions required for spatially extensive layers, such as quantum wells. Defects within these layers deteriorate the optical and electrical characteristics of the device and may lead to reduced reliability. In recent years, however, nanostructures have been shown to be suitable for successfully realizing light emitters on silicon [12–17]. Their facet edges and sidewalls can minimize or eliminate the formation of dislocations [14], and, due to the reduced contact area, nanostructures are little affected by dislocation networks [17]. Here we demonstrate the potential of indium phosphide (InP) quantum dots (QDs) as a promising base for lasers and non-classical light emitters on CMOS-compatible Si(001). Using QDs as the active light emitting medium provides the...
possibility to build up semiconductor lasers with superior performance, such as low thresholds and broad gain spectra. Over and above this, self-assembled QDs are excellent candidates for single-photon sources, which are essential for quantum information science and technology [18–20]. The emission wavelength of the InP QDs in this work lies in the red spectral region around 650 nm, which makes them interesting for chip-to-chip optical interconnects, as for this wavelength Si avalanche photodiodes have their highest detection efficiency, and also polymer optical fibres (POFs) have an attenuation minimum [21].

2. Sample fabrication

The growth of III–V structures was performed by metal–organic vapour-phase epitaxy (MOVPE) using exactly oriented Si(001) substrates and standard precursors (trimethylgallium, trimethylindium, trimethylaluminium, arsine and phosphine). Structures for photoluminescence (PL) measurements were deposited on top of a III–V buffer, as illustrated in figure 1(a). This buffer had the purpose of reducing the high defect density accompanying III–V/Si heteroepitaxy and was built up of a strained-layer superlattice (SLS) embedded in two GaAs layers of 1 µm thickness. The SLS consisted of 10 periods of 10 nm In$_{0.45}$Ga$_{0.55}$As and 20 nm GaAs. The strain introduced by the SLS interacts with dislocations and suppresses propagation into overlying layers. Temperature cycle steps during buffer growth, performed in a range between 200 °C and 790 °C, further support dislocation reduction. On top of this buffer, self-assembled InP QDs were grown for optical investigation. The QDs were embedded in barriers made of (Al$_{0.5}$Ga$_{0.5}$)$_{0.51}$/In$_{0.49}$P and (Al$_{0.2}$Ga$_{0.8}$)$_{0.51}$/In$_{0.49}$P which provide efficient carrier confinement up to elevated temperatures. For details of the growth process, please refer to previous work [22].

For electrically pumped samples, the pin structure (see figure 4(a)) was built up of 80 nm of GaAs$_{0.51}$/In$_{0.49}$P:Si and 200 nm of AlInP:Si, followed by the deposition of 50 nm of intrinsic Al$_{0.55}$Ga$_{0.45}$InP. QDs were grown by depositing two monolayers of InP onto 10 nm of intrinsic Al$_{0.20}$Ga$_{0.80}$InP. The QDs were capped by a further 10 nm of intrinsic Al$_{0.20}$Ga$_{0.80}$InP and 50 nm of intrinsic Al$_{0.55}$Ga$_{0.45}$InP. 200 nm of AlInP:Zn, 50 nm of GaInP:Zn and 200 nm of GaAs:Zn completed the pin structure. For device processing, Cr/Zn/Au/Pt/Au-strips were evaporated to form the p-contact, and an Al-layer deposited on the back of the sample provided the n-contact.

3. Characterization methods

Low-temperature ($T = 5$ K) cathodoluminescence (CL) measurements were performed in a home-built setup based on a modified JEOL 6400 scanning electron microscope. The kinetic energy of the incident electron beam was set to 5 keV ($I_{\text{beam}} = 730$ pA). This yielded a maximum penetration depth of $d = 350$ nm for Al$_{0.5}$Ga$_{0.5}$InP [23] and the highest energy dissipation up to a depth of about 100 nm below the surface, generating the excited carriers directly inside the AlGaInP barriers of the InP dots.

PL measurements were performed in a He-flow cold finger cryostat. A heater inside the cryostat enabled temperature control from 4 to 300 K. The ensemble luminescence was investigated under optical excitation by a frequency-doubled Nd:YAG laser in continuous wave (CW) mode at 532 nm with a spot size of 50 µm in diameter and an intensity of 650 W cm$^{-2}$ on the sample.

For $\mu$-PL investigations, a pulsed white light laser, based on a nonlinear photonic crystal fibre, was used for optical excitation, with a repetition rate of 50 MHz and a pulse width of 150 ps. The excitation wavelength (580 nm (4 K) or 570 nm (80 K)) was selected via either a 150 mm focal length monochromator or an acousto-optical tunable filter. Illumination and detection of the sample was carried out confocally through a 50× microscope objective, focusing the laser spot down to a diameter of approximately 1 µm. Two stepper motors allowed horizontal and vertical movement of the cryostat, with an effective spatial resolution of 50 nm in each direction. As will be seen later, the density of optically active QDs was sufficiently low to investigate single dot emission without the need for shadow masks.

