Research Article

Epitaxial Growth of Optoelectronically Active Ga(As)Sb Quantum Dots on Al-Rich AlGaAs with GaAs Capsule Layers

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1. Introduction

Inorganic quantum dots (QDs), especially concerning their potential use in semiconductor lasers, have been in the research focus for 25 years now [1, 2]. In particular, Ga(As)Sb quantum dots (QDs) grown on GaAs [3–13] are of interest as an active material for efficient lasers in the near- and midinfrared. These lasers could, for example, be used for sensitive spectroscopy or even long-distance optical communications via glass fibers [11]. On account of the strong hole confinement and (due to a type-II band alignment) no electron confinement, each QD might also be suitable for use as a binary storage cell in a quantum dot memory device [11, 14–16]. On the one hand, to increase the degree of design freedom, in principle, the QDs could also be grown onto an Al-rich Al1-xGa1-xSb layer (with x >10%) as the barrier layer. In the case of antimonide QD applications in semiconductor lasers, this would potentially increase the range of available emission wavelengths. In the case of antimonide QDs as binary storage cells, this approach would raise the confinement energy $C$, but it would also result in a transition from a type-II to a type-I band alignment. This means that not only the holes but also the electrons will be confined, such that—by trend—the carrier lifetime will be reduced due to an increased rate of electron-hole recombination.

On the other hand, the biggest challenge in using Al-rich barrier layers for Ga(As)Sb QDs is the fact that they are
usually not optoelectronically active [17, 18], although QD-like objects might evolve at these barriers during Stran斯基–Krastanov growth (SK); see Figure 1.

This has been attributed to crystal defects at the QDs’ bases, which lead to mismatch dislocations, which dislocate the whole QDs by 1 ML [19]. The resulting stress/strain cannot relax, especially in the case of Al-rich surrounding material.

Thus, despite their promising potential applications, the growth of active antimonide QDs at and in between Al-rich barriers with strong luminescence does not seem to be feasible.

In this contribution, we show that the thin, so-called “capsule layers” made from GaAs between the QD layers and the original Al-rich barriers can solve this problem.

In this case, each capsule layer has to be considered part of a new (double-layer) barrier. With that, however, the confinement energy and the photon energy of the luminescence emission might be altered to some extent. In the case of antimonide QD semiconductor lasers, this varies the range of possible emission wavelengths. For the use of QDs as binary storage cells, two aspects have to be considered: either type-II band alignment can be maintained. At the same time, the hole lifetime can even be increased further, or the band alignment inadvertently changes to type-I, making electron-hole recombination more likely and thus reducing the carrier lifetime.

2. Antimonide QD Growth

Each sample is grown on an undoped (001) GaAs substrate with an R450 molecular beam epitaxy (MBE) machine from DCA Oy, Finland. The beam equivalent pressure of Ga differs slightly from sample to sample, that is, between 1.6 and 1.8×10⁻⁷ hPa. The Sb/Ga (V/III) flux (or partial pressure) ratio is adjusted with a valved Sb cracker from ADDON, Carrières-Sur-Seine, France. Wafers are bought “epi-ready” from Wafer Technology Ltd., Milton Keynes, UK.

For each sample, first, a 200 nm thick GaAs buffer layer is grown on the substrate with 0.3 ML/s (monolayer per second) at a GaAs substrate temperature of 630°C, the latter measured with a pyrometer. Then, the sample is cooled down to the chosen quantum dot growth temperature, that is, to 530°C for all cases shown here. The GaSb material is deposited on an As-rich surface with a nominal QD growth rate of also 0.3 ML/s.

The Stranski–Krastanov (SK) growth of binary GaSb QDs on GaAs would be associated with a 7.8% lattice mismatch. Due to As and Sb intermixing (resulting in GaAsSb QDs) [20, 21], the growth of these QDs is more complex than that of InAs QDs on GaAs. The epitaxial process depends on the exact control of the Sb/Ga (V/III) flux ratio, the growth temperature, and the nominal coverage [16, 18, 22].

