Narrow photoluminescence linewidth (<17 meV) from highly uniform self-assembled InAs/GaAs quantum dots grown by low-pressure metalorganic chemical vapor deposition

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(Received 2 October 2003; accepted 25 February 2004)

We report highly uniform self-assembled InAs quantum dots (QDs) emitting at 1.3 μm, grown on GaAs substrates by low-pressure metalorganic chemical vapor deposition. By optimizing the InAs growth rate and capping the QDs with GaAs using triethylgallium as a gallium source, we have achieved a narrow photoluminescence (PL) inhomogeneous linewidth of 16.5 meV (at 7 K) from QDs with a density of $1.7 \times 10^{10} \text{ cm}^{-2}$. Furthermore, we show by temperature-dependent PL measurements that the QDs exhibit almost no dependence of linewidth on temperature due to their high uniformity. © 2004 American Institute of Physics. [DOI: 10.1063/1.1711163]

The demand for high-performance semiconductor lasers operating at long emission wavelengths such as 1.3 or 1.55 μm has strongly stimulated research on 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 However, a large photoluminescence (PL) linewidth (typically 40–60 meV),2 which results from size, shape, and composition variation in QDs, has been one of the main obstacles limiting them from practical applications. Much effort has been made to decrease the large PL linewidth, and it has been improved to typically 19–25 meV by using various special growth techniques such as a low InAs growth rate,7,8 a low arsenic pressure,9 an InGaAs strain-reducing capping layer,2 close vertical stacking,10,11 and punctuated island growth.12 Very recently, a narrow PL linewidth of 14 meV at 10 K was reported, but it was achieved from a complicated bilayer of InAs QDs.13 For practical device applications, and especially for multilayer stacking of QDs to increase QD density and uniformity and density is more desirable. In addition, there are very few reports on highly uniform InAs/GaAs QDs grown by metalorganic chemical vapor deposition (MOCVD).8 In terms of high growth rates in volume production requirements or application to optical integrated devices that require regrowth or selective area growth, it would be greatly beneficial to achieve highly uniform QDs grown by MOCVD. Finally, owing to the lack of highly uniform QDs, traditional self-assembled InAs/GaAs QDs typically exhibit an unusual decrease of PL linewidth with increasing temperature.14–16

In this letter, we report the achievement of a narrow PL

linewidth of 16.5 meV at 7 K from a single layer of highly uniform self-assembled InAs/GaAs QDs with a density of $1.7 \times 10^{10} \text{ cm}^{-2}$, grown by low-pressure MOCVD. Furthermore, we show by temperature-dependent PL measurements that the QDs exhibit almost no dependence of linewidth on temperature due to their high uniformity.

Samples with different growth rates of InAs QDs were grown on GaAs (001) substrates by low-pressure MOCVD at a chamber pressure of 76 Torr, using trimethylindium (TMI), trimethylgallium (TMG), triethylgallium (TEG), and tertiary-butylarsine (TBA) as source materials. After thermal etching at 800 °C, a 200-nm-thick GaAs buffer layer was deposited on the substrate at 700 °C using TMG and TBA. Then InAs QDs were grown at 500 °C using TMI and TBA. After a growth interruption of 20 s, InAs QDs were capped with a GaAs layer (100 nm) using TEG and TBA at the same temperature as the QD growth. It has been demonstrated that the use of TEG in place of TMG as a gallium source during the deposition of a GaAs capping layer can result in both a narrower PL linewidth and longer-wavelength emission due to the much greater growth rate of GaAs attainable using TEG and TBA, rather than TMG and TBA.8 For example, at 500 °C, the growth rate of GaAs using TEG and TBA is about eight times larger than for TMG and TBA. The V/III ratio during the growth of InAs QDs and GaAs (buffer and cap layers) was approximately 0.3 and 25, respectively. The growth rates of GaAs for the buffer and cap layers were about 0.53 and 1.12 nm/s, respectively. The nominal thickness of InAs QDs was 2.64 ML for all of the samples. Optimization of the growth conditions and capping layer properties, combined with small InAs growth rates, enables production of InAs/GaAs QDs with inhomogeneous linewidths typically less than 20 meV, as discussed below. Optical properties of the samples were evaluated by PL measurements. A He–Ne laser (633 nm) was used as a light source for small excitation powers, while a Ti: sapphire laser (800 nm) was employed for larger excitation powers. Structural properties of InAs QDs in the samples were characterized by measuring uncapped samples with an atomic force microscope (AFM).

