Post-thermal-Induced Recrystallization in GaAs/Al$_{0.3}$Ga$_{0.7}$As Quantum Dots Grown by Droplet Epitaxy with Near-Unity Stoichiometry

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ABSTRACT: Here, we investigate the stoichiometry control of GaAs/Al$_{0.3}$Ga$_{0.7}$As droplet epitaxy (DE) quantum dots (QDs). Few tens of core nonstoichiometries in the GaAs atomic percent are revealed in as-grown “strain-free” QDs using state-of-the-art atomic-scale energy-dispersive X-ray spectroscopy based on transmission electron microscopy. Precise systematic analyses demonstrate a successful quenching of the nonstoichiometry below 2%. The control of the chemical reactions with well-controlled ex situ annealing sheds light on the engineering of a novel single-photon source of strain-free DE QDs free of defects.

INTRODUCTION

Self-assembled semiconductor quantum dots (QDs) have gained much attention due to their rich variety of nanotechnological applications in photonic quantum technologies (e.g., information processing, cryptography, and quantum teleportation).1−6 Quantum simulators of chemical and physical systems,7 optoelectronics,8−14 and chemical15−17 or biosensing18,19 Conventionally, three-dimensional islands of Stranski–Krastanov (SK)-type QDs are formed on lattice-mismatched heteroepitaxial systems driven by the elastic relaxation of strain. Experiencing a substantial (intrinsic) strain mismatched heteroepitaxial systems driven by the elastic relaxation of strain. Experiencing a substantial (intrinsic) strain, the chemical composition profiles of SK QDs unfortunately significantly modify their optoelectronic properties, which is an obstacle to the integration of QD single-photon sources in on-chip optical circuits. For decades, droplet epitaxy (DE) has attracted attention as an alternative technique for fabricating deterministic QD structural morphologies that are free of strain.20−30 In the DE mode, liquid droplets of a metallic material (Ga) are formed on a group-III-terminated surface, followed by crystallization under the flux of a group-V element (As) (Figure 1a). While the structural morphology of QDs can be precisely controlled by growth parameters, such as the substrate temperature, the flux of As$_x$, and the surface stoichiometry, the DE technique enables the fabrication of “strain-free” GaAs QDs on a lattice-matched AlGaAs matrix. Such controllability (solely via the crystallization conditions) and the precedent characteristics of DE QDs hold substantial promise for on-chip implementations of QD single-photon sources.

The presence of a small amount of defects in DE QDs, however, can strongly modify their optoelectronic properties and their chemi-physical nature, drastically decreasing the efficiency of photon emitters. Suffering from the low efficiency of optical emission in QD photoluminescence (PL), many studies have reported improving the quality of DE QDs by performing ex situ thermal treatments.22−24,26,27,29,31−33 The optimization of the post-thermal process (relative to improving the crystallinity of QDs) relies on phenomenological enhancements in the intensity of the QD PL. Such analyses need to be put in structural and chemical contexts that nanoscopic studies have been lacking.31−37 Atomic-resolution systematic analyses of such DE QDs are therefore in high demand to acquire a precise chemi-physical understanding to engineer defect-free and strain-free QDs.

In this study, we thoroughly investigate the nonstoichiometry in GaAs DE QDs with and without post-thermal-induced recrystallization processes. Precise structural and chemical investigations are systematically performed for 40 individual QDs using atomically resolved transmission electron microscopy (TEM) energy-dispersive X-ray spectroscopy (EDS). Different degrees of nonstoichiometric cores are revealed in strain-free QDs as a function of the post-thermal-induced recrystallization temperature. While resolving a few to a few tens of compositional changes in the atomic percent, near-
unity stoichiometric QDs are acquired via a careful post-thermal-induced recrystallization process. In addition, we examine the $T_{\text{ex-an}}$-dependent effect of Al interdiffusion on the electronic structures of DE QDs via an envelope function model. Defect-free and strain-free DE QDs of high optical quality would ultimately lead to the on-chip implementation of QD single-photon sources.

