Photoluminescence and time-resolved photoluminescence characteristics of $In_xGa_{(1-x)}As/GaAs$ self-organized single- and multiple-layer quantum dot laser structures

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The characteristics of ground and excited state luminescent transitions in $In_{0.4}Ga_{0.6}As/GaAs$ and $In_{0.35}Ga_{0.65}As/GaAs$ self-organized single- and multiple-layer quantum dots forming the active regions of lasers have been studied as a function of incident excitation intensity, temperature and number of dot layers. The results have been correlated with molecular beam epitaxial growth conditions. The threshold excitation density for the saturation of the ground state increases with the number of dot layers and no saturation is observed in samples with more than six dot layers up to an excitation power density of 2 kW/cm^2. The luminescent decay times for the ground and excited states are around 700 and 250 ps, respectively, almost independent of the number of dot layers. © *1997 American Institute of Physics.* [S0003-6951(97)03733-9]

Formation of quantum dots by self-organization during molecular beam epitaxial (MBE) growth of highly lattice mismatched layers has gained considerable interest in recent years[1–7] due to the ease with which quasi-zero dimensional structures can be formed, compared to the complexities in nanometer scale processing. High quality room temperature lasers with self-organized InGaAs/GaAs quantum dots as the gain medium have been demonstrated.[8–11] A small-signal modulation bandwidth of 7.5 GHz and a differential gain of 1.7×10^{-14} cm^2 is measured in single mode ridge waveguide lasers with a single layer of InGaAs dots.[11] Since the confinement factor in these lasers is very low (2.7×10^{-3}), due to the relatively low fill factor, a considerable improvement in both the threshold current and modulation characteristics is expected from multiple layers of quantum dots. It has been theoretically predicted[12] and experimentally observed[13,14] that quantum dots formed in successive layers are vertically aligned and electrically coupled when the barrier layers are sufficiently thin. This effect of spatial correlation and self-organization in the growth direction is expected to improve the size uniformity of the dots. We report here the results of a systematic study of the photoluminescence (PL) and time-resolved photoluminescence (TRPL) from single and multiple dot layer laser heterostructures in which we have correlated some of the results with growth phenomena. The characteristics of lasers are described elsewhere.[9,11]

The quantum dot heterostructures were grown in a GEN-II MBE system with uncracked As_4 source. The separate confinement heterostructure (SCH) lasers have $In_{0.4}Ga_{0.6}As$ quantum dots and GaAs barriers in the center of a 0.2 μm GaAs core with 1 μm $Al_{0.4}Ga_{0.6}As$ clad on each side [Fig. 1(a)]. A growth temperature of 540 °C was chosen for InGaAs based on optimum luminescence intensity, while the remainder of the structure was grown at 640 °C. A growth rate of 0.72 μm/h for GaAs and 1 monolayer (ML)/s for InGaAs, and a V/III ratio of 10–15 were used. Under these conditions, a single layer of quantum dots are formed between 7 and 8 MLs of InGaAs, as observed by the change in the reflection high energy electron diffraction (RHEED) spectrum from a streaked to a spotty pattern. Clear formation of dots are observed only with a growth interruption after the dot layer when the thickness of $In_{0.4}Ga_{0.6}As$ is 7 MLs, while it is observed without an interruption when the thickness of InGaAs is 8 MLs. Subsequent layers of quantum dots are formed with only 3 MLs and 5 MLs of $In_{0.4}Ga_{0.6}As$, when the barrier GaAs layers are 1.5 and 2.5 nm thick, respectively. The growth and optimization of multiple dot layers are discussed later. The number of dot layers is varied from

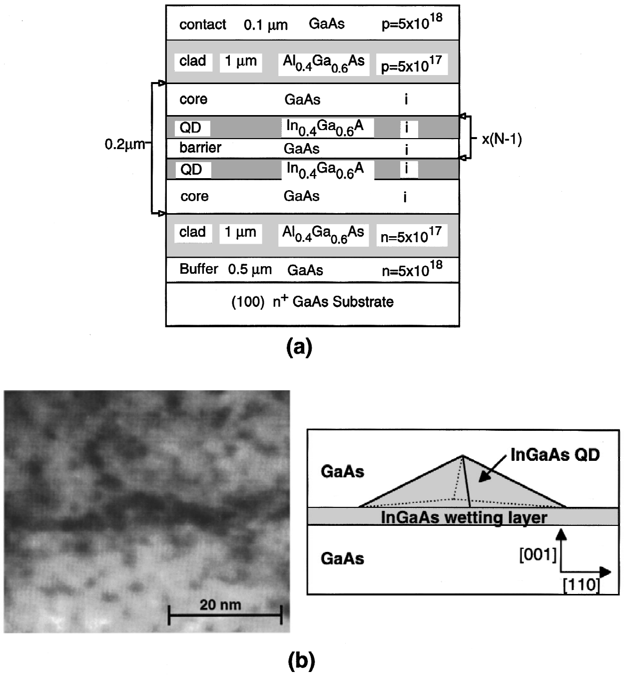

FIG. 1. (a) Separate confinement heterostructure with $In_{0.4}Ga_{0.6}As$ quantum dot layers and GaAs barrier layers where *N* is the number of dot layers; (b) cross sectional transmission electron microscope image of single layer $In_{0.4}Ga_{0.6}As$ quantum dot showing the pyramidal cross section.

