2 Ultra-High Density Lightwave Communication

2-1 Fabrication Technique of Ultra-high Density Semiconductor Quantum Dot

AKAHANE Kouichi, YAMAMOTO Naokatsu, GOZU Shin-ichiro, UETA Akio, OHTANI Naoki, and TSUCHIYA Masahiro

Strain-compensation methods were developed to multiply self-assembled semiconductor quantum dot (QD) layer. Although self-assembled technique in lattice mismatched material systems have been attracted much attentions to fabricate semiconductor nano-structures, for example, semiconductor QD, it is difficult to fabricate high density QD because of accumulation of strain energy. In this research, we fabricated ultra-high density QD which was stacking of 150 QDs layers by using strain compensation methods. The density of QDs exceeds 5×10^{12} /cm² which is world's highest density. A strong emission from this sample was observed at around 1.5 µm even at room temperature. The emission wavelength is suitable for fiber-optic communications systems. Therefore, our technique for growing stacked QDs has potential in applications for constructing novel high-performance QD devices for these communications systems.

Keywords

Semiconductor quantum dot, Self-assembling, Strain compensation

1 Introduction

A semiconductor QD is a three-dimensional nano-scale structure that confines small particles, such as carriers (electrons and holes) doped in semiconductor materials and the excitons generated by the electron-hole pair. Structures that can restrict the spatial degree of freedom of electrons and other particles are referred to as quantum structures. When the restricted spatial degrees of freedom are onedimensional, two-dimensional, and threedimensional, the structure is referred to as a quantum well (QW), a quantum wire (QWr), and a QD, respectively. As shown in Fig. 1, the density of states of these quantum struc-



tures—in other words, the number of states at a certain energy—changes from a parabolic shape for the bulk structure to a step shape (QW), a saw-tooth shape (QWr), or a deltafunction shape (QD). Accordingly, the behavior of light absorption and emission are considered to change sequentially, which is expected to induce a change in the optical response.

In QDs, the carriers are concentrated at a certain energy value, theoretically leading to laser diodes with extremely low thresholds. Moreover, the temperature dependence of the threshold current disappears when using QDs with semiconductor lasers. This characteristic is due to the change in density of the states of the OD. In other words, the density of states changes continuously in a normal bulk material or in a QW structure. (The density of states of a QW change stepwise at certain energy values but maintains a consistent value at other energies.) Thus, when the device temperature increases, the injected carriers are redistributed, and the carrier density-which contributes to laser oscillation-decreases. As a result, new carriers need to be injected for proper laser oscillation. Therefore, the threshold current for laser oscillation generally shows a tendency to increase as the temperature increases. On the other hand, a QD laser features a delta-function-form density of states, so that only discrete values are allowed for the carrier energy, even if the carriers attempt to redistribute themselves when the temperature increases. In other words, the redistribution of the carriers is suppressed. This suppression maintains the density of carriers of a certain energy value before and after the temperature increase, which suppresses the increase in the threshold current that would otherwise result from the increase in temperature. Thus, we can create a situation in an ideal QD laser in which the threshold current is completely independent of temperature[1]. Of course, it is difficult to fabricate completely ideal QDs, and we have not yet produced a laser with a threshold current that is completely independent of temperature. Nevertheless, among semiconductor lasers today the laser with the smallest temperature dependence on threshold current is achieved by the QD laser.

When we use QDs, we can implement a highperformance semiconductor laser that does not require a cooling mechanism, so that the system configuration will be simple and inexpensive when it is applied to optical communication systems.

Further, there are high expectations for semiconductor QDs in the application of future information communication technologies such as quantum information processing and quantum communication. As discussed earlier, QDs have a three-dimensional confinement structure, and many research groups all over the world are now attempting to perform quantum information processing by using confined excitons-combined states of electrons and holes-in this confinement structure and by applying coherent control to the excitons. Attempts are also underway at applying QDs to interception-free quantum communication by controlling each of the photons generated by QDs[2]. The advantage of implementing these processes with semiconductor QDs is that doing so will enable the production of smaller devices compared to other methods.

QDs have various particular characteristics as shown above. This introductory research reports on a method for extremely dense fabrication of QDs, with a view to application in fiber-optic communications.

