The epitaxial growth and unique morphology of InAs quantum dots embedded in a Ge matrix

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Abstract
In this work, we investigate the epitaxial growth of InAs quantum dots (QDs) on Ge substrates. By varying the growth parameters of growth temperature, deposition thickness and the growth rate of InAs, high density (1.2×10¹¹ cm⁻²) self-assembled InAs QDs were successfully epitaxially grown on Ge substrates by solid-source molecular beam epitaxy and capped by Ge layers. Pyramid- and polyhedral-shaped InAs QDs embedded in Ge matrices were revealed, which are distinct from the lens- or truncated pyramid-shaped dots in InAs/GaAs or InAs/Si systems. Moreover, with a 200 nm Ge capping layer, one-third of the embedded QDs are found with elliptical and hexagonal nanovoids with sizes of 7–9 nm, which, to the best of our knowledge, is observed for the first time for InAs QDs embedded in a Ge matrix. These results provide a new possibility of integrating InAs QD devices on group-IV platforms for Si photonics.

Keywords: quantum dots, germanium, molecular beam epitaxy, nanovoids

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

1. Introduction
The epitaxial growth of InAs quantum dots (QDs) on GaAs substrates and GaAs/(Ge)/Si substrates has been thoroughly investigated for the pursuit of optoelectronic applications such as lasers [1] and photodetectors on Si [2]. The almost identical lattice constants and thermal expansion coefficients (TECs) of Ge and GaAs make Ge a promising substitute to GaAs substrates. Ge substrates are not only advantageous in mechanical strength and crystal quality, but also are available in larger scale wafers of 8 inches in diameter. More importantly, they are capable of being integrated with a Si substrate with a Ge thin buffer [3].

Until now, the heteroepitaxial growth of InAs QDs directly on Si or Ge substrates has been rarely reported by several groups, where different dot morphology and density were investigated. For example, large dome-shaped InAs QDs with sizes of 30–70 nm were grown at 400 °C on a hydrogen-terminated Si (100) substrate with a density of 1.2×10¹¹ cm⁻² [4]. Later, high uniformity InAs QDs grown on Si substrates were demonstrated with a dot density of ~10¹¹ cm⁻² and 1.3 μm wavelength photoluminescence emission [5, 6]. Recently, nearly strain-relaxed InAs QDs...
embedded in a defect-free Si matrix have been demonstrated by a combination of several steps of overgrowth and post-growth annealing processes [7]. As for the epitaxial growth of InAs QDs directly on Ge, InAs and InGaAs islands were grown on a 6° offcut Ge (100) substrate by metalorganic vapour phase epitaxy. The highest island density achieved was $2.5 \times 10^{10}$ cm$^{-2}$ and photoluminescence from the InAs islands was only observed when embedded in GaAs capping layers [8, 9].

Compared with InAs QDs directly embedded in Si, where the critical radius of nanoclusters is just a couple of nanometres due to the large heteroepitaxial strain of 11.6% [10], the size tunability of InAs QDs in Ge is comparatively higher [5, 9]. In addition, with the highly lattice-mismatched InAs-Si heteroepitaxial growth, it is easy to produce mesoscopic dislocated clusters instead of nanoscale QDs [11]. The InAs/Ge system can also be easily buried into a Si matrix to provide better carrier confinement. Inspired by the abovementioned advantages, in this paper we have investigated the growth mechanism of InAs QDs directly grown on a Ge substrate by varying the growth temperature, deposition thickness and growth rate of the InAs, which results in high density InAs/Ge QDs of $1.2 \times 10^{11}$ cm$^{-2}$. Furthermore, the overgrowth of the Ge capping layer was examined with 40 nm and 200 nm, respectively. The QD morphology was changed during the different capping processes. By understanding the mechanism of InAs QDs grown on a Ge substrate, the Ge/InAs QDs/Ge stack can potentially be a candidate for new generation Si-based optoelectronic devices in the future, of which the growth is fully complementary metal-oxide-semiconductor compatible, low-cost and feasible.

