Long-wavelength emission in structures with quantum dots formed in the stimulated decomposition of a solid solution at strained islands

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When an array of strained InAs nanoislands formed on a GaAs surface is overgrown by a thin (1–10 nm) layer of an indium-containing solid solution, stimulated decomposition of the solid solution is observed. This process causes the formation of zones of elevated indium concentration in the vicinity of the nanoislands. The volume of newly formed InAs quantum dots increases as a result of this phenomenon, producing a substantial long-wavelength shift of the photoluminescence line. This effect is enhanced by lowering the substrate temperature, and it depends weakly on the average width of the band gap of the solid solution. The indicated approach has been used successfully in achieving room-temperature emission at a wavelength of 1.3 μm. © *1999 American Institute of Physics.* [S1063-7826(99)02208-5]

INTRODUCTION

The self-organizing phenomena responsible for the creation of semiconductor heterostructures with quantum dots (QDs) and quantum wires[1] are intriguing not only from the standpoint of revealing fundamental aspects of epitaxial growth, but also in regard to the fabrication of new-generation optoelectronic and microelectronic devices. The greatest progress in device applications is achieved utilizing the phenomenon of spontaneous conversion of a strained thin film to an array of coherent islands.[2–4] Injection lasers have been constructed utilizing QDs and exhibiting extremely high temperature stability of the threshold current density J_{th}, a low value of J_{th} (Refs. 4–6), and continuous-wave lasing at room temperature with a power output in excess of 3 W (Ref. 7).

One advantage of using QD structures is the possibility of broadening the optical range of emission in comparison with structures based on quantum wells. For example, structures containing In(Ga)As/(Al)GaAs QDs are capable of lasing in the range $1-1.6$ μm (Refs. 4 and 8–10). In particular, QD structures hold considerable promise for the fabrication of devices emitting at a wavelength of 1.3 μm (Refs. 9 and 10). Lasers utilizing InGaAs QDs and emitting at a wavelength of 1.3 μm have now been constructed.[10] Nonetheless, because of rapid gain saturation, lasing via the ground state of QDs has been achieved only by means of very long cavities and with the deposition of high-reflectivity coatings. The problems of attaining the 1.3-μm range for structures utilizing InGaAs/GaAs QDs stem from the rather large average thickness of InAs in the active zone and the high probability of formation of misfit dislocations. It is therefore critical at this time to look for new approaches to the construction of QD devices emitting at 1.3 μm, with a need to minimize the quantity of In in the active zone. Huffaker *et al.*[10] have deposited 5.5 InAs monolayers to achieve emission at a wavelength of 1.3 μm. In the present study we have reduced this number to four monolayers.

We have investigated the structural and optical properties of QD arrays overgrown by a layer of an (In, Ga, Al)As solid solution. The resulting QDs stimulate decomposition of the solid solution, thereby increasing the In concentration in the vicinity of the QDs and producing a long-wavelength shift of the emission line. This approach can be used to achieve emission from InAs QDs at wavelengths up to 1.32 μm while significantly reducing the total content of In in the active zone.

EXPERIMENT

The investigated structures were grown by molecular-beam epitaxy on GasAs (100) semi-insulating substrates by means of a Riber 32P MBE machine. The active zone was placed in a GaAs host and consisted of an array of quantum dots formed in the deposition of an InAs thin film. The effective thickness of the InAs QD layer d_{QD} varied from two to three monolayers (ML) for different structures. The QDs were overgrown by an InGaAs or InGaAlAs solid solution. The thickness of the (In, Ga, Al)As layer (L) varied from 2.5 nm to 8 nm, and the In concentration in the solid solution (x)

varied from 0.1 to 0.4. To prevent the transport of nonequilibrium carriers into the surface zone and into the substrate zone during optical investigations, the active zone was bounded on both sides by short-period AlAs/GaAs superlattices. The growth temperature was 485 °C for the active zone and 600 °C for the rest of the structure. The formation of QDs was monitored from the fast-electron diffraction pattern in reflection.