4. Results and discussion

For atomic force microscopy (AFM) investigations, QD structures were grown without capping layers. Figure 1(b)
Figure 2. Investigations of InP QD luminescence. (a) Spectra of optically excited InP QDs, showing the emission from the buffer (GaAs), the wetting layer (WL) and two types of QD (type-A and type-B). (b) Ensemble PL spectrum of InP QDs at 4 K (green boxes: measured data, black lines: Gaussian fit curves representing contributions from the wetting layer, type-A and type-B QDs). The locations of emission centres on the sample surface were mapped by cathodoluminescence microscopy. The results for two selected wavelength regions are illustrated in the SEM image depicted in (c) (blue: 665–675 nm, red: 725–750 nm).

shows a typical AFM scan of a QD layer on top of the extended buffer structure. We mainly observe lens-shaped structures of two different sizes. While the smaller ones are found all over the investigated area, the larger ones are predominantly arranged in chains along the edges of trenches or valleys. This ordering phenomenon can be explained by effective local strain fields [24] at these locations leading to a preferred nucleation and assembly of InP. We will see later that the structures observed can be identified as two different types of QD. By means of AFM analysis we can estimate the density of small QDs, in the following named ‘type-A’ QDs, to be $4.2 \pm 1 \times 10^{10}$ cm$^{-2}$, and for the large QDs (‘type-B’) we find $(1.9 \pm 1) \times 10^9$ cm$^{-2}$. The density of QDs is of the same order of magnitude as for QDs on GaAs substrates, deposited within the same barrier structure and with the same growth parameters. On GaAs substrates, we also find a bimodal distribution, however shifted to higher emission energies, which indicates that the QDs are smaller.

The temperature-dependent ensemble PL spectra are shown in figure 2(a). At 4.6 K, QD emission is observable as broad emission peaks around 1.70 eV (type-B) and 1.88 eV (type-A). With increasing temperature the emission intensity decreases, since charge carriers are thermally activated out of the QDs and get lost for radiative recombination. Due to the stronger quantization, this loss process is more severe for the smaller sized type-A QDs, having a smaller energetic distance between ground state and barrier. Type-B QDs have a smaller energy-level spacing and therefore more energy levels in the QDs, which may lead to a higher population number of carriers. In addition, carrier capture and relaxation processes are more efficient in type-B QDs, enabling luminescence up to 300 K. Comparing the integrated intensities of both QD types at 4 K with QDs deposited on a GaAs substrate (same growth parameters, same barrier structure), we find that the QDs on Si yield about 35% of the intensity of QDs on GaAs.

In order to confirm the correlation between the structures observed by AFM and the PL results, the sample was investigated by spatially and spectrally resolved CL microscopy [25]. This method allowed the creation of luminescence maps within a particular wavelength region. Based on the PL results, we chose narrow spectral regions in which the emission of one QD type dominates, with negligible influence of the wetting layer or the other QD type, respectively (figure 2(b)). The local distribution of the luminescence of both QD types is illustrated in a scanning electron microscope (SEM) image of the investigated area. As can be seen, there is almost no overlap of the luminescence patterns of the two spectral regions, indicating that the two QD types are located in different areas. Moreover, we find that type-B QDs generate a strikingly horizontally or vertically oriented pattern, which coincides with the distribution of trenches, along which AFM analysis had shown an agglomeration of large QDs. Over and above this, the CL maps give an impression of the density of optically active QDs, which was estimated by $\mu$-PL measurements to be of the order of $10^9$ cm$^{-2}$. The relatively high proportion of optically inactive QDs is assumed to be due to the still existing defects in the active layer.

Since light emitted by single QDs shows significant non-classical characteristics, second-order autocorrelation measurements $g^{(2)}(\tau)$ were performed in a Hanbury-Brown and Twiss type setup [26] to verify single dot emission [18]. The count rates on the avalanche photodiodes (APDs) during autocorrelation measurements were 26 000 for the sample measured at 4 K and 20 000 for the one at 80 K (corresponding to 208 000 and 222 000 photons collected in the first microscope objective (NA = 0.45)). The realizable total time
resolution of the setup was around 500 ps, limited by the time resolution of the APDs. The number of coincidence events versus the delay time \( \tau \) is shown in figure 3 for optically pumped QDs under pulsed excitation. Both measurements show a pronounced suppression of coincidences at \( \tau = 0 \). The experimental data were fitted using

\[
g^{(2)}(\tau) = P \left( 1 - ae^{-\frac{\tau}{\tau_d}} \right),
\]