**RESULTS AND DISCUSSION**

**Control of Nonstoichiometry in Strain-Free DE QDs.**

Because the GaAs/AlGaAs DE QD system is known to be strain-free, the elemental distribution of GaAs QDs and any chemical changes in a QD are primarily of concern. To atomically resolve the structural and chemical characteristics of strain-free GaAs DE QDs, TEM EDS is performed on cross-sectional specimens with embedded GaAs islands (cf. Figure 1). For the pyramidal/conical shapes of the GaAs DE QDs, atomic-scale analyses of the in-depth profiles reveal a remarkable inhomogeneity in their chemical distributions (cf. Figure 1b,c). In particular, an As-deficient (Ga-rich) QD is resolved in the EDS maps. Here, the control parameters of the structural morphology include the surface stoichiometry, the crystallization temperature, and the As$_4$ flux.\textsuperscript{22} To resolve the two-dimensional (2D) elemental distributions more precisely (with a high signal-to-noise ratio), we also perform line scans of the chemical constituents across the center of each QD along the growth axis (cf. Figure 2). A significant non-stoichiometry ($\sim7:3$ in the Ga/As atomic ratio) is revealed to
be present at the core of as-grown DE QDs. The primary mechanism responsible for metal-rich nonstoichiometric QDs is the insufficient diffusion of As atoms into the Ga liquid droplets (i.e., the Ga reservoirs) under the surface impinging density of As.\textsuperscript{22,27} The incomplete crystallization of GaAs DE QDs results in a gradient of the As composition in the droplet leaving the nonstoichiometric core. The As-deficient (Ga-rich) droplets are seen to turn into stoichiometric QDs at an enhanced temperature in the post-thermal-induced recrystallization process. The exemplary Ga depth profiles in Figure 2 show a remarkable improvement in the crystallinity by a few tens of atomic percent for four distinct temperatures of ex situ annealing.

To gain more systematic and profound insights into this process, we perform a statistical EDS of 40 individual QDs, 10 QDs for each of the cases with and without post-thermal-induced recrystallization treatments at various $T_{\text{ex-an}}$. The maximum deviations between Ga and As in the atomic percent ($C_{\text{Ga–As}}$) are displayed in Figure 3 for the four distinct treatments. With no deliberate ex situ annealing treatment, the GaAs DE QDs present a significant mean nonstoichiometry of 37% ($C_{\text{Ga–As}}$) with an atomic ratio of Ga/As ranging from 1.5 to 3.7 ($C_{\text{Ga–As}} = 10.1–28.5$). Conversely, Keizer et al.\textsuperscript{34} reported an X-ray scanning tunneling microscopy (X-STM) analysis of as-grown DE QDs with no recognizable nonstoichiometry. While the X-STM probes the surface characteristics at the atomic scale, the additional capability of TEM EDS identifies the elemental compositions by measuring the energies of the characteristic X-ray peaks. At the same time, $C_{\text{Ga–As}}$ is measured as varying from $\sim$20 to $\sim$58% in a given specimen (cf. Figure 3). The measured PL signals of as-grown QDs\textsuperscript{31,32} seem to arise relative to QDs with a low degree of nonstoichiometry. A nearly identical trend is seen in the elemental profiles of GaAs DE QDs with thermal treatment at 750 °C, even though the observed nonstoichiometry ratios fall in a smaller variance from 1.9 to 3.1. $C_{\text{Ga–As}}$ is seen to diminish by more than 16% with an increase of 50 °C in $T_{\text{ex-an}}$ from 750 °C. Encouragingly, a few tens of the still existing core nonstoichiometry are dramatically quenched below $\sim$4% for GaAs DE QDs post-thermal recrystallized at 850 °C. The average standard deviation of $C_{\text{Ga–As}}$ is extracted to 1.6% for DE QDs annealed at 850 °C. Here, the overall analytical accuracy of EDS analysis is approximately 2% (see details in Experimental Section and Computational Methods). Further reaction of the As atoms, therefore, eliminates the As composition gradient within the dot. Such recrystallization processes can be understood as reordering the As and Ga atoms dissolved within the GaAs DE QD. The diffusion coefficient of As (e.g., $5.8 \times 10^{-14}$ at 850 °C) has been estimated to be too low to permit bulk equilibrium via vacancy diffusion from the surface at the annealing temperatures.\textsuperscript{35} Accordingly, the local rearrangement of defects has been identified as a viable mechanism for producing uniformity during post-growth annealing.\textsuperscript{33,36} The localized dissolution of As atoms from the substrate would create nonstoichiometric defects, whereas near-stoichiometry in GaAs is observed at the bottom of the QDs (cf. Figure 1). Remnants of metallic Ga within the GaAs DE QDs have also been reported by Mano et al.\textsuperscript{37} We note that individual phases of Ga and GaAs have been resolved to coexist within the GaAs nanostuctures with no post-thermal treatment. The in situ annealing process seems to induce the nonstoichiometric phases of Ga and As for our as-grown DE QDs. Compared to the ambiguous optimization processes of post-thermal annealing (relying on the phenomenological enhancements in the QD PL intensity), the EDS statistics reveal a dramatic recovery of the crystallinity in 50 °C from 800 °C (cf. Figure 3). We note that the operating temperature of rapid thermal annealing (RTA) is measurement-based (i.e., based on the position of the temperature sensor and the Ar environment), and therefore no absolute value is given. For example, Bocquel et al.\textsuperscript{27} performed post-thermal annealing at 750 °C to complete the crystallization.