Transmission electron microscope (TEM) examinations were made on a Philips EM 420 electron microscope with an accelerating voltage of 100 kV. Photoluminescence was excited by an Ar^+ laser ($\lambda = 514.5$ nm, excitation density $\sim 100\,\mathrm{mV/cm^2}$). In the measurement of the luminescence excitation spectrum light from a halogen lamp was transmitted through a monochromator. The luminescence was detected by a cooled Ge photodiode.

RESULTS AND DISCUSSION

Today the greatest progress in the fabrication of (In, Ga)As/GaAs QD lasers is achieved using QDs in the initial stage of their formation,[6,7] which are characterized by a high density, an absence of dislocations, and short carrier trapping and relaxation times. For the implementation of long-wavelength emission in the InAs QD structures the QD arrays were overgrown with a layer of $In_xGa_{1-x}As$ solid solution during the initial stage of their formation ($d_{QD} \sim 2$ ML).

Figure 1a shows the photoluminescence spectra of InAs QD structures prepared by the deposition of ~ 2 ML of InAs and overgrown by a layer of $In_xGa_{1-x}As$ solid solution with various In concentrations (x). The thickness of the $In_xGa_{1-x}As$ layer for all the structures was 4 nm. The photoluminescence spectrum contains a line, labeled *QW* in Fig. 1, in addition to the *QD* line associated with carrier recombination in the resulting QDs. To investigate the nature of this line, we have analyzed the luminescence excitation spectra (Fig. 1b). In the excitation spectra of the *QD* line there are discernible peaks $P1$, $P2$, and $P3$, which on the energy scale are situated at distances from the detection energy of 35 meV, 74 meV, and 102 meV, respectively. This form of the luminescence excitation spectra is typical of QD structures[11] and is associated with a multiphonon mechanism of carrier relaxation from excited QD states. The luminescence excitation spectra also reveal a *QW* line whose position coincides with the position of the *QW* line in the photoluminescence spectra and does not depend on the detection energy. This result shows that the *QW* line is associated with carrier excitation in the continuum of the InGaAs solid solution.

It is evident from the photoluminescence spectra (Fig. 1) that when the In concentration in the InGaAs solid solution increases, the *QW* line exhibits a long-wavelength shift and, accordingly, is shifted toward the long-wavelength end relative to the wetting-layer line observed in the spectrum of ordinary InAs/GaAs QDs (*WL* line in Fig. 1a). Consequently, the QDs reside in the InGaAs matrix, which is a narrower-gap medium than GaAs.

Note that despite the overgrowth of the QDs by the narrower-gap (in comparison with GaAs) material, the energy distance between the QD photoluminescence maximum (*QD* line) and the *QW* peak is roughly 200 meV, which is comparable with the values obtained for ordinary QDs in a GaAs host.[4] It also shows that the carriers localize effectively in the QDs.

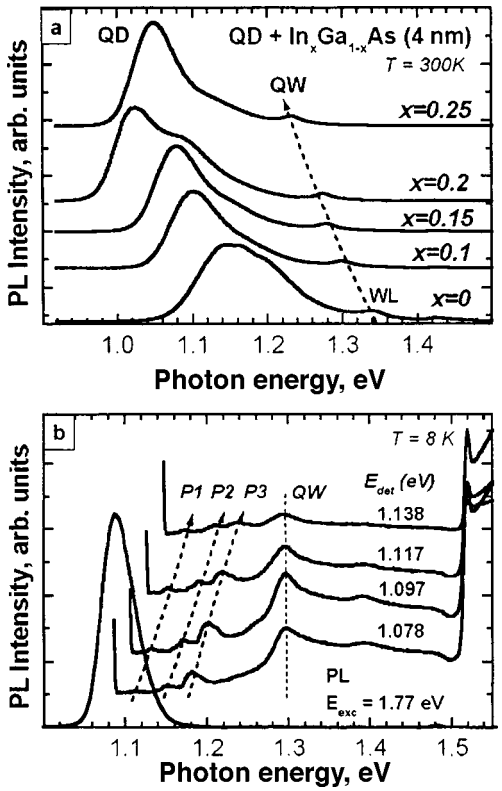

FIG. 1. (a) Photoluminescence spectra for structures containing InAs QDs ($d_{QD} = 2$ ML) overgrown by an $In_xGa_{1-x}As$ layer, $L = 4$ nm. (b) Photoluminescence spectrum and luminescence excitation spectra for structures containing InAs QDs ($d_{QD} = 2$ ML) overgrown by an InGaAs layer. The excitation and detection energies are indicated alongside the spectra.