### 2. Material epitaxial growth and characterisation

The material epitaxial growth was carried out in a twin-chamber Veeco Gen-930 solid-source molecular beam epitaxy (MBE) system, where wafers can be transferred under ultra-high vacuum conditions between III–V and group-IV growth chambers. In the III–V growth chamber for QD growth, the III–V growth chamber for QD growth. The growth was initiated based on our optimised growth techniques for III–V on Ge by applying a group-III pre-layer [14]. For each

### Table 1. Parameters of layer-by-layer deposited Ge/lnAs/Ge substrates.

| Sample | Substrate temperature, $^\circ$C | InAs coverage, ML | Growth rate, ML (s$^{-1}$) | Cap layer | Dot density, $\times 10^{10}$ cm$^{-2}$ |
|--------|-------------------------------|-----------------|-----------------|-------------|---------------------------------------|
| A      | 450                           | 2.1             | 0.15            | No          | 0.40                                  |
| B      | 430                           | 2.1             | 0.15            | No          | 0.43                                  |
| C      | 410                           | 2.1             | 0.15            | No          | 1.38                                  |
| D      | 390                           | 2.1             | 0.15            | No          | 1.65                                  |
| E      | 360                           | 2.1             | 0.15            | No          | 2.21                                  |
| F      | 360                           | 1.8             | 0.15            | No          | 6.18                                  |
| G      | 360                           | 1.65            | 0.15            | No          | 0.37                                  |
| H      | 360                           | 1.49            | 0.135           | No          | 2.88                                  |
| I      | 360                           | 1.32            | 0.12            | No          | 11.6                                  |
| J      | 360                           | 1.32            | 0.12            | 40 nm       | —                                     |
| K      | 360                           | 1.32            | 0.12            | 200 nm      | —                                     |

### 3. Results and discussion

#### 3.1. Growth optimisation

To find the optimal growth conditions for InAs QDs on Ge, three series of samples were grown, which are summarised in table 1. Prior to the growth details, as mentioned above, the Ge wafers were all deoxidised in a group-IV MBE chamber instead of direct deoxidisation in the III–V chamber for the growth, aiming to avoid As etching of the Ge surface [12]. To verify the effect of the As-rich environment on Ge, one p+ Ge (100) wafer was loaded into the III–V growth chamber and heated to $\sim 400^\circ$C with an As background. As$_2$ molecules with a beam-equivalent pressure (BEP) of $5 \times 10^{-6}$ Torr were supplied for 10 min. After that, the sample was taken out for surface probing. Figures 1(a) and (b) show $1 \times 1 \mu$m$^2$ and $5 \times 5 \mu$m$^2$ AFM results, respectively. The AFM image of the Ge surface just after deoxidation without any As background is also presented as a reference, as shown in figure 1(c), which shows no ordered surface patterns. In contrast, etched step edges can be clearly seen in figure 1(a). Such etching happened in directions both along and perpendicular to the step edge. The height profile across the surface, as indicated by the white line in figure 1(a), is shown in figure 1(d), with a step height of approximately 2.36 Å, which is near to the theoretical double atomic step (2.83 Å). In larger scale AFM images, as shown in figure 1(b), sawtooth-like surface constructions are observed, which are similar to the dendritic single steps in a hydrogen-annealed Si surface explained to be the result of dimer-vacancy rows crossing the terraces in either a [110] or [1-10] direction [13].

After deoxidation, wafers were directly transferred to the III–V growth chamber for QD growth. The growth was initiated based on our optimised growth techniques for III–V on Ge by applying a group-III pre-layer [14]. For each
AFM images of central parts of the background As-etched Ge surface: (a) $1 \times 1 \mu m^2$ and (b) $5 \times 5 \mu m^2$; and (c) $1 \times 1 \mu m^2$ AFM image of the Ge substrate surface after deoxidation without any As background. (d) Surface height profile of the white dashed line in (a).

sample, 1 ML In was deposited on the Ge first without As flux at the same temperature and growth rate as QD growth. Then As$_2$ overpressure was provided for 10 s to fully passivate the In monolayer. Although the In BEP varied with different In growth rates, a fixed V/III ratio of 55 was used for QD growth. For example, for QD growth with an In growth rate of 0.15 ML s$^{-1}$, the BEP of In was $9 \times 10^{-8}$ Torr and that of As$_2$ was $5 \times 10^{-6}$ Torr. Three series of samples were grown to investigate the effects of substrate temperature, InAs coverage and growth rate on dot morphology and density. In the first series (samples A–E), an In pre-layer, an InAs coverage of 2.1 ML and a growth rate of 0.15 ML s$^{-1}$ were used as a starting point to find the optimal growth temperature. Figures 2(a)–(e) show $1 \times 1 \mu m^2$ AFM images of samples A–E, respectively.