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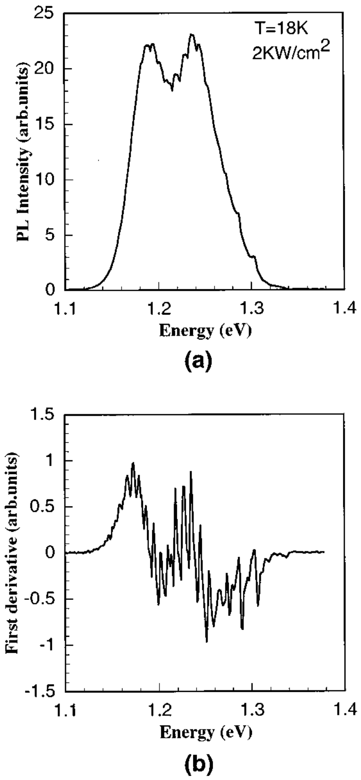

FIG. 2. (a) Low temperature photoluminescence spectrum from $In_{0.35}Ga_{0.65}As$ quantum dots under high excitation intensity ($2\ kW/cm^2$) showing both ground and excited state transitions; (b) first derivative of the spectrum in (a) clearly elucidating the fine structures.

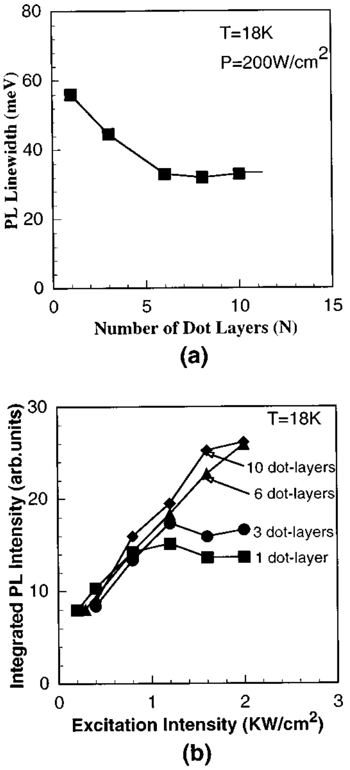

FIG. 3. (a) Photoluminescence linewidth full width half maximum (FWHM) of the ground state emission vs number of dot layers N; (b) excitation dependence of integrated PL intensity of the ground state emission for various number of dot layers.

1 to 10 in our experiments. Some PL measurements were also made in $In_{0.35}Ga_{0.65}As$ dot layers.

Cross-sectional transmission electron microscopy (XTEM) and atomic force microscopy (AFM) images were taken from the layers discussed above and from layers with exposed quantum dots, respectively. The XTEM image shown in Fig. 1(b) indicates the pyramidal cross section of the dot with a base diagonal of 20 nm and a height of 7 nm. The AFM images indicate a narrow range of distribution of the dot size. The density of the dots, measured from a typical AFM image, is $5\times10^{10}\ cm^{-2}$.

PL measurements were made with a 632.8 nm He–Ne laser excitation, 1 m scanning spectrometer, and photomultiplier detection with lock-in amplification. The spectra were recorded at varying excitation power densities and temperatures. Low temperature (18 K) PL spectra from all the samples show a broad peak at relatively low excitation intensity ($20\ W/cm^2$), corresponding to the ground state transition of the quantum dots. Figure 2(a) shows such a spectrum from a single layer of $In_{0.35}Ga_{0.65}As$ quantum dots under high excitation intensity ($2\ kW/cm^2$). The spectrum shows ground state and an excited state transition at around 1.18 and 1.24 eV, respectively. Although the overall linewidth [full width at half maximum (FWHM)] of the transitions is relatively large (30–55 meV), we have observed structures with linewidths as narrow as 4 meV superimposed on these spectra, which we believe is due to the singular density of states in the quantum dots.[15,16] This is elucidated in the first derivative spectrum Fig. 2(b).