(1)

where \( a \) is the value of the antibunching dip, \( \tau_d \) is the antibunching dip time constant and \( P \) is the Poisson level. The relevant \( g^{(2)}(0) \) can be calculated using \( g^{(2)}(0) = 1 - a \). The QD in figure 3(a) reveals a \( g^{(2)}(0) \) value of 0.08 at 4 K (\( \tau_d = 0.68 \) ns), which indicates a decrease in multi-photon emission events by a factor of approximately 12 when compared to a Poissonian source of the same average intensity. The deviation from \( g^{(2)}(0) = 0 \), as would be expected for an ideal source, is caused by the limited temporal resolution of the experimental setup, the non-zero probability of multi-photon emission, and uncorrelated background emission. The latter becomes more severe with increasing temperature, as apparent in the PL spectrum at 80 K (figure 3(b)). Nevertheless, autocorrelation measurements showed single-photon emission with \( g^{(2)}(0) = 0.37 \) (\( \tau_d = 0.94 \) ns). Considering the contributions of the background emission [27], we obtained a corrected value...
for $g^{(2)}(0)$ of 0.15 at 80 K. Although the measurements were carried out under pulsed excitation, no correlation peaks can be identified at 4 K (figure 3(a)). This behaviour can be explained by charge traps in the barrier material, in which electrons and holes are stored after optical excitation. After the decay of the exciton in the QD, the charge carriers diffuse into the QD and thus populate it again before the next excitation pulse [28]. This refilling effect becomes less pronounced at higher temperatures, as the traps are then less populated. Consequently, at 80 K, correlation peaks are observable at multiples of the laser repetition rate (figure 3(b)).

The results shown so far clearly demonstrate the applicability of InP QDs to build up light emitters even in a difficult environment. However, any application demands electrical pumping. Therefore, our next step was to transfer the active zone into a pin-diode on n-doped Si(001). A sketch of the sample structure is depicted in figure 4(a). The III–V buffer was kept simple in this case, solely consisting of a GaAs:Si layer of 1 $\mu$m thickness. Figure 4(b) shows the temperature-dependent ensemble luminescence under electrical excitation. Again we see two types of QD contributing to the emission, observable as two broad peaks at 1.73 and 1.91 eV at 4 K. The emission of type-A QDs can be observed up to 80 K, and type-B QDs again show luminescence even up to room temperature.

The electrical properties of the diodes are quite comparable with those of diodes realized on GaAs substrates, as apparent in figure 4(c). Here we see the diode characteristics of InP QD-based LEDs, on Si and GaAs, respectively. The characteristics of both LEDs are in good accordance, both at 4 K and at room temperature.

Furthermore, samples were subjected to $\mu$-EL measurements, using a current source with a resolution of 0.1 mA for excitation. $\mu$-EL measurements were then performed similarly to the $\mu$-PL measurements described above. Figure 5(a) depicts the emission characteristics of a single QD while varying the bias voltage. Starting at 2 V, we see a narrow line at 1.889 eV, which can be attributed to an excitonic transition, i.e. the recombination of an electron–hole pair. Increasing the voltage to 2.02 V, and thereby also increasing the injection current, an additional line emerges with an energy difference of 5 meV. This value fits well to the exciton–biexciton binding energy of approximately 4–6 meV in this material system [29] and thereby indicates the zero-dimensionality of the light emitter. Typically, with increasing injection current, the gain in emission intensity is greater for the biexciton compared to the exciton (see the inset in figure 5(a)). Figure 5(b) shows the temperature-dependent emission from a single QD. Luminescence can be observed up to 60 K, keeping up with electrically pumped InP QDs grown on GaAs substrates [27].

Autocorrelation measurements on an electrically pumped QD under DC current excitation were performed at count rates of 18 000 on the APDs (225 000 photons collected in the first microscope objective). As displayed in figure 5(c), the measurements show also non-classical photon statistics, with a $g^{(2)}(0)$ value of 0.52 ($\tau_{el} = 0.48$ ns). Here, background correction yields only a slight reduction down to $g^{(2)}(0) = 0.48$. Since we did not deconvolute the instrument response function, all $g^{(2)}(0)$ values given here should be understood as upper limits. Compared to the results of optically pumped QDs we find significantly higher $g^{(2)}(0)$ values for electrically pumped structures which we mainly attribute to a higher defect density due to the simplified buffer structure. An improved buffer should therefore lead to enhanced single-photon emission characteristics.

5. Conclusions

We have investigated the growth and emission characteristics of InP QDs on Si(001), showing photoluminescence
up to room temperature. Second-order autocorrelation measurements prove single-photon emission with a $g^{(2)}(0)$ value of 0.08 at 4 K. Moreover, InP QDs in a pin-diode structure were successfully realized.

We attach great importance to CMOS process compatibility and therefore used only exactly oriented Si(001) substrates. The buffer structures allowed high resolution lithography; however, there is surely room for improvement regarding thickness and defect suppression. The growth temperature did not exceed 700 °C. Annealing steps performed at higher temperatures may be modified to comply with CMOS restrictions.

With this first presentation of electrically driven single-photon emission on CMOS-compatible Si substrates, InP QDs have proven to be highly attractive light sources for future Si based photonic integrated circuits and quantum information technology.

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