An increased Sb/Ga ratio results in a red-shift of the luminescence or laser emission due to lower As content in the QDs and the size of the QDs, which tend to get larger. However, an increase in the growth temperature for a constant Sb/Ga ratio leads to a blue-shift due to stronger As in-diffusion into the QDs. This internmixing of Sb and As dominates the luminescence peak shift; that is, the corresponding blue-shift usually overcompensates for the red-shift of larger dots.

Thus, for example, to achieve a maximum emission wavelength, the As content within Ga(As)Sb QDs has to be as low as possible. Other authors introduced pre- and postsoaking steps with Sb before and after QD growth [23]. In work leading to [11, 16, 18], we successfully achieved strong photoluminescence (PL) emission from QDs with a wavelength peak at 1.309 μm. To do so, we applied a growth protocol with a 2:1 Sb/Ga flux ratio, a substrate growth temperature of 530°C, and a nominal coverage of the growth front of 2 ML. Furthermore, we interrupted the QD growth for 10 s while keeping the Sb flux turned on after the deposition of the QD material as a long postsoaking step. This growth interruption stabilizes the surface and suppresses As/Sb intermixing to some extent [11]. With this optimized growth protocol, we could obtain GaAsSb QDs with an As content of only around 30% and Sb content maximized to about 70% [16, 18]. This protocol is mentioned here exemplarily to report the growth parameters which are optimal for long-wavelength emission. For the results described in this contribution, slightly modified growth protocols have been applied.

3. Antimonide QD Design

The design of QDs also includes the design of the barriers, by which the dots become quantum dots, that is, three-dimensional potential wells. Figure 2 reveals the energy level/band schemes (under flat band conditions) for three different barrier materials and a hypothetical pure GaSb QD in each case, that is, an antimonide QD without any As contents. These values have been calculated with the nextnano++ software by nexnano GmbH, Poing, Germany, which will be explained in some more detail below. The valence band offset $D = C + L$ of the bulk material for the hole QDs can be divided into the confinement energy $C$ (energy difference of the hole state in the QD and the bulk valence band edge of the material used for the QDs) and the localization energy $L$ (energy difference of the hole state in the QD and the potential barrier edge). The larger the Al content in the barriers, the larger both $D$ and $L$—assuming the same QD size and shape, that is, for our calculations: pyramidal QDs with a base width of 25 nm and a height of 3 nm.

Since it does not seem possible to grow optoelectronically active antimonide (Ga(As)Sb) QDs on Al$_{x}$Ga$_{1-x}$As layers with high Al-contents ($x > 10\%$), here we suggest capsule layers in between the Al-rich (formerly single-layer) barrier and the pure QD layer. These capsules to the QDs are designed to be made from pure GaAs and to have a thickness of 5–20 nm. To protect the Al$_{x}$Ga$_{1-x}$As layers (especially with high Al contents) from oxidation, the surfaces can be passivated to ensure the long-term stability of the components [24–27].
4. Numerical Results on QD Coupling

There are two main reasons for the coupling of QDs, both “horizontally” (within a QD layer) and “vertically” (from one QD layer to the next): mechanical stress/strain exerted from neighboring QDs and quantum-mechanical coupling due to the overlap of the wave functions.

To get an understanding (before investigating double-layer barriers) on how strongly both of these reasons for coupling affect the spectral luminescence peak position, we have pursued numerical calculations again using the nextnano++ software by nextnano GmbH, Poing, Germany, for the case of pure GaAs (single-layer) barriers. The program is based on an 8-band $k\cdot p$ algorithm [28, 29].

Mechanical stress/strain is automatically considered in the program, but stress relaxation by the evolution of crystal defects (one-dimensional or dislocations) is not because specific positions of the atoms corresponding to the $\alpha$ zinc blende crystal structure are assumed.