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We first investigated low-temperature (7 K) PL spectra from the samples. Figure 1 plots the variation of full width at half maximum (FWHM) of PL peaks from the samples as a function of InAs growth rate, obtained under an excitation density of about 0.5 W/cm² at 7 K. It can be easily seen from the figure that the FWHM first decreases and then increases with decreasing growth rate, reaching a minimum for a growth rate of 0.011 ML/s. The narrowing of the linewidth as the growth rate decreases from 0.022 to 0.011 ML/s can be ascribed to the formation of more uniform InAs QDs, which we have confirmed through AFM measurements of uncapped InAs QDs having the same growth conditions as the samples studied here. These AFM measurements show that the size variation of the QDs decreases as the growth rate decreases from 0.022 to 0.011 ML/s. In contrast, the broadening of the linewidth as the growth rate decreases further to 0.0055 ML/s is attributable to the increased importance of desorption over adsorption at such slow growth rates, which can degrade the uniformity of the QDs. Figure 2 shows the FWHM dependence of QDs with the growth rate of 0.011 ML/s on excitation density, in which a linear variation of the FWHM with excitation density is distinctly visible. The narrowing of the FWHM as the excitation density decreases for extremely low excitation densities may be due to the suppression of dephasing mechanisms since excited states of the QDs do not appear under the extremely low excitation densities. Furthermore, from Fig. 2, a FWHM as narrow as about 16.5 meV is obtained under the extremely low excitation density of ~0.05 mW/cm². The inset in Fig. 2 shows the PL spectrum of the QDs with the narrowest FWHM of 16.5 meV. Figure 3 shows an AFM image of the highly uniform uncapped QDs, measured in a 1 µm × 1 µm area. From this image, we estimate the density of the QDs to be 1.7 × 10¹⁰ cm⁻², and measure an average height of approximately 8.4 nm, and an average diameter of about 34.7 nm.

Next, we studied room temperature (RT) spectra of the highly uniform QDs. The inset in Fig. 4 shows a RT spectrum obtained with an excitation density of about 0.5 W/cm², showing a sharp peak similar to that observed at 7 K. The FWHM is now about 18.0 meV, and the peak energy is approximately 0.94 eV (about 1.32 µm). The achievement of the emission beyond 1.3 µm can be attributed to the rapid capping of the QDs, due to the greater GaAs growth rate.

**FIG. 1.** Variation of PL FWHM as a function of InAs growth rate, obtained under an excitation density of about 0.5 W/cm² at 7 K.

**FIG. 2.** FWHM dependence of QDs with the growth rate of 0.011 ML/s on excitation density. A linear variation of the FWHM with excitation density is distinctly visible. The inset is a 7 K PL spectrum of the QDs, showing the narrowest FWHM of 16.5 meV, obtained with an excitation density of approximately 0.05 mW/cm².

**FIG. 3.** AFM image (1 µm × 1 µm) of the highly uniform QDs. The density of the QDs is 1.7 × 10¹⁰ cm⁻², and the QDs have an average height of approximately 8.4 nm and base diameter of about 34.7 nm.

**FIG. 4.** RT PL spectrum from the highly uniform QDs, for an excitation density of about 50 W/cm². The energy spacing between the first excited and ground states and between the first and second excited states is approximately 70.7 and 67.8 meV, respectively. The inset shows a RT PL spectrum of the highly uniform QDs, obtained with an excitation density of about 0.5 W/cm². The peak centered at approximately 0.94 eV (~1.32 µm) has a FWHM of 18.0 meV.
obtained using TEG as a gallium source material. This rapid capping suppresses the size reduction and shape change of QDs and In/Ga intermixing that can occur during capping. Figure 4 shows a RT spectrum measured with a much higher excitation power (about 50 W/cm²), where emission from the first and second excited states of the QDs can be clearly recognized. The energy spacing between the first excited and ground states and between the first and second excited states is approximately 70.7 and 67.8 meV, respectively.

We also investigated temperature-dependent optical properties of the highly uniform QDs under an excitation density of about 0.5 W/cm². Figure 5 shows the change of the FWHM with temperature. It is found that the FWHM remains constant within a range of about 0.7 meV (the difference of FWHM between the maximum and minimum) when the temperature is raised from 7 to 300 K. This temperature-independent behavior is distinctly different from that reported previously. Previous reports show an abnormal decrease of PL linewidth with increasing temperature, which mainly results from thermal transfer of carriers from smaller QDs into nearby larger QDs having smaller ground-state energies. This effect is associated with large QD size variations.

In summary, we have demonstrated highly uniform self-assembled InAs/GaAs QDs emitting at 1.3 µm, grown by low-pressure MOCVD. A narrow inhomogeneous linewidth of 16.5 meV was achieved from QDs with a density of \(1.7 \times 10^{10}\) cm⁻². Furthermore, the QDs exhibit almost no dependence of PL linewidth on temperature due to their high uniformity. The results obtained here have significant implications for the realization of high-performance QD lasers for applications in high-speed optical communication systems.

This work was supported by the “Nano-Photonic and Electronic Devices Technology,” Focused Research and Development Project for the Realization of the World’s Most Advanced IT Nation, MEXT.