It is evident from the photoluminescence spectra (Fig. 1a) that as x increases in the interval from 0 to 0.2, the photoluminescence *QD* line is observed to shift toward lower photon energies. However, when x is further increased to 0.25, the *QD* line shifts toward the short-wavelength end. On the other hand, when the thickness of the InGaAs layer is increased from 4 nm to 8 nm at a constant concentration x (0.15 or 0.2), we also observe a short-wavelength shift of the *QD* peak. The variation of the energy positions of the *QD* and *QW* lines in the photoluminescence spectra as functions of the parameters of the $In_xGa_{1-x}As$ layer is shown in Fig. 2a. A further increase in the In concentration to 0.35–0.4 at $L = 4$ nm still enabled us to achieve emission at a wavelength of 1.3 μm, but the TEM examinations show that the density of penetrating dislocations increases sharply in this case.

Figure 2b shows the dependence of the total photoluminescence intensity on the effective InAs thickness in the active zone ($d_{eff} = d_{QD} + L \times x$), which for the given samples increases as a result of the increase of the In concentration in the solid solution. It is evident that for concentrations $x > 0.3$ (which corresponds to $d_{eff} = 18$ Å for $d_{QD} = 2$ ML and $L = 4$ nm), the photoluminescence intensity decreases considerably.

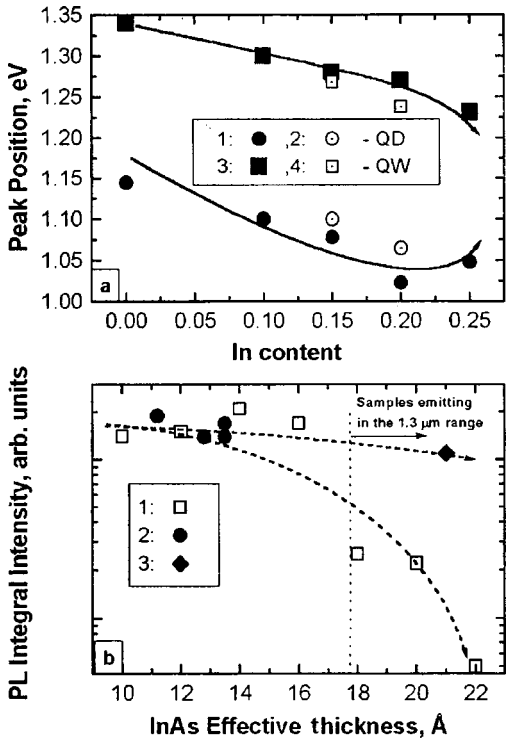

FIG. 2. (a) Positions of the QD photoluminescence peaks (*QD* line) and an InGaAs layer (*QW* line) versus the In concentration in the InGaAs layer: (*1, 3*) $L=4$ nm; (*2, 4*) $L=8$ nm. (b) Dependence of the total photoluminescence intensity on the effective thickness of the InAs layer: (*1*) Structures with $d_{QD}=2$ ML, $L=4$ nm, and various In concentrations in the solid solution; (*2*) structures with various effective InAs thicknesses in the active zone (the parameters of the structures are given in Table I; (*3*) structures with InAs QDs ($d_{QD}=3$ ML) deposited in the middle of an $In_{0.12}Ga_{0.88}As$ layer.

It is evident from the investigations that for structures with 2 ML of InAs QDs with overgrowth of the QDs by an InGaAs solid solution it is possible to obtain long-wavelength emission, but for a certain critical In concentration in the InGaAs layer ($x\sim 0.25$ in the given situation) the resulting effect is such as to produce a short-wavelength shift of the QD photoluminescence line. To obtain long-wavelength emission in this case requires a significant increase of the In concentration in the InGaAs layer, but this leads to the formation of dislocations and, in the final analysis, degradation of the optical properties of the structures (Fig. 2b).