2 Fabrication methods for semiconductor QDs

Quantum structures including QDs can be fabricated through the manipulation of the energy band gaps in semiconductors. Namely, by fabricating a structure in which a material with a larger energy gap surrounds a material with a smaller energy gap, electrons and holes are confined in the material with the smaller energy gap. However, to obtain sufficient quantum effects, these structures must be on the order of several tens of nanometers or smaller. We require extremely precise techniques with which to fabricate arbitrary nanostructures. In recent years, advanced semiconductor crystal growth technology has led to a mature technology for fabricating semiconductor films at a precision of 1 nm or less, based on molecular beam epitaxy (MBE) and other techniques. We can thus now easily fabricate QWs, the one-dimensional carrier confinement structure. These QWs can be fabricated by precisely controlling growth rate and growth time in semiconductor crystal growth. This technique has led to the realization of semiconductor lasers that operate at room temperature, and these lasers have been applied to various devices, including those aimed at many applications other than optical communications.

In contrast to the thin-film deposition technology for fabricating QWs, QDs are more difficult to fabricate. As QDs require a confinement structure in all three dimensions, we now require a structure fabrication technology on the order of several tens of nanometers or less also in directions parallel to the surface. The first attempt at fabricating QDs involved the formation of a QW, patterning of the well with electron-beam lithography equipment, and etching of the pattern. However, this topdown method presents problems in that it cannot produce QDs of high quality, as it damages the sample during etching; additionally, it cannot produce high-density QDs.

In the early 1990s, a new method of QD fabrication was invented that made use of the self-assembling nature of semiconductor crystal growth. In crystal growth in a lattice mismatched material system, structures are selfassembled at sizes on the order of several tens of nanometers in directions parallel to the surface. The QDs obtained in this manner are referred to as self-assembled QDs[3]. Normally, in crystal growth of a lattice mismatched material system, defects and dislocations are formed to relax the strain energy of growth film when this film cannot withstand the lattice strain. Crystal quality deteriorates, with defects and dislocations, and thus materials with low lattice mismatching are generally selected for semiconductor crystal growth. On the contrary, self-assembled QDs make positive use of this strain in a lattice mismatched system. One of the most popular material systems for self-assembled ODs consists of a combination of GaAs and InAs. In this combination, InAs forms the ODs. GaAs and InAs have lattice constants of 5.653 Å and 6.058 Å, respectively, and the lattice mismatch between them is approximately 7%. When InAs is grown on GaAs, InAs first grows two-dimensionally and moves to three-dimensional growth when the layer exceeds approximately 1.5 monolayers (one monolayer corresponds to half the lattice constant.). This sort of growth mode is referred to as a Stranski-Krastanow mode (S-K mode). This three-dimensional growth forms InAs island structures (InAs QDs) on the sample surface, each with a diameter of approximately several tens of nanometers. When InAs is grown without being stopped, defects and dislocations are formed in the crystal, as discussed earlier, and the crystal quality deteriorates. However, defects and dislocations do not arise directly following the formation of the InAs QDs, and if the growth of InAs is stopped at the appropriate growth amount and the sample is embedded with GaAs (or another material with a band gap larger than that of InAs), InAs QDs can be successfully produced. As an example, Fig. 2 shows an atomic force microscope (AFM) image of the self-assembled InAs QDs fabricated on GaAs with an MBE at NICT facilities. In this sample, QDs with an



average diameter of 40 nm, an average height of 9 nm, and a density of 2.5×10^{10} /cm² were obtained. It is characteristic of the self-assembling method that high-quality ODs were obtained without damage, as this is a complete vacuum process; additionally, the QDs obtained feature higher density than that seen in the top-down fabrication method. Using this type of QD, diverse applications are now under development. Among these applications, use in optical communication devices is just a step from practical use. In particular, the QD laser in the 1.3-mm band and the QD semiconductor optical amplifier operate at a lower threshold current, depend less on temperature, and offer better high-speed signalprocessing performance than lasers and optical amplifiers based on QWs.

In these QD devices, the QDs function as the gain medium; thus, fabrication technology for denser QDs stands as a key technology in improving device performance. However, even when simply considering increasing the in-plane density, spatial limitations arise. For example, assuming that we can fabricate QDs with a diameter of 20 nm in a closely packed structure as shown in Fig. 3, surface density is limited to approximately 3×10^{11} /cm². Further, it is not easy to fabricate such a structure. When one pursues a denser QD structure, one may stack layers consisting of ODs. However, QD fabrication based on the S-K mode uses the strain energy of the lattice mismatched material system as the driving force in QD formation. Thus, when the density of the QDs is to be increased by stacking, accumulation of strain is a problem. This accumulation of strain may lead to problems such as changes in the size and shape of the QDs; further, excessive accumulation of strain generates defects and dislocations, so that the number of stacked layers is generally limited to 10 or less.