Three-dimensional composition profiles of DE QDs have also been retrieved using atomic probe tomography (APT) across a volume of several tens of nanometers. The TEM EDS employed in this study facilitates the comparative statistical analysis by scanning a sample cross section of 2 orders of magnitude larger than that of APT. Here, our methodology shows how strain-free but nonstoichiometric QDs are converted into stoichiometric DE QDs at well-controlled temperatures of ex situ annealing. This provides powerful new insights into the engineering of strain-free QDs with unity stoichiometry.

**Interdiffusion of Detrimental Al Atoms in Strain-Free QDs.** Along with the control of the nonstoichiometry, the interdiffusion of detrimental Al is also a critical issue in DE QDs,\textsuperscript{27,29,31,32} and disturbs the optoelectronic properties of single-photon emitters. We carefully examine whether the ex situ annealing process degrades the purity of the strain-free DE QDs. Figure 2 confirms a low level of Al intermixing for most of the QDs. Limited by the inevitable position drift and damage of the thin samples, the average Al elemental profiles of the EDS line scans level off at 4% in the central regions of QDs with widths of 2 nm. We note that $\sim$1.4% of the average Al composition was retrieved with an average deviation of $\sim$0.4% on the host material of $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ for a scanning strip with a width of $\sim$20 nm. The Al defects underneath the QDs originate from the recrystallization of Al atoms, which dissolve into the Ga droplet.\textsuperscript{27,28} Meanwhile, the local enrichment of Al at the top facet is ascribed to the different mobilities of Ga and Al atoms. We also examine the influence of Al interdiffusion in the electronic structures of DE QDs employing the envelope function scheme within the $k\cdot p$ theory (see details in Experimental Section and Computational Methods). The sub-band energies $E_{\text{sub}}^{\text{Ga/As}}$ in the conduction/valence band and...
the direct Coulomb interaction energy $J$ of an electron–hole pair are evaluated for lens-shaped QDs. The average radius of the DE QDs is taken to be 15 nm, and the average heights are found to be 5.5, 7.7, 7.5, and 8.9 nm for the as-grown QD and the QDs post-annealed at 750, 800, and 850 °C, respectively. The results of $E_{\text{conf}}$ and $J$ for the ground sub-bands are summarized in Table 1 for various temperatures of post-thermal-induced recrystallization. $E_{\text{conf}}$ and $J$ denote the results of an ideal confinement potential in the absence of Al interdiffusion. To configure a more realistic profile of band gap, the experimental mole fraction $x(z_{\text{QD}})$ of the Al inquired in EDS is applied to an empirical band gap $E_{\text{fl}}$ of $E_{\text{fl}}(x)=1.519+1.155x+0.37x^2$. Here, $z_{\text{QD}}$ denotes the position measured from the bottom to the top facet along the growth axis of a QD. Our analysis shows that due to the interdiffusion of Al within the QDs, the QD recombination energy is modified by $\sim 1.5\%$ for the as-grown sample. The QD excitonic energy is reduced by $\sim 2.7\%$ via the post-thermal-induced recrystallization treatment up to 850 °C (see theoretical details in Experimental Section and Computational Methods). The interdiffusion effect on the band gap via $x(z_{\text{QD}})$ yields $\sim 46.4\%$ of the quantum confinement correction (i.e., the sum of the excitonic sub-band energy and the Coulomb interaction of $E_{\text{sub}}+J$) for the annealed samples, while it yields $\sim 32\%$ of the quantum confinement correction for as-grown QDs. In our calculation, a finite $T_{\text{ex-an}}$ treatment gives rise to a minor change of $\pm 0.2\%$ in $E_{\text{sub}}+J$ for the specimens treated at the three distinct temperatures. Therefore, we demonstrate that the optimal post-thermal-induced recrystallization process quenches the detrimental core nonstoichiometry of strain-free DE QDs. The $T_{\text{ex-an}}$-dependent degradation of the QD purity by Al is believed to be minor.