4.1. Scheme of Numerical Calculations. The nextnano++ software, which is based on a perturbation-theoretical approach, is useful to determine the quantum-mechanical eigenfunctions and eigenstates of the charge carriers, for example, within QDs. The solutions are developed locally around and at any specific point in momentum space, for example, at the $\Gamma$ point (as we do here). The nextnano++

Figure 1: Atomic force micrographs of (a) active Ga(As)Sb QDs on GaAs and (b) nonactive QD-like objects grown from Ga(As)Sb on Al$_{0.5}$Ga$_{0.5}$As. Be aware of the different scale in the height dimension as opposed to both lateral dimensions along the growth front.

Figure 2: Electronic/hole QD energies and bulk material band edges for three QD heterostructures with different Al contents $x$ in the single-layer barriers (flat band conditions including stress/strain). Pyramidal QDs with 25 nm base width and 3 nm height are assumed.
software takes care of any mechanical stress/strain and the localization of the carriers in all three dimensions in the QDs.

Physical parameters, such as temperature, have to be chosen, as well as the model to describe the lattice stress/strain.

A three-dimensional spatial mesh is defined. For each point of the mesh, the material composition has to be chosen. Schroedinger’s and Poisson’s equations are solved iteratively until a self-consistent solution is reached (if at all). For different spatial areas, the software user can determine whether the calculation should be performed considering quantum-mechanics (i.e., Schroedinger and Poisson) or classically (i.e., Poisson) only. In the first case, both stress/strain and quantum-mechanical coupling will be taken into account; in the second case, coupling via stress/strain only will be taken into account.

For the calculations, we assume an orderly 3 × 3 array of QDs arranged in a square in all of the QD layers.

When quantum-mechanical effects need to be considered, the volume of the quantum dot itself and the volume around it for a distance of up to 6 nm in all six directions from the outer edges (or the tip) of the quantum dot are included. Calculations for distances of more than 6 nm, however, would result in a tremendous increase in computing time.

This approach means that both the coupling via stress/strain and the quantum-mechanical coupling is taken into account as long as distances between the QDs of less than 6 nm are assumed. For larger QD distances, however, which resemble the situation of our grown samples more closely, the calculations only reflect the influence of stress/strain on the carriers’ energy levels.

4.2. In-Plane Coupling. For the calculations, we assume GaAs$_{0.3}$Sb$_{0.7}$ QDs of pyramidal shape with a base width of 25 nm, a height of 3 nm, and an Sb content of (1-y) = 0.7.

A square-shaped 3 × 3 array of these GaAs$_{0.3}$Sb$_{0.7}$ QDs is assumed for each calculation. Their “horizontal distance” is defined as the geometrical distance between the edges of any two nondiagonally neighboring QDs. For example, for a nominal distance of 12 nm, the tips of two such QDs are [(25/2) + 12 + (25/2)] nm = 37 nm apart. Our calculations are done for nominal horizontal QD distances of 1 nm, 2 nm, 12 nm, and 27 nm.

The results are summarized in Figure 3. The quantities $\Delta E_{2s}$, $\Delta E_{hh}$, and $\Delta E_{so}$ denote the shift of the bulk conduction band edge, the bulk valence band edge for heavy holes (hh), and that of the bulk valence band edge for split-off holes (so), respectively.

The differences in energy shifts for the 3 × 3 QDs are negligible on this scale, and only the mean values are given here. For the luminescence emission wavelength shift $\Delta \lambda$, the differences in the results for the nine QDs are larger so that the range is given here via the light-red shaded area. Differences between the values for the central QD and the four QDs at the corners of the 3 × 3 array/square and the four QDs in the middle of the square sides should be expected from the geometrically different coupling to adjacent QDs. However, even within any of these QD classes (e.g., QDs at the corners of the square), there are slight differences, which are related to the way the nextnano++ software algorithm computes the spatial mesh.

