Improvement of the uniformity of self-assembled InAs quantum dots grown on InGaAs/GaAs by low-pressure metalorganic chemical vapor deposition

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We report an approach to improve the uniformity of self-assembled InAs quantum dots (QDs) grown on a strained In0.12Ga0.88As buffer layer on GaAs substrates by low-pressure metalorganic chemical vapor deposition. By inserting a thin GaAs layer between the InAs QD layer and the In0.12Ga0.88As buffer layer and examining its thickness effect, we demonstrate that the photoluminescence (PL) inhomogeneous linewidth from the QDs can be improved by increasing the thickness of the thin GaAs layer. The PL inhomogeneous linewidth is significantly decreased from about 70 to 20 meV at 7 K as the thickness is increased from 0 to 2 nm. This significant improvement of the PL inhomogeneous linewidth is due to the fact that the QDs change from a bimodal distribution to a monomodal distribution consisting only of large QDs as a result of the inserted thin GaAs layer. © 2004 American Institute of Physics. [DOI: 10.1063/1.1802376]

The demand for high-performance semiconductor lasers operating at long emission wavelengths such as 1.3 or 1.55 µm has stimulated considerable interest in self-assembled InAs/GaAs quantum dots (QDs), because this system has the possibility of extending the emission of GaAs-based optical devices to these wavelengths.1–4 Due to the three-dimensional confinement of carriers, QDs are predicted to improve the performance of semiconductor lasers compared to quantum wells,5 and GaAs-based InAs QD lasers emitting at 1.3 µm with low thresholds have already been demonstrated.6 Nevertheless, the performance of the QD lasers must be further improved to put them into practical use, and the fabrication of QDs with high density as well as high uniformity is of critical importance for achieving the further performance improvement. Much effort7–13 has been made to develop such QDs with high density and uniformity. One way to increase the density of QDs is to grow InAs QDs on a strained InGaAs buffer instead of a GaAs buffer.11–13 As InGaAs matches InAs more than GaAs, it lowers surface energy or reduces indium migration length during the deposition of InAs QDs, the density of QDs is increased. However, the QDs usually exhibit a large photoluminescence (PL) inhomogeneous linewidth (typically greater than 50 meV1–13) that can greatly degrade the zero-dimensional nature of the density of states, resulting in higher threshold currents and lower temperature stability for QD lasers.14 Therefore, it is necessary to decrease the large PL inhomogeneous linewidth. In addition, the origin of this large PL inhomogeneous linewidth remains unclear.

In this letter, we present a method to improve the uniformity of self-assembled InAs QDs grown on a strained In0.12Ga0.88As buffer layer on GaAs substrates by low-pressure metalorganic chemical vapor deposition (MOCVD). By introducing a thin GaAs layer between the InAs QD layer and the In0.12Ga0.88As buffer layer and investigating its thickness effect, we demonstrate that the uniformity of the InAs QDs can be improved by increasing the thickness of the thin GaAs layer. The PL inhomogeneous broadening from the QDs is dramatically reduced from 70 to 20 meV at 7 K as the thickness is increased from 0 to 2 nm. A detailed analysis of the structural properties of the QDs measured by atomic force microscope (AFM) reveals that this significant improvement of the PL inhomogeneous linewidth is due to the fact that the QDs change from a bimodal distribution to a monomodal distribution consisting only of large QDs, as a result of the insertion of the thin GaAs layer.

All samples used in the study were grown on GaAs (001) substrates by low-pressure MOCVD, using trimethylindium (TMI), trimethylgallium (TMG), or triethylgallium (TEG), and tertiarybutylarsine (TBA) as source materials. Figure 1 schematically illustrates the cross-sectional structure of the samples. A 200-nm-thick GaAs buffer layer was first grown on the GaAs substrate at 700 °C, using TMG and TBA. Then, a 5-nm-thick In0.12Ga0.88As layer was grown at 500 °C, followed by a thin GaAs layer at 600 °C, using TMI or TGA and TBA. The thickness (t) of the thin GaAs layer for the samples was varied from 0 to 5 nm to examine its effect on the formation of subsequent InAs QDs. The subsequent InAs QDs were deposited at 500 °C using TMI and TBA. To achieve long wavelength emission at 1.3 µm, the QDs were finally capped with a 100-nm-thick GaAs at the same temperature as the QD growth, using TEG and TBA. The use of TEG in place of TMG as a gallium source during

![Figure 1](image-url)

FIG. 1. Schematic diagram of the cross-sectional structure of samples, where t varies from 0 to 5 nm for the samples.

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the deposition of a GaAs cap cannot only shift the emission to longer wavelengths but also narrow PL linewidths due to the much greater growth rate of GaAs attainable using TEG and TBA, compared to TMG and TBA.3,15 This much greater growth rate allows the rapid capping of the QDs and thereby sufficiently suppresses the size reduction and shape change of QDs as well as In/Ga intermixing that can occur during capping. The growth rate of the QDs was 0.011 monolayers (MLs)/s and the nominal thickness was 2.6 MLs for all the samples. Optical properties of the samples were evaluated by PL measurements. A He–Ne laser (633 nm) was used as a light source for smaller excitation powers, while a Ti:sapphire laser (840 nm) was employed for larger excitation powers. Structural properties of InAs QDs in the samples were characterized by measuring uncapped samples with an AFM.