**CONCLUSIONS**

We have systematically demonstrated full control of the detrimental nonstoichiometric defects in strain-free DE QDs by employing a post-thermal-induced recrystallization process. Harnessing unambiguous high-resolution TEM EDS, the nonstoichiometries present in the core of strain-free DE QDs have been quantitatively revealed. Chemically and spatially resolved complexes of as-grown DE QDs have an average nonstoichiometry of 6.85:3.15 (Ga/As) in the atomic percent. Using statistical analyses of TEM EDS for 40 individual DE QDs, we have demonstrated a successful approach to eliminate the core nonstoichiometry via a careful ex situ annealing process. At the threshold temperature, a near-unity stoichiometry has been attained in our strain-free QDs within the analytical accuracy of EDS. The effect of Al interdiffusion (versus the treatment temperature) on the quantum confinement characteristics is believed to be minor. In addition to shedding light on the $T_{\text{ex-an}}$-dependent nanoscopic machinery, this novel engineering of strain-free QDs with unity stoichiometry is critical for the practical on-chip implementation of QD single-photon sources.

**EXPERIMENTAL SECTION AND COMPUTATIONAL METHODS**

Fabrication of GaAs DE QDs. The GaAs QDs under investigation were grown by DE using molecular beam epitaxy. As illustrated in Figure 1, the epitaxial growth of GaAs QDs in the DE mode is based on the crystallization of Ga droplets via the injection of an As$_4$ flux. The crystallized GaAs QDs are then capped with an AlGaAs protective layer. To form the Ga droplet, the partial pressure of As ($P_{\text{As}}$) in the chamber was maintained below $\sim 2 \times 10^{-12}$ Torr prior to the injection of the Ga flux. We fabricated the Ga liquid droplets on an Al$_{0.3}$Ga$_{0.7}$As/GaAs substrate by supplying two monolayers of Ga at $T \sim 220$ °C. The growth chamber background pressure was below 10$^{-10}$ Torr during the injection of the Ga atoms. As depicted in Figure 1a, the As atoms were subsequently supplied to impinge on a Ga-terminated (Ga-rich) surface with the reconstruction of (4 × 6) under a moderate As$_4$ beam equivalent pressure (BEP) of 1.0 × 10$^{-5}$ Torr. The morphological reconstruction of the DE droplet involves the migration, accumulation, and desorption of Ga on the (4 × 6) Ga-rich surface (diffusion-stopping processes). After the crystallization time of 60 s, the GaAs DE QDs were thermally annealed at an increased substrate temperature of $\sim 560$ °C (high temperature) with a reduced As$_4$ BEP of 6.0 × 10$^{-6}$ Torr. While controlling the shapes of nanostructures with higher thermal stability, we maintained the crystallization process of GaAs droplets for 720 s. We note that the migration of the Ga droplet is regulated with the control of the substrate temperature and the surface impinging density. The GaAs DE QDs were then protected with a 20 nm-thick Al$_{0.3}$Ga$_{0.7}$As layer at 560 °C. The average QD density of $\sim 3$ × 10$^{10}$ cm$^2$ was obtained by measuring the images of a charge-coupled device for optically excited samples. A detailed description of the growth mechanism and parameters can be found in our previous works. We note that the structural morphology and optical characteristics have also been reported for the similar type of DE QDs.