It is obvious from Figure 3 that there is a nonnegligible luminescence emission wavelength shift due to horizontal coupling via stress/strain as well as quantum-mechanically. For nominal horizontal QD distances of some nanometers, the wavelength shift can even exceed values of 50 nm. The shift is always positive and thus has the same qualitative impact as an increase in the size of the QD or a reduction in As content. For horizontal QD distances > 6 nm (per definition of the calculation conditions) only stress/strain is incorporated for the calculated wavelength shift, which can still exceed tens of nanometers.

4.3. Cross-Plane Coupling. In Figure 4, the results of the calculations for an additional vertical coupling of the GaAs$_{0.3}$Sb$_{0.7}$ QDs are shown. “Additional” means that the values given in the diagram are stated relative to the “new” levels, that is, already shifted due to horizontal coupling according to Figure 3. The calculations are pursued by the numerical implementation of two 3 × 3 square arrays of the same orientation on top of each other. The nominal horizontal distance between QDs is kept constant at 2 nm within both 3 × 3 arrays, in equivalence to a situation with QDs situated very close to each other.

The mean energy shifts for the two central QDs in the two 3 × 3 arrays are given in Figure 4 since they resemble the more realistic situation where others surround every QD. The fact that there are differences between the results for the two central QDs has to be attributed to the direction of working through the mesh by the software algorithm.

For these calculations, the nominal vertical distance of QDs on top of each other has been varied. This distance is defined by the one between the tip of the QDs in the “lower” QD layer and the base of the QDs in the “upper” QD layer. The values, which have been assumed for the calculations, are 2 nm, 22 nm, and 47 nm, respectively.

For the heavy hole (hh) and the split-off hole (so) energy level shifts, the differences for both central QDs are negligible. For the bulk conduction band edge shift, the differences are not negligible, but for the sake of readability of the diagram, only the mean values are shown in Figure 4 together with the mean values for the wavelength shifts $\Delta \lambda$.

As can be deduced from this figure, vertical coupling also leads to considerable (positive-valued) luminescence emission wavelength shifts. For small vertical QD distances of a few nanometers, the additional shift comes close to shifts due to the horizontal interaction; that is, it reaches values of around 40 nm. Therefore, for small QD distances, the overall wavelength shift due to both in- and cross-plane coupling (via mechanical stress/strain and quantum-mechanically) can show values of 80 to 90 nm.

This (positive) wavelength shift value comes close to the (negative) value, which can be expected from Figure 5 according to [16, 18] for the case of uncoupled QDs with
25 nm base length and 3 nm height and a reduction of the Sb content in the QDs from 65% to 55%, as calculated with
nextnano++.

For larger QD distances, where only coupling by stress/strain is taken into account, but not quantum-mechanical coupling, the wavelength shift can still reach up to 25 nm, which is in good quantitative agreement with the experimental results further below.

5. Photoluminescence (PL) Results

For comparison, to gain some understanding of the influence of the GaAs capsule layers, we experimentally investigate eight different samples with 4 Ga(As)Sb QD layers and pure GaAs (Al-free single-layer) barriers in between. For any one of the eight samples, the barrier thickness is kept constant. The barrier thickness is varied, from 10 to 80 nm in
steps of 10 nm. The base width of these QDs is 25 nm, and their height is 3 nm.

Figure 6 reveals experimental photoluminescence (PL) results for these eight samples. Although there are growth tolerances, which cause unexpected deviations from the general trend (e.g., the PL peak intensity for a barrier thickness of 60 nm falls short), there is evidence that coupling is weak for barrier thicknesses $>50$ nm, where the PL peak positions and heights do not change strongly anymore.

The value of $50$ nm barrier thickness gives an approximate lower limit for the single-layer barrier thickness desirable for growth processes. For double-layer barriers, the height and thickness of the GaAs capsule layers have to be considered, too.

Therefore, we start to use double-layer barriers made from $\text{Al}_{x}\text{Ga}_{1-x}\text{As}$ on the far side of any QD layer and the GaAs capsule layer adjacent to any QD layer. So in between any two of the 4 QD layers (grown as before), there are now two GaAs capsule layers (one from each QD layer) and one $\text{Al}_{x}\text{Ga}_{1-x}\text{As}$ layer in between. A corresponding energy band diagram for $\text{Al}_{0.9}\text{Ga}_{0.1}\text{As}$ layers is shown in Figure 7.