The dot diameter and height histograms of samples A–E are presented in figure 3. A clear trend of increasing dot density with decreasing substrate temperature was found (from samples B–E). The dot density is increased from $(4.28 \pm 0.38) \times 10^9$ cm$^{-2}$ (sample B) to $(2.21 \pm 0.13) \times 10^{10}$ cm$^{-2}$ (sample E), as shown in figure 2(f). For samples A and B, the dot densities are almost the same, which are $(4.0 \pm 0.44) \times 10^9$ cm$^{-2}$ and $(4.3 \pm 0.38) \times 10^9$ cm$^{-2}$, respectively. Normally, the dot density should increase with decreasing growth temperature because of suppressed InAs desorption and an increased sticking coefficient. This anomalous behaviour could be attributed to the coalescence of the adjacent dots [9] and/or a coarsening dynamics of Ostwald ripening, due to material exchanges between the growing QDs, which is favoured at high mobility. Further decreases in growth temperature showed no significant positive effect on dot density, indicating that around 360°C is the optimal temperature for InAs QD/Ge growth. A typical size of QD from samples A–E (except sample B) has a diameter of 35–60 nm and a height of 4–12 nm.

Later, to examine the effect of InAs coverage on QD density, samples F and G were grown with decreased InAs coverages of 1.8 ML and 1.65 ML, respectively, by shortening the deposition duration while maintaining other growth parameters. An AFM image of sample F is shown in figure 4(a). Compared with sample E, a significant improvement of dot density is identified. However, although sample F has reached a high dot density, a large amount of defect dots are also observed. This can also be seen from the diameter histogram of sample F in figure 5; a long tail up to 60 nm is presented. Those defect dots are several times larger in dimension than others and may contain lots of crystal defects, deteriorating the optical properties of the QDs. A possible formation mechanism of these defect dots is the coalescence of adjacent dots.

By further decreasing the InAs coverage to 1.65 ML, the dot density in the central area of sample G, as shown in figure 4(b), drops dramatically to $3.7 \times 10^9$ cm$^{-2}$. Indeed,
Figure 3. Dot diameter and height histograms of samples A–E.

Figure 4. (a)–(d) AFM images (1 \times 1 \mu m^2) of the central parts of samples F, G, H and I, respectively.

Figure 5. Dot diameter and height histograms of samples F–I. Gaussian distribution can be found for the diameter histogram of sample I and the height histograms of samples F and I.

Figure 6. (a) Schematic illustration of the grown structure. (b)–(e) AFM images (1 \times 1 \mu m^2 and 5 \times 5 \mu m^2) of the samples are shown in figures 6(b) and (c) (sample J), and figures 6(d) and (e) (sample K).