It is observed that there is a large decrease in the luminescence intensity, an increase in linewidth, and a large increase in the emission wavelength, with increasing number of dot layers, when the thickness of all the layers of $In_{0.4}Ga_{0.6}As$ is held constant at 7 MLs. The energy difference between the ground state PL emission peaks of single and two dot-layer samples is about 90 meV. This is due to the increasing size and possible coalescing of the dots in the subsequent layers which have lower wetting layer thickness. When the $In_{0.4}Ga_{0.6}As$ layers are grown just enough to form the dots, which in our case is 7 MLs for the first layer of dots and 3 MLs for subsequent layers of dots, with 1.5 nm GaAs in between, there is no significant decrease in the peak PL intensity at 18 K, while the PL linewidth decreases with increasing number of dot layers. This is shown in Fig. 3(a). The linewidth decreases from 56 meV for a single dot layer to 33 meV for a six dot layer sample. We believe this is due to the alignment of dots in the growth direction and the resulting size coherence in lateral and vertical directions. The strain driven self-alignment in the growth direction in multilayer quantum dots leads to a filtering action wherein the dots in subsequent layers align to the larger dots in the previous layers.[12] This process eliminates smaller dots and improves the overall size uniformity. Furthermore, with the optimized thickness of $In_{0.4}Ga_{0.6}As$ for the first layer (7 MLs) and subsequent layers (3 MLs) of quantum dots, the shift in the ground state PL emission energy between single and multiple dot layers is significantly smaller (only about 65 meV for the 10 dot layer sample) than the constant thickness case mentioned earlier.

The integrated PL intensity at 18 K, obtained by a

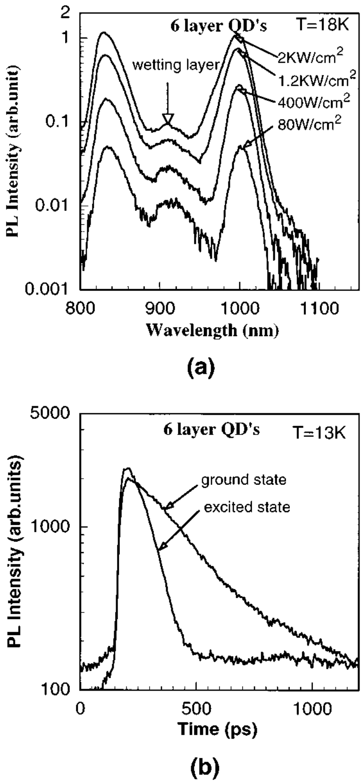

FIG. 4. (a) Low temperature photoluminescence spectra at different excitation levels from the six dot layer sample showing emission from the wetting layer even at the lowest excitation level; (b) time resolved PL intensity from the ground- and excited-state transitions in a six dot layer sample.

Gaussian fit to the ground state emission, is shown in Fig. 3(b) as a function of excitation intensity for samples with varying number of quantum dots layers. The single dot layer sample shows saturation of the ground state emission at fairly low intensities, whereas no saturation is observed in the case of six and 10 layer samples up to an excitation intensity of 2 kW/cm^2. This strongly suggests that lasers with multiple layer quantum dots in the gain region will emit at a wavelength corresponding to the ground state and not from an excited state, as is commonly observed in single dot layer lasers.[9,10] An important observation is that the low temperature PL spectra of three dot layer samples is dominated by ground state and excited state transitions at around $\lambda = 1.0$ and 0.95 μm, respectively. For samples with six or more quantum dot layers, a distinct emission at ~ 0.9 μm, corresponding to the two-dimensional wetting layer, is observed. This is shown in the 18 K PL spectra, for various incident excitation intensities, in Fig. 4(a). Furthermore, the intensity of the wetting layer emission becomes very significant as the sample temperature is increased. These features will have important consequences in the design of multi-quantum dot layer lasers.

The carrier recombination dynamics related to the ground- and excited-state transitions were investigated by TRPL measurements at 13 K with a streak camera. A Gaussian fit to the time integrated PL is used to identify the spectral position of the ground and excited state transitions. The measured PL decay times yield recombination lifetimes of 700 and 250 ps for the ground state and excited state, respectively, almost independent of the number of dot layers. The measured PL decays of the ground and excited state emissions for a six dot layer sample are shown in Fig. 4(b). The short decay time of the excited state is believed to be due to loss of carriers to the ground state and to carrier reemission to the GaAs layers. The larger emission time of the ground-state emission indicates that the oscillator strength of the transition is small, which could result from the pyramidal shape and the complex strain tensor within the quantum dots. We have used the lattice gas computer simulation model to calculate the strain tensor in the self-organized dots. The electronic spectra for the conduction and valence bands are then calculated using a eight-band $\mathbf{k}\cdot\mathbf{p}$ model. The theoretically calculated ground- and excited-state transition energies show good agreement with the measured energies.

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