3 Stacking of QD multi-layer by strain-compensation method

To solve this problem, we developed the strain-compensation method when stacking QDs; this approach thus enabled stacking of multi-layer QDs. Figure 4 is a schematic diagram of the strain-compensation method. We used an InP(311)B substrate for fabricating the QDs. As shown in Table 1, the lattice constant





of InP is between that of GaAs or AlAs and InAs, and as a result various material systems can be grown on an InP substrate. In the present study we have devised a structure in which the InAs QDs fabricated on InP are embedded in InGaAlAs, which has a slightly smaller lattice constant than InP. In this manner, the tensile strain generated in InAs is compensated for by the compressive strain in InGaAlAs, which solves the problem of accumulated strain energy when fabricating the stacked structure. We determined the conditions for strain compensation based on the following equations.

$$d_{QD} \cdot \epsilon_{QD} = -d_s \cdot \epsilon_s$$
$$\epsilon_{QD} = (a_{InAs} - a_{InP})/a_{InP}$$
$$\epsilon_s = (a_s - a_{InP})/a_{InP}$$

where dop and ds are the film thicknesses of the QD layer and of the strain-compensation layer, respectively. aInAs, aInP, and as are the lattice constants of InAs, InP, and the InGaAlAs strain-compensation layer, respectively. EQD and ε_s are the amounts of strain with respect to the InP substrate in the InAs QDs and in the strain-compensation layer, respectively. The sample is prepared as follows. The InP(311)B substrate is placed in an MBE growth chamber and thermally cleaned at 500°C for 10 minutes to produce a clean surface. Then, a 150 nmthick lattice-matched InAlAs buffer layer is grown. Finally, the InAs QDs and the InGaAlAs strain-compensation layer are grown alternately, to produce the stacked structure.

First, to verify the effect of strain compensation, the InGaAs QDs fabricated on a GaAs(311) substrate and the InAs QDs fabricated on the InP(311)B are compared using an AFM. Figure 5 shows the results. In the figure, (a) shows the single layer InGaAs QDs fabricated on the GaAs(311)B substrate. In this single layer, relatively uniform selfassembled QDs are formed. In (b) we see the results of AFM observation for a sample stacked with 20 layers of these InGaAs QDs with 10 nm GaAs spacer layers. In this case, strain compensation is not introduced, so that



(a) Single layer InGaAs QDs on GaAs, (b) 20layer stacked InGaAs QDs on GaAs, (c) single layer InAs QDs on InP, and (d) 20-layer stacked InAs QDs on InP.

the accumulation of strain leads to large size distribution in the QDs. Many regions without ODs are also observed. We can surmise that defects and dislocations have formed in these areas, preventing the formation of ODs. In any case, without strain compensation in the stacking, the QDs produced clearly show degradation. Continuing, (c) shows the surface morphology of the single-layer InAs QDs on the InP(311)B substrate, and (d) shows a sample stacked with 20 layers of InAs QDs with 10 nm InGaAlAs spacer layers, which satisfies the strain compensation conditions. Uniform QDs are also formed for the single layer InAs QDs on the InP substrate, and strain compensation maintains uniformity in the size distribution of the QDs after stacking 20 layers. These results clearly indicate that strain compensation plays an important role in stacking multi-layer QDs. As QD distribution is more uniform than that without strain compensation, we may further conclude that this method also suppresses the formation of defects and dislocations. It is also known that adding Al to the strain-compensation layers suppresses In surface segregation. It is thus important to use InGaAlAs containing Al in the intermediate layers in order to form a uniform stacked structure of QD layers [4].

To confirm that dislocations are not formed, we also observed a sample cross-section with a scanning transmission electron microscope (STEM). Figure 6 shows the results. In this sample, 30 layers are stacked with 20 nm of InGaAlAs strain-compensation layers. The inserted figure is the result of AFM measurement of the sample surface. Uniform QDs have formed on the sample surface, and cross-sectional STEM measurement does not indicate the formation of dislocations. Thus, it is clear that the creation of a stacked structure with strain compensation not only maintains the uniformity of the ODs but also suppresses the formation of dislocations, leading to the creation of high-quality QDs. The advantage of stacked structures based on the strain-compensation method lies in the fact that the number of stacked layers is, in principle, unlimited, as long as the strain compensation condition is satisfied. Thus, stacking can be repeated many times, making it possible to fabricate extremely dense QDs. The present study led to the fabrication of a stacked structure of up to 150 layers. Figure 7 shows the surface morphology of the InAs QDs stacked at 150 layers. Despite the extremely large number of stacked QD layers, surface morphology shows no degradation. Figure 8 shows the dependence of size and density of the QDs on the number of stacked layers. The diameter, height, and density of the QDs changes little as a function of the number of stacked layers, which indicates the effective-