3.2. Ge overgrowth

Based on the optimal growth conditions for QDs (sample I), 40 nm (sample J) and 200 nm (sample K) Ge capping layers were grown in the group-IV MBE chamber using a two-step growth method. A schematic illustration of the grown structure is shown in figure 6(a). AFM images (1 \times 1 \mu m^2 and 5 \times 5 \mu m^2) of the samples are shown in figures 6(b) and (c) (sample J), and figures 6(d) and (e) (sample K). A smooth epi-surface with atomic steps could be clearly seen for both samples, indicating the QDs have been fully covered by a Ge layer, which is different from Si/InAs/Si where numerous nanoholes as large as QDs were depicted after Si overgrowth [7]. However, surface threading dislocations (TDs) with a density as the dot density varies substantially, which is not as expected, larger scale AFM scans were performed to check the general morphology. It was found that the surface shows large undulations of height around 30 nm, decorated with clustered QDs, therefore giving rise to the large discrepancy of surface dot density. The possible reason for this could be accidental surface contaminations acting as nucleation centres for the large undulations [15], but this needs to be investigated further in future work. Nevertheless, from comparing samples E and F, the beneficial impact of reducing the InAs coverage can be inferred, as the dot density is significantly increased. Consequently, in the third group (samples H and I), the growth rates were varied from 0.135 ML s\(^{-1}\) to 0.12 ML s\(^{-1}\), with a deposition time of 11 s and a substrate temperature of 360 °C. A high QD density of \(1.2 \times 10^{11} \text{cm}^{-2}\) with a dramatically reduced number of defect dots was achieved in sample I, as demonstrated from figure 4(d). The diameter and height histograms of samples F–I are shown in figure 5. Wide diameter distribution has also been found for samples F, G and H. The height histogram improves dramatically for sample F, where Gaussian distribution centred at 4.3 nm can be seen. For sample I, both diameter and height demonstrate Gaussian distribution, centred at 10.2 nm and 6.5 nm, respectively. A typical QD size in sample I is 5–18 nm in diameter and 5–8 nm in height. These results lead us to draw the conclusion that the growth dynamics of the QDs is a result of the coordinative changes of the growth temperature, material coverage and dot growth rate rather than the monotonic change of one variable.
of $3 \times 5 \times 10^8$ cm$^{-2}$ were estimated for both 40 nm and 200 nm Ge samples by counting the dark holes in the AFM images. These TDs mainly originate from misfit dislocations at the InAs/Ge interface and later travel to the surface. It is also noteworthy that the defect morphology for the thin and thick capping layer samples is different. In addition to surface TDs, as observed in sample J with a 40 nm Ge capping layer (see figures 6(b) and (c)), short-line indentation defects with similar lengths were also found in sample K with 200 nm Ge capping, as shown in figures 6(d) and (e). The line indentation defects may be related to stacking faults (SFs) extending to the surface [16]. To find out the potential explanation for the different surface morphology of the overgrowth samples and to have a closer investigation of the novel material system, STEM measurements were applied.

4. STEM analysis

To investigate the morphology of the embedded InAs QD structures, large-scale STEM was first conducted. Figures 7(a) and (b) show HAADF and BF-STEM images of InAs QDs with 40 nm-thick Ge capping layers, respectively. The well-defined pyramid- and polyhedral-shaped InAs QDs with a brighter colour than the Ge were observed in figure 7(a). A continuous wetting layer and defect-free Ge matrix were also confirmed. In contrast, the InAs QD with a 200 nm-thick Ge capping layer presents clear differences in terms of the buried dot morphology and the surrounding Ge matrix, as shown in figures 7(c) and (d). In figure 7(c), approximately one-third of QDs are found with a considerably reduced HAADF-intensity nanostructure on the top or top corner, indicating the existence of nanovoids. Indeed, [111] defects of similar and even larger (with a reversed-truncated cone shape) dimensions are ubiquitous in the 200 nm capped sample, ascribed to the latter capping process. The size of the abovementioned reversed-truncated cone shape defect is consistent with the surface indentation defect shown in the AFM images (figures 6(d) and (e)). Note that numerous tiny spots observed in the Ge matrix for both samples are caused by sample cleavage and thus should be exempt from the structural analysis.

Figures 8(a)–(c) show high magnification HAADF-STEM images of selected typical QD structures in the 40 nm Ge capping layer of sample J. Several features of these QDs can be summarised from the images, as below. (a) The QDs have a good profile, showing no Ge-InAs inter-diffusion. (b) Although most of the QDs contain crystal defects, visually, none of them has propagated to the above Ge matrix. (c) The aspect ratio of the QDs is apparently smaller than that of InAs QDs grown on GaAs [17–20]. The truncated pyramid shape has been reported for InAs QDs embedded in a GaAs surrounding [20]. In our case, dots either remain pyramid-shaped (figure 8(a) dot on the right and figures 8(b)–(c)) or have a spherical shape (figure 8(a) dot on the left). A smaller dot 6.2 nm in diameter and 5.8 nm in height and a larger dot 24 nm in diameter and 11.4 nm in height are found, as shown in figure 8(a). The observed sizes of the QDs are comparable to the uncapped QDs (sample I) depicted by the AFM results in section 3.1, which means that the strain of the embedded QDs is well relaxed. The observed crystal defects of SFs, twin boundaries (TBs) or misfit dislocation loops are marked by white dashed lines, orange solid lines and small white triangles, respectively, in figure 8(c).