In a first test run, we use 20 nm thick GaAs capsule layers and $\text{Al}_{0.5}\text{Ga}_{0.5}\text{As}$ layers ($x = 0.50$) with varying thicknesses from sample to sample, that is, 10 nm, 30 nm,
and 40 nm. The PL results are given in Figure 8. The results are similar to those in Figure 6 for the case of single-layer barriers. If the Al\(_{0.5}\)Ga\(_{0.5}\)As layers are too thin (<40 nm), stress/strain cannot be relieved from one QD layer to the next one, and the PL intensity is weak. Thus, from now on, we choose Al\(_{x}\)Ga\(_{1-x}\)As layers of a minimum thickness of 40 nm.

Next, we employ three samples with 4 QD layers (grown as before) and 40 nm thick Al\(_{0.5}\)Ga\(_{0.5}\)As layers as parts of the double-layer barriers. The GaAs capsule layers are 5, 10, and...
20 nm thick, respectively, for the three different samples. The ploluminescence results in Figure 9 reveal that a capsule thickness of 5 nm leads to a relatively low PL peak intensity. Obviously, for too thin GaAs capsule layers, the detrimental influence of the large interface energy of the Al$_{0.5}$Ga$_{0.5}$As layers is still too strong. We conclude that the GaAs capsule layer thickness should be at least 10 nm, in the case of an Al$_{0.5}$Ga$_{0.5}$As layer as the Al-rich barrier part.

For the final sample run, we use a 100 nm thick Al$_{0.9}$Ga$_{0.1}$As (now $x = 0.9!$) barrier layer, in two samples with different GaAs capsule layer thicknesses of 10 nm and 20 nm, respectively. The electronic/hole energy band scheme
corresponds to that given in Figure 7, but now only 1 QD layer (grown as before) is used in these two samples. The PL results in Figure 10 reveal that now the capsule layers should rather have a thickness of 20 nm or more in order not to let the interface energy of the Al$_{0.9}$Ga$_{0.1}$As layers deteriorate the optoelectronic activity of the QDs.

From the results in Figures 8 through 10, it can be deduced that a capsule layer will make the epilaxial growth of Ga(As)Sb QDs “on” Al-rich barrier layers possible, if thick enough GaAs capsule layers are introduced, with each of the latter being part of a new (double-layer) barrier. For increasing the Al-contents of the Al-rich layers, the thickness of the capsule layers has to be increased to keep the QDs optoelectronically active.

6. Conclusions

To increase the degree of freedom for antimonide (Ga(As) Sb) quantum dot (QD) design, Al-rich Al$_{1-x}$Ga$_x$As barriers would be helpful. Unfortunately, those QDs cannot be grown next to such Al-rich layers since the interface energy would make the QDs optoelectronically nonactive.

In this contribution, we have verified experimentally that pure GaAs capsule layers between the QD layer and the Al-rich layers on both sides of the QD layers can solve this problem. The QDs will be optoelectronically active, as long as the capsule layers are thick enough, depending on the Al contents of the Al-rich barrier layers.

Each capsule layer then forms a part of any barrier; that is, each barrier is made up of two layers, which has to be taken into account for the QD design, for example, when aiming at a specific wavelength for the luminescence emission peak.

As our numerical calculations indicate, coupling of neighboring QDs via mechanical stress/strain and quantum-mechanical has to be taken into account in the numerical design since this can cause a spectral shift of the luminescence emission peak of several tens of nanometers. QD layer sequences as active material in semiconductor lasers are useful for applications where specific emission wavelengths are needed, such as infrared spectroscopy.

Data Availability

The data used to support the findings of this study are included within the article. Should further data or information be required, these are available from the corresponding author upon request.

Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this paper.

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