An alternative possibility for achieving emission at a wavelength of 1.3 μm is to increase the effective thickness of the InAs layer (d_{eff}) without increasing the In concentration in the solid solution. We have grown a series of samples in which the effective thickness of the InAs layer was varied by varying the amount of InAs during deposition of the QDs (d_{QD}) and by varying the thickness of the InGaAs layer (L) without the value of x ever exceeding 21 %. Table I gives the parameters of the active zone of these structures, along with TEM data [the density and diameters of the QDs and the density of misfit dislocations formed at the interface in the plane of the QDs]. Figure 3 shows the photoluminescence spectra of these samples. The following conclusions can be drawn from a comparison of the TEM and photoluminescence results. First, increasing the total quantity of In during the successive deposition of QDs and the InGaAs layer increases the size of the islands and produces a long-wavelength shift of the QD photoluminescence line. However, at a certain critical concentration of In atoms ($d_{QD}=2.7$ ML, $L=25$ Å, and $x=21$ % in the given situation) the density of misfit dislocations increases, accompanied by a 25-meV shift of the QD photoluminescence line toward higher photon energies. This effect is similar to the effect observed for 2 ML of QDs.

TABLE I.

Sample	d_{QD} (ML)	x	L, nm	Lateral QD diameter, nm	QD density, cm^{-2}	Density of misfit dislocations, cm^{-1}
A	~2.2	0.18	2.6	15	3.4×10^{10}	2.3×10^{4}
B	~2.7	0.18	2.6	18	4.3×10^{10}	2.7×10^{4}
C	~2.7	0.18	3	18	4.5×10^{10}	3.2×10^{4}
D	~2.7	0.21	2.6	17	4.6×10^{10}	6.5×10^{4}

We assume that when the QD array is overgrown by an InGaAs layer, the In atoms are distributed nonuniformly in the growth plane, owing to QD-induced stresses. As a result, zones of elevated In concentration form in the vicinity of the resulting QDs, in effect increasing the size of the QDs. The phenomenon of decay of a quantum well under the influence of a QD array is described in Refs. 12 and 13 for various materials. If the In content exceeds a certain value, the In concentration near the largest dots can exceed the critical value, and misfit dislocations are formed in the plane of the QDs. Since radiative recombination does not take place at these dots, the intensity of the long-wavelength part of the photoluminescence spectrum decreases, and the photoluminescence line shifts toward the short-wavelength end. Another cause of the short-wavelength shift of the QD photoluminescence line with a simultaneous long-wavelength shift of the photoluminescence line of the InGaAs layer could be a change in the growth kinetics as the islands are overgrown by an InGaAs solid solution having a higher In content. In this event the fraction of In atoms deposited in the vicinity of the QDs decreases, raising the average In content.

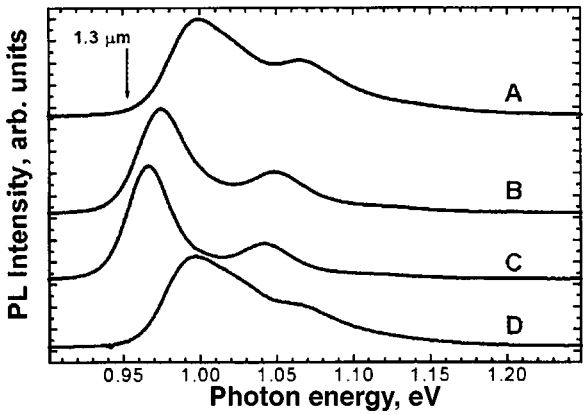

FIG. 3. Photoluminescence spectra for structures with various effective InAs thicknesses in the active zone. The parameters of the structures $A-D$ are given in Table I.