It is also noteworthy that the morphology discrepancy between the surface dots and buried dots may point to
behaviour is also dissimilar to that in a GaAs matrix where circles, and in the basal centre (figure not shown here). This is seen through the darker intensity inside the wetting layers in some Ge-InAs interdiffusion is recognised, which can be explained by the formation of dislocation half-loops. They are reported to nucleate from 60° dislocations at a critical thickness, then elongate along the {1,1,1} plane forming half-loops and keep expanding the diameter with the help of the strain field within the material [21]. Normally, these dislocation half-loops will ultimately reach the material interface and either become linear misfit segments or travel upwards as threading dislocations. In our case, the dislocation loops seem mostly to form misfit segments at the InAs/Ge interface. However, there is a possibility that the threading dislocations still originate from the buried dots because of the existence of the threading segments.

The structures of the embedded QDs under a 200 nm Ge cap are even more unique. First, the most unusual observation is the formation of the nanovoids, as presented in figures 9(a)–(c). Only one-third of nanovoids are of an ellipse cross section (figure 9(a)), and the rest have a hexagonal shape with (100) facets forming the upper bound of the void and the interface with the QD, as can be seen in figures 9(b) and (c). Both the pyramidal dots and the hexagonal void facets have a ~55° angle with a (100) crystal plane, which correspond to {1,1,1} facets. The hexagonal nanovoids have a sharp interface with the Ge matrix, in which the facets facing each other are parallel and the two facets on each side form an obtuse angle of approximately ~108°, rendering one to link the formation of the nanovoids to the possible defect- or strain-related movement of the crystalline plane during the capping process. Besides, another important observation is that the chance to find a ‘perfect’ dot, i.e. without any defect, is increased for the 200 nm Ge cap sample compared with the 40 nm Ge cap sample. Two such examples are shown in figures 9(d) and (e), where large (21.6 nm) and small (12.5 nm) dots are chosen. For QDs with a nanovoid on the top, the dot itself (which means the dot material, except the void defect) tends to be defect-free, as shown in figures 9(a) and (b). Nonetheless, SFs and misfit dislocations can occasionally be seen in dots with or without a nanovoid, as presented in figures 9(c) and the hexagonal void facets (f), respectively.

Although the formation mechanism of the nanovoid is unclear as yet, two possible explanations are proposed as follows. First, the voids are caused by the In segregation during the high temperature Ge overgrowth. As mentioned in section 3.2, the Ge overgrowth was performed by the two-temperature step method, of which the high temperature used was 500 °C and the growth duration was 50 min for sample K. Kept at this high temperature for a long time, the embedded QDs may consequently experience structural changes. Likewise, materials missing from QDs were also reported by Lenz et al, where after the deposition of InGaAs QDs onto a GaAs substrate, about 5 nm GaAs was overgrown at 500 °C followed by a 600 s long interruption at an elevated temperature of 600 °C [22]. From their observations, small dots were not affected, while larger and thus more strained dots first were affected.

The important role of the TEC of the dot material and the surrounding matrix. In the InAs/Si system, surface dots are mostly lens-shaped, whereas the buried dots are more spherical [7]. This is because dots are almost unstrained at the growth temperature and only strained again during aftergrowth cooling due to the difference in the linear TEC [7]. In contrast, InAs and Ge have a similar TEC, inferring less discrepancy between the morphology of the surface and buried dots, which is in good agreement with that observed for sample I (uncapped dots) and J (capped dots). In addition, the wetting layer is continuous without defects even though some Ge-InAs interdiffusion is recognised, which can be seen through the darker intensity inside the wetting layers in figure 8. This behaviour is distinct from the InAs QD on Si where a discontinuous wetting layer was reported [7]. Besides, In enrichment is detected in some dots in the basal corner of the dots, as shown in figures 8(a) and (c) marked by red circles, and in the basal centre (figure not shown here). This behaviour is also dissimilar to that in a GaAs matrix where reversed truncated pyramid-shaped In enrichment is found. Last, defects are observed both in smaller and in larger dots, while a dot without any defect is only found for smaller dots (~15.5 nm in diameter and 10.4 nm in height, e.g. shown in figure 8(b)). The significantly reduced defect propagation can be explained by the formation of dislocation half-loops. They are reported to nucleate from 60° dislocations at a critical thickness, then elongate along the {1,1,1} plane forming half-loops and keep expanding the diameter with the help of the strain field within the material [21]. Normally, these dislocation half-loops will ultimately reach the material interface and either become linear misfit segments or travel upwards as threading dislocations. In our case, the dislocation loops seem mostly to form misfit segments at the InAs/Ge interface. However, there is a possibility that the threading dislocations still originate from the buried dots because of the existence of the threading segments.