Ex Situ Thermal Annealing Process. For systematic control of the stoichiometry of DE QDs, post-growth RTA processes were performed on as-grown QDs as a function of temperature in a standard RTA equipment. Each specimen was heated up to 750, 800, and 850 °C within 60 s and maintained for 240 s in ambient Ar. Then, the sample was cooled below 100 °C over 15 min. The cycling of ex situ RTA is optimized based on the phenomenological enhancement of PL intensity. We note that previous studies have shown a decrease of PL intensity for DE QDs post-annealed above 850 °C.

**TEM and EDS Analyses.** The high-resolution structural analysis was performed by employing a Titan 80-300 TEM operating at 300 kV. We used focused ion beam (FIB) techniques for the preparation of specimens, in which QD droplets are randomly embedded. To resolve structural profiles of all constituents in DE QDs precisely, chemical characterization with compositional mapping was carried out using a Talos F200X of 200 kV. The EDS elemental maps and line scans were measured in FIB-prepared cross-sectional specimens with a thickness of less than 50 nm. While 50% of the As concentration is theoretically estimated in the matrix of...
Al$_x$Ga$_{1-x}$As, the average concentration profiles of As were measured to be 52.3% with a standard deviation error of 1.8% for a scanning strip with a width of 20 nm. The 2D mapping could include an incorrect distribution of elements due to the unavoidable spatial drift and damage of thin samples during several minutes of data collection time. For statistical analyses of 40 individual QDs, we performed the EDS line scans for a faster elemental mapping to increase the contrast of elemental distributions. Given the point resolution of 1.6 Å in scanning transmission electron microscope mode, the in-depth concentration profiles of DE QDs were seen by scanning across the center of the QDs in the growth direction [001]. The TEM EDS maps and line scan profiles in Figures 1 and 2 probe small compositional changes of Ga, As, and Al in the DE QDs post-annealed at distinct temperatures.

**Calculation of QD Quantum Confinement Energies.**
We consider a lens-shaped DE QD of GaAs/Al$_x$Ga$_{1-x}$As with in-plane cylindrical symmetry, whose base length and height are extracted from the cross-sectional TEM images. We employ the envelope function scheme based on the k-p theory to calculate the quantum confinement characteristics of DE QDs. The excitonic recombination energy $E_X$ of QD is given by the sum of the fundamental band gap $E_{sub}$ of QD, the sub-band confinement energies $E_{sub}$ of carriers, and the electron–hole Coulomb interaction energy $J$: $E_X = E_{sub} + E_{sub} + J$. The latter two terms (i.e., $E_{sub}$ and $J$) in $E_X$ describe the quantum confinement characteristics of an electron–hole system. The QD sub-band energies and wavefunctions are calculated by numerically solving a Schrödinger equation written as

$$\begin{align*}
-\frac{h^2}{2m^*} & \left( \frac{1}{m^*} \frac{\partial}{\partial \rho} \left( \frac{\rho}{m^*(\rho, z)} \frac{\partial}{\partial \rho} \right) + \frac{\nu^2}{m^*(\rho, z)z^2} \right) + \frac{1}{m^*(\rho, z)} \frac{\partial}{\partial z} \left( \frac{1}{m^*(\rho, z)} \frac{\partial}{\partial z} \right) + V(\rho, z) \right) \psi(\rho, z) \\
&= E \psi(\rho, z)
\end{align*}$$

To conform realistic quantum confinement potentials inside the DE QDs, the empirical distributions of Al in the atomic percent are least-squares-fitted with a polynomial function of $x(z_{CD})$ to an empirical gap formula of $E_{sub,Al_{1-x}}(z_{CD}) = 1.519 + 1.155x + 0.37x^2$. The conducton band offset is taken to be 60% of the band gap difference. The direct Coulomb energy $J$ projected onto an exciton state $|\psi_{\alpha,\beta}(\rho, z)\rangle$ is written as

$$J = -e^2 \sum_{\alpha,\beta} \int \int d^3 r' d^3 r \times \frac{\psi_{\alpha,\beta}^* (\rho, r') \psi_{\alpha,\beta} (\rho, r)}{|r-r'|^2},$$

where the Coulomb interaction is screened by the local dielectric function $\varepsilon(r-r')$ for a given position $r$ with spin $\sigma$. The material parameters used in the calculation can be found in refs 38–40.

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**Notes**
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