Fig.7 AFM image of 150-layer stacked QDs

ness of the strain-compensation method in fabricating a stacked structure. The cross-sectional STEM measurement discussed above also supports the maintenance of QD size distribution. Figure 8 shows that approximately 4×10^{10} /cm² of QDs are formed per layer. Thus, over 5×10^{12} /cm² of QDs are formed in a sample with 150 stacked layers, a result that would prove impossible using ordinary methods. This number of stacked QD layers is the largest in the world to date. The inserted figure in Fig. 7 shows a two-dimensional fast Fourier transformation (2DFFT) image. This 2DFFT image clearly indicates higher-order satellite peaks. As is also clear from the AFM image, this indicates a two-dimensional array structure of the QDs. The symmetry of the 2DFFT shows that the QDs are formed in a closely packed structure with six-fold symmetry. The QD array formation as the number of



stacked layers increase is probably due to the redistribution of strain after the formation of each strain-compensation layer. In other words, although the strain energy is counterbalanced and prevented from accumulating in the material system as a whole, a non-uniform strain is distributed on the sample surface after the growth of a strain-compensation layer, due to variances in the positions of the QDs embedded. The lattice constant is slightly larger directly above the QDs, so these serve as the dominant sites for the generation of the QDs in the next layer. This phenomenon can also be confirmed in cross-sectional STEM measurement, which shows that the QDs in the next layer are formed above the QDs of the lower layer. As the strain compensation condition is satisfied by necessity, the lattice constant is slightly smaller at positions other

than those directly above the ODs, compensating for the tensile and compressive strains in the entire system. If we look at a single QD, we see that the OD in the next layer is formed above the QD of the lower layer, continuing the morphology of the first layer. However, the fact that an array formation is promoted as the number of stacking layers increases indicates that the strain distribution formed on the spacer layer is due to the strain interactions involving the QDs in the nearby area as well as the QD directly below. Thus, the formation of a strain field involving multiple dots is important in OD array formation. It is easily deduced that the propagation of the strain field strongly depends on the thickness of the intermediate layer. In other words, thinner intermediate layers will lead to a stronger effect of the QD directly below relative to the surrounding QDs, so the system will continue the morphology of the first layer as is. Thicker intermediate layers involve an extremely large number of ODs in the generation of the strain field, so the strain field is averaged and the QD array formation may disappear. It should also be noted that the substrate (311)B surface tends to cause the formation of QD arrays [5]. This is also considered to lend additional strength to the formation of a QD array structure. These phenomena have been well studied with respect to the PbSe/PbEuTe material system [6]. To investigate this aspect, the present study also involved the fabrication of samples stacked with 150 layers of 10-nm and 60-nm strain-compensation layers. Figure 9 shows the results of AFM observation. In the figure, (a) shows the results with the 10-nm straincompensation layers and (b) those with the 60nm strain-compensation layers. As expected, the array structure eventually dissipates with both thick and thin strain-compensation layers. Thus, it is clear that film-thickness control of the strain-compensation layer is an important element of QD array formation. As discussed at the outset, QDs have discrete energy levels with a delta-function-form density of states. As the energy can be manipulated, the QDs are sometimes likened to artificial atoms.



If we can fabricate a three-dimensional array structure of QDs based on the stacked QDs, we can therefore obtain an array structure of artificial atoms—in other words, artificial crystals. Here, we consider that the following three factors will be of particular importance: stacking based on strain compensation, QD array formation based on thickness control of the intermediate layers, and control of coupled states between the QD layers.