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truncated and then suffered from severe In eruption, which was confirmed from the occurrence of sub-monolayer In, and afterwards were unable to be fully filled due to the formation of either a concave top facet or void within the QD [22]. Moreover, a hexagonal-shaped void has also been documented in metalorganic chemical vapour deposition of N-polar AlN film on a sapphire substrate, where the oxygen out-diffusion was accused of the formation of the void and further related to the generation of inversion domains during the overgrowth [23]. However, in our experiment, no In sub-monolayer or small In clusters are detected in the surrounding matrix, and the missing materials are on the top or top corners of the dots instead of in the middle. Besides, if we treat the 40 nm cap sample as an early stage of the 200 nm cap sample, the In enrichment behaviour from the beginning stage is divergent from that in the GaAs case. In some HAADF images of dots with a void, In enrichment is also observed just below the void, challenging the reliability of this material segregation explanation. Nonetheless, the observed SFs in the Ge matrix and short-line indentation defects from the AFM images (figures 4(d) and 4(e)) should be related to the existence of the nanovoids.

The second possibility is related to the heteroepitaxial and capping process strain-induced melting of InAs during overgrowth. For example, the large lattice mismatch-induced heteroepitaxial strain between InAs and GaAs will cause the melting of InAs during the growth on GaAs at 770 K [24]. With almost the same lattice constant of Ge with GaAs, Ge and InAs will also experience quite large heteroepitaxial strain (7.1%) during growth. Although the deposition of InAs QDs on Ge was carried out at 633 K, the strain caused by the lattice-mismatched heteroepitaxial growth and capping process (at 773 K) might lead to the melting of the dots. Because of the higher density of the InAs liquid phase than the solid phase, the volume of the dots shrinks, and the surrounding Ge, which is stiff to resist a structural change, leads to the formation of voids thereafter. During the cooling procedure, in addition, Ge has an even larger TEC than InAs, and thus will shrink faster to its bulk lattice value, locking the location of the nanovoids. This hypothesis can explain the size of the nanovoid and why they did not propagate to the embedment, but it is unable to clarify the hexagonal shape of the void. The formation of the void facets may involve the varied etching rates of As on InAs along different crystallographic directions during the cooling process, which have been well established in III–V materials [25–27]. Other possibilities of void formation can be associated with defect-induced deployment of the growing material, but no direct evidence has been found to support all these possibilities.

5. Conclusion

High-density InAs QDs were successfully grown directly on Ge (100) substrates without any buffer layers by solid-source MBE. The overgrowth of Ge was also investigated in detail. In general, the structure of QDs buried in a Ge matrix is more pyramidal and polyhedral, which is different from that in a GaAs or Si matrix and can be mainly attributed to the differences of the TECs of the InAs/Ge and InAs/GaAs systems and suppressed In diffusion from the dots. It was also found that the embedded QD morphology was modified to a large extent during the capping process. While crystal defects, especially SFs and misfit dislocations, are ubiquitous in QDs in both the 40 nm and 200 nm capping layer samples, nanovoids of 7–9 nm were presented only for the 200 nm capping layer sample. Possible formation mechanisms are proposed but detailed investigation is demanded in the future. For the 200 nm capped sample, short-line indentation planar defects were detected from AFM images, and hints of its formation were also witnessed from STEM images as the propagation of the SFs as the propagation of the SFs.

Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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