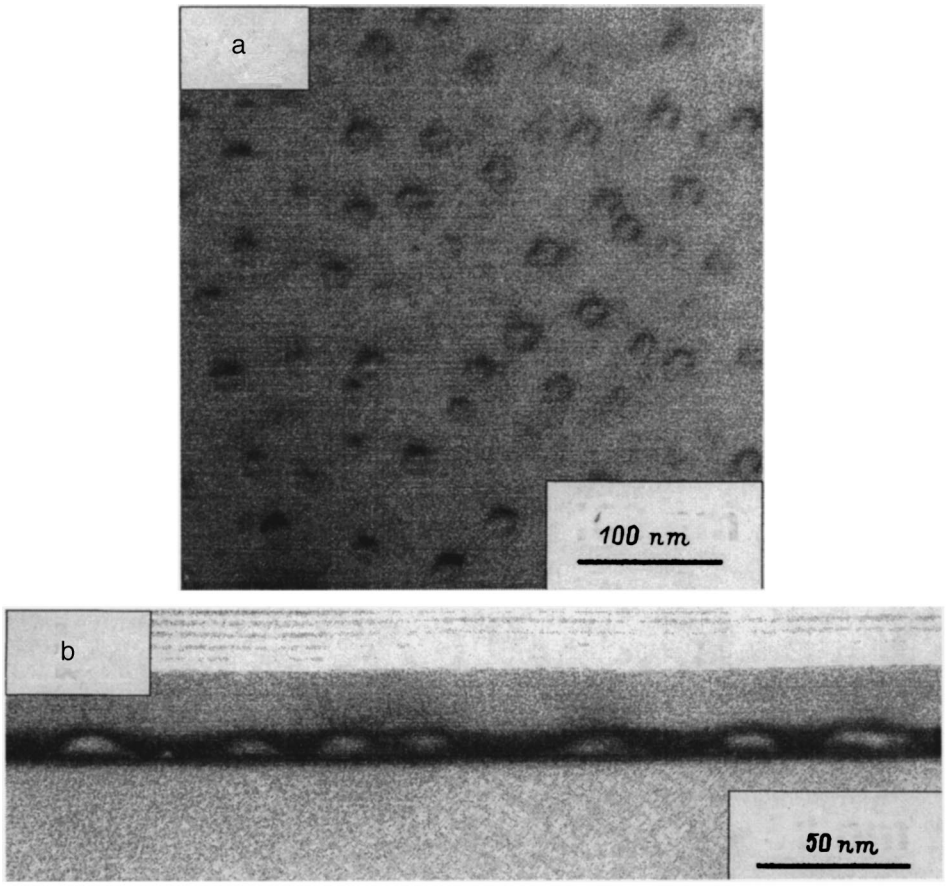

FIG. 4. TEM images in the growth plane (a) and in the cross section (b) of a structure with InAs QDs ($d_{QD}=3$ ML) deposited in the middle of an $In_{0.12}Ga_{0.88}As$ layer.

Since the increase in the QD diameter as the InAs content in the QDs increases from 2.2 Ml to 2.7 ML does not cause the density of misfit dislocations to increase, we have sought to further increase the long-wavelength shift by growing a structure with 3 ML of InAs QDs deposited in the middle of a wide (10 nm) $In_{0.12}Ga_{0.88}As$ layer. Figure 4 shows TEM images in the growth plane and in the cross section for the indicated structure. It is evident from the image of the cross section that the height of the resulting islands is comparable with the thickness of the InGaAs layer, indicating essentially complete overgrowth of the QDs by the InGaAs layer. The newly formed QDs therefore reside in the narrow-gap InGaAs host. Moreover, a comparison of the TEM images in the growth plane for structures containing 3 ML of InAs with and without[4] a layer of InGaAs solid solution shows that the deposition of InGaAs increases the lateral dimensions of the QDs from ~15 nm to ~20 nm.

The indicated structure exhibits emission at a wavelength of 1.3 μm at room temperature (Fig. 5). We also note that when the effective thickness of the InAs layer increases as the result of an increase in d_{QD} and L and a simultaneous lowering of the In concentration in the solid solution, the total photoluminescence intensity is essentially unchanged (Fig. 2b). This approach can therefore be used to achieve emission at 1.3 μm and at the same time to avoid degradation of the optical properties of the structures.

We subsequently attempted to lower the total quantity of In in the active zone while maintaining long-wavelength emission at a wavelength of approximately 1.3 μm. To do so, we investigated the influence of the growth temperature during overgrowth of the QDs by an InGasAs layer on the optical properties of the structures. Figure 6 shows the photoluminescence spectra of samples with the InGaAs layer grown at temperatures of 485 °C and 455 °C, respectively. It is evident that lowering the growth temperature imparts a long-wavelength shift to the emission. This result can be attributed to the influence of temperature on the decay kinetics of the InGaAs layer and demonstrates the feasibility of achieving a further long-wavelength shift of the photoluminescence line with the right choice of temperature regimes.