Finally, we will discuss the optical properties of the stacked QDs. Strain-compensation layers of 20 nm are used and the photoluminescence (PL) of the sample stacked at 150 layers is measured at room temperature. To excite the sample, the 532-nm second harmonic of a diode-pumped Nd:YVO4 laser is used. A 250-mm monochromator and an electrically cooled PbS photodetector are used for spectroscopy and detection of the emitted light. Figure 10 shows the results of measurement. The figure shows strong emission even at room temperature. These results also demonstrate the effectiveness of suppressing dislocation by strain compensation and of enhancing emission intensity using increased density. This PL measurement shows a spectrum with a main peak near approximately 1.5 µm and a shoulder structure on the higher energy side. Based on a simple calculation of



quantum levels, these peaks are found to agree with the energy levels of the ground state, the first excited state, and the second excited state of the QDs. The corresponding values are indicated in the figure. The full width at half maximum of the ground state is approximately 40 meV. The emission wavelength of this sample corresponds to that of fiber-optic communication and can be expected to be applied to QD lasers and semiconductor optical amplifiers. The density of the QDs is particularly important in QD lasers and semiconductor optical amplifiers, as this is the source of gain. Thus, the fabrication technology for high-density QDs used in the present study has the potential to improve the performance of conventional semiconductor devices to a significant degree. Further, InAs QDs on GaAs are subject to large compressive stress caused by GaAs and lead to formation of a large band gap, causing a problematic restriction of emission to the 1-µm band. However, as the InP substrate has a larger lattice constant than GaAs, the compressive stress applied to InAs on an InP substrate is smaller; accordingly, we have obtained emissions of the InAs QDs in the fiber-optic communication wavelength. These emission characteristics can be modified by changing the volume of the InAs QDs and the barrier height of the intermediate layers. If we combine QDs with various emission wavelengths and increase the density while satisfying the strain compensation condition, we believe that it will be possible to produce a semiconductor optical amplifier with higher efficiency and broader bandwidth than those available with current products.

In addition, studies are progressing to apply the ultra-high-density QDs fabricated in the present study to areas other than semiconductor lasers and optical amplifiers, such as application as a saturable absorber (SA) and experiments for measuring the dephasing time of the excitons in QDs[7][8]. In these activities, we have obtained superior saturable absorption characteristics and longer exciton dephasing time than those obtained in other reports. These results are expected to be applied to the implementation of high-performance mode lockers for mode lock lasers and quantum information processing. For further details on these results, please see the references.

4 Conclusions

We have developed a strain-compensation method for increasing the density of selfassembled QDs and have enabled the stacking of QDs based on this method. We have shown that the size and distribution of the QDs are kept uniform even with multi-layer stacking in samples using strain-compensation layers. Based on this method, we have achieved a world record of 150 stacked layers with respect to self-assembled QDs, and have successfully developed ultra-high-density ODs with a density of 5×10^{12} /cm² or larger (two orders of magnitude larger than with the conventional method). By stacking ODs using this method, we have also confirmed the emergence of an array structure for the QDs, which is considered to be due to modulation in the distribution of strain. This array structure of QDs may also be applied to the fabrication of artificial crystals. We also obtained emissions in the 1.5-mm band at room temperature with the fabricated ultra-high-density QDs. This wavelength corresponds to the fiber-optic communication wavelength, and this technology is considered to offer promising potential use in improving the performance of QD lasers and semiconductor optical amplifiers, which require high-density QDs.

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AKAHANE Kouichi, Ph.D Researcher, Advanced Communications Technology Group, New Generation Network Research Center (former : Researcher, Photonic Information Technology Group, Basic and Advanced Research Department)

Semiconductor Photonic Device, Crystal Growth of Semiconductor



YAMAMOTO Naokatsu, Dr. Eng.

Researcher, Advanced Communications Technology Group, New Generation Network Research Center (former: Researcher, Photonic Information Technology Group, Basic and Advanced Research Department)

Optoelectronics Materials and Novel Devices



GOZU Shinichiro, Ph.D.

Limited Term Reseacher, Advanced Communications Technology Group, New Generation Network Research Center (former: Researcher, Photonic Information Technology Group, Basic and Advanced Research Department) Semiconductor Optical Device, Crystal Growth.

Nano Fabrication Process

OHTANI Naoki, Ph.D.

Associate Professor, Department of Electronics, Doshisha University

Semiconductor Photonic Device, Condensed Matter Photophysics



UETA Akio, Ph.D.

Limited Term Reseacher, Advanced Communications Technology Group, New Generation Network Research Center (former: Researcher, Photonic Information Technology Group, Basic and Advanced Research Department)

Semiconductor Crystal Growth, Photonic Device



TSUCHIYA Masahiro, Dr. Eng.

Group leader, Advanced Communications Technology Group, New Generation Network Research Center (former: Group leader, Photonic Information Technology Group, Basic and Advanced Research Department)

Photonics, Electronics