Figure 2(a) shows PL spectra from the samples with different $t$, obtained at 7 K under an excitation density of about 1.5 W/cm$^2$. For $t$=0 nm (the QDs grown directly on the In$_{0.12}$Ga$_{0.88}$As buffer layer), it can be seen that the PL exhibits a broad and asymmetric peak with a linewidth as large as about 70 meV. Furthermore, two subpeaks in the broad and asymmetric peak can be clearly recognized although they are closely overlapped. As $t$ increases, the intensity of the peak at the side of short wavelength rapidly decreases, and almost vanishes when $t$ ≥ 2 nm. At the same time, the peak becomes more and more symmetric and the peak linewidth narrows. In Fig. 2(b), the full width at half maximum (FWHM) of the peaks in Fig. 2(a) is plotted as a function of $t$, where the FWHM dramatically decreases from about 70 to 20 meV as $t$ increases from 0 to 2 nm and remains almost unchanged with the further increase of $t$. Therefore, applying a thin GaAs layer can effectively improve the uniformity of the InAs QDs.

In order to clarify the PL behavior observed above, we investigated structural properties of uncapped QDs, corresponding to the samples used for PL studies, by AFM. Figure 3 presents surface AFM images and corresponding height histograms of the QDs, obtained in 1 μm×1 μm-areas. From both the surface AFM images and corresponding height histograms, a bimodal distribution consisting of small and large QDs is clearly visible for $t$=0 and 1 nm, while the small QDs are hardly observed for $t$=2 and 3 nm (or there are only the large QDs). Furthermore, the height distribution in the histograms can be well fitted by two Gaussians for $t$=0 and 1 nm, while for $t$=2 and 3 nm it can be well described only by one, as shown by the fits (solid curves) to the histograms. This further confirms that the QDs for $t$=0 and 1 nm are bimodal while for $t$=2 and 3 nm, the QDs are monomodal. In particular, the fitting to the height distribution for $t$=2 and 3 nm yields an identical FWHM of about 1.6 nm, indicating that the QDs for $t$=2 and 3 nm have same size variation. The density of QDs from the surface AFM images is $3.1 \times 10^{10}$, $2.2 \times 10^{10}$, $1.8 \times 10^{10}$, and $1.5 \times 10^{10}$ cm$^{-2}$ for $t$=0, 1, 2, and 3 nm, respectively. Although the QD density decreases with increasing $t$, the small QDs can be seen, both from the surface AFM images and corresponding height histograms, to decrease dramatically. The AFM results can be understood by the fact that a thinner GaAs is stretched more by the underlying In$_{0.12}$Ga$_{0.88}$As.
buffer layer, while a thicker GaAs layer grades out and compensates more of the strain accumulated in the In$_{0.12}$Ga$_{0.88}$As buffer layer. As a result, the thinner GaAs layer (e.g., $t \approx 1$ nm) has a larger lattice size and matches InAs more closely, leading to denser InAs QDs like those grown directly on the In$_{0.12}$Ga$_{0.88}$As buffer layer ($t=0$ nm). On the other hand, the thicker GaAs layer (e.g., $t=2$ nm) has a smaller lattice size, resulting in more uniform and greater InAs QDs due to its relatively higher surface energy, but with a decreased density. Clearly, the AFM results obtained above can be responsible for the observed PL behavior. For $t=0$ nm, the broad and asymmetric PL peak can be attributed to QDs with a bimodal and broad size variation. Furthermore, the significant decrease in intensity at the shorter wavelength side as well as the narrowing of the peak with increasing $t$ can be explained by the fact that the number of QDs with the small size rapidly decreases and the large QDs become dominant. Finally, the constant FWHM with the further increase of $t$ in Fig. 2(b) is attributable to the QDs with an identical size variation.

We also studied room temperature (RT) spectra from the samples, as shown in Fig. 4. It can be seen that the RT spectra are very similar to those obtained at 7 K [Fig. 2(a)], except that emissions (around 1220 nm) from first excited states are always observable due to thermal excitation at RT. Furthermore, it is found that the FWHM value of the peaks (ground states) in Fig. 4 is almost the same as the corresponding one obtained at 7 K [Fig. 2(b)], but a reduction from 70 to 56 meV for $t=0$ nm. This reduction for $t=0$ nm is due to thermal transfer of carriers from smaller QDs to nearby larger QDs having lower ground-state energies in the QD ensemble, which can result in a decrease in FWHM. This effect is associated with large QD size variations. As an example, RT excitation-dependent spectra from the sample with $t=2$ nm are shown in Fig. 5, where the state-filling effect involving up to the second excited state of the QDs. Moreover, the peak wavelength is found to be around 1220 nm for the ground, first excited, and second excited states of the QDs.

In summary, we have demonstrated a growth technique to improve the uniformity of self-assembled InAs QDs grown on a strained In$_{0.12}$Ga$_{0.88}$As buffer layer on GaAs substrates by low-pressure MOCVD. By inserting a thin GaAs layer between the QD layer and In$_{0.12}$Ga$_{0.88}$As buffer layer, the uniformity of the QDs can be improved by increasing the thickness of the thin GaAs layer, and the PL inhomogeneous broadening from the QDs is decreased from 70 to 20 meV at 7 K when the thickness is increased to 2 nm. As demonstrated by AFM observations, this significant improvement of the PL inhomogeneous broadening arises from the fact that the QDs change from a bimodal distribution to a monomodal distribution of large QDs, as a result of the inserted thin GaAs layer. The results obtained here suggest that self-assembled InAs QDs with high uniformity and density on GaAs substrates may be fabricated by optimizing the structure of the strain-coupled GaAs/In$_{0.12}$Ga$_{0.88}$As buffer layer.

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FIG. 4. RT PL spectra from the samples under an excitation density of about 3.8 W/cm$^2$. The peaks centered at about 1042 nm are from the underlying In$_{0.12}$Ga$_{0.88}$As layer.

FIG. 5. Excitation dependence of RT spectra from the sample with $t$ = 2 nm; $P$ is about 15 W/cm$^2$. Arrows from right to left indicate the peak position of ground, first excited, and second excited states of the QDs.