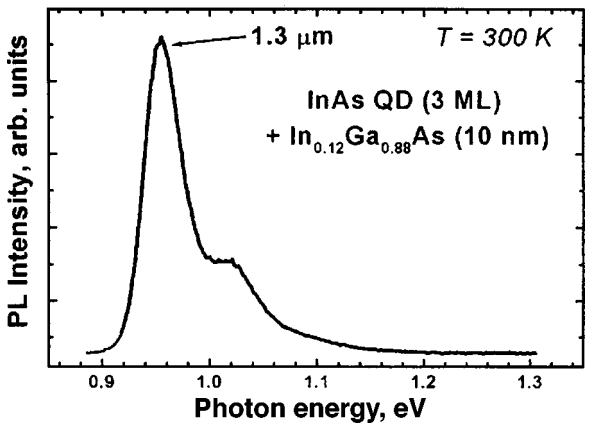

FIG. 5. Photoluminescence spectrum for a structure with InAs QDs ($d_{QD}=3$ ML) deposited in the middle of an $In_{0.12}Ga_{0.88}As$ layer.

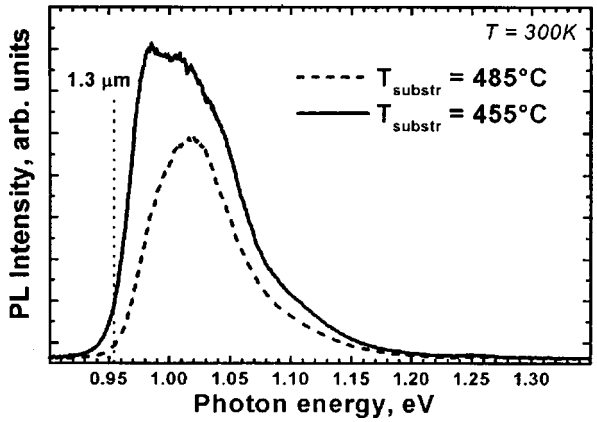

FIG. 6. Photoluminescence spectra for InAs QDs ($d_{QD}=2.2$ ML) overgrown by an $In_{0.12}Ga_{0.88}As$ layer at various temperatures. The overgrowth temperatures are indicated alongside the spectra.

In addition, we proposed to exploit a phenomenon that we had observed previously for spontaneously decomposing layers[1,14]: decomposition of an InGaAlAs/GaAs layer. We grew structures with InAs QDs overgrown by an InGaAlAs solid solution. The investigations show that the introduction of Al into the solid solution produces a significant long-wavelength shift of the QD emission line despite major broadening of the band gap of the host. This effect can be explained on the assumption that the introduction of Al significantly enhances the decomposition of the solid solution and increases the In concentration in the vicinity of the QDs. It should be noted that the total quantity of In in the active zone for this structure corresponds to 12.5 Å of InAs, which is far below the value of 16.5 Å given in Ref. 10, in which lasing at a wavelength of 1.3 μm is reported. We have thus succeeded in substantially lowering the total quantity of In in the active zone while achieving emission at a wavelength greater than 1.3 μm at room temperature.

CONCLUSIONS

The reported investigations have shown that the overgrowth of an array of InAs quantum dots by a thin film of (In, Ga, Al)As solid solution leads to decomposition of the solid solution and the formation of local zones near the QDs with an elevated concentration of In atoms. This phenomenon produces an effective increase in the size of the QDs and a significant long-wavelength shift of the photoluminescence emission line to 1.3 μm at room temperature. We have investigated the dependence of the optical properties of such structures on the growth conditions and have demonstrated the feasibility of achieving a long-wavelength shift of the emission without degradation of the optical properties of the structures. We have shown that the addition of small concentrations of Al to the solid solution can significantly reduce the total quantity of InAs in the active zone while maintaining an emission wavelength greater than 1.3 μm. This result demonstrates the possibility of constructing lasers utilizing InAs QDs on GaAs substrates and emitting at a wavelength of 1.3 μm.

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