Temperature-dependent photoluminescence of $In_{0.5}Al_{0.5}As/Al_{0.25}Ga_{0.75}As$ self-organized quantum dots

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The photoluminescence spectra of large ensembles of self-organized $In_{0.5}Al_{0.5}As/Al_{0.25}Ga_{0.75}As$ quantum dots were studied. The spectra reveal a number of sharp peaks with energy separation and full width at half maximum values of $\sim$20 meV for $150\,K \leqslant T \leqslant 275\,K$. Significant changes are observed in the luminescence characteristics with varying temperature, which are attributed to a changing distribution of photoexcited carriers between dots. © *1999 American Institute of Physics.* [S0021-8979(99)02505-0]

A highly successful method of realizing quantum dots is through strained layer growth in the Stranski–Krastanow growth mode, commonly referred to as self-organized growth.[1–3] The formation of quantum dots through self-organized growth has been demonstrated with a variety of heterostructure materials, leading to luminescence in a wide range of emission wavelengths. Luminescence measurements on small ensembles of quantum dots have shown sharp and narrow luminescent peaks,[4] as expected for quantum dots. To improve the usefulness of self-organized quantum dots, it is important to understand the growth and optical properties of the dots such that the size distribution is reduced and narrow luminescence peaks are obtained for a large ensemble of dots. Recently, narrow electroluminescence peaks have been reported for large dot ensembles emitting in the infrared.[5] In this communication, we report the observation of narrow luminescence peaks from red emitting self-organized $In_{0.5}Al_{0.5}As/Al_{0.25}Ga_{0.75}As$ quantum dots and changes in the nature of the emission with variation of temperature.

Heterostructures for photoluminescence (PL) and atomic force microscopy (AFM) measurements were grown by molecular beam epitaxy. The grown structure is shown in Fig. 1 and consists of a GaAs buffer layer followed by a 0.2 μm $Al_{0.25}Ga_{0.75}As$ barrier, both grown at 630 °C. The $In_{0.5}Al_{0.5}As$ quantum dots were then grown at 540 °C with a nominal thickness of 10 monolayers (ML). The quantum dot growth was monitored by examining the reflection high energy electron diffraction (RHEED) pattern and three-dimensional (3D) island growth was found to commence after $\sim$8 ML, which is in agreement with other reports.[6] A 15 s growth interruption at this point allowed further dot formation and was followed by the growth of 0.2 μm of $Al_{0.25}Ga_{0.75}As$ at 630 °C. A layer of $In_{0.5}Al_{0.5}As$ quantum dots was then grown on the surface under conditions similar to the buried dots for AFM measurements, which reveal a typical island size of $\sim$17 nm in base width and 8 nm in height and a density of $\sim 10^{11}$ dots/cm^2.

Photoluminescence measurements were made using an Ar^+ ($\lambda = 488$ nm) laser, a 1 m double grating monochromator, and a cooled photomultiplier with photon counting. The spectral resolution of the measurements performed corresponds to 0.6 meV. The laser spot diameter is $\sim$50 μm, which corresponds to a region containing an estimated 2×10^6 dots. The low temperature ($T = 12$ K) PL spectra for an excitation intensity of $P = 1500$ W/cm^2 shows a broad luminescence peak centered at 1.70 eV. The peak is asymmetric with a long tail at the low energy side. Weak luminescence from the $Al_{0.25}Ga_{0.75}As$ barrier is also observed at 1.86 eV. The PL spectra at $T = 12$ K were examined for a range of lower excitation intensities, down to $P = 15$ W/cm^2, and do not reveal a change in peak shape or position. The broad luminescence is believed to originate from quantum dot ground state transitions from an ensemble of quantum dots of varying size, and hence varying transition energies. The luminescence peak is asymmetric with a low energy tail. We believe that this reflects the dot size distribution with a larger number of smaller dots emitting at higher energies, and a smaller number of larger dots emitting at lower energies. Near $T = 100$ K, we begin to observe sharper features in the luminescence spectra which become more pronounced with increasing temperature. The sharp luminescence peaks also become more pronounced with increasing excitation intensity, which is in contrast to a recent report where sharp features were observed in electroluminescence spectra,[5] but only for smaller current densities. The sharper features are superimposed on a broad luminescence similar to that observed at $T = 12$ K. The overall luminescence feature is from the ground state transition of the quantum dots and the origin of the narrow peaks will be discussed later. As the measurement temperature is increased to near $T = 210$ K and above, we begin to see an additional broad luminescence at higher energies with sharp features superimposed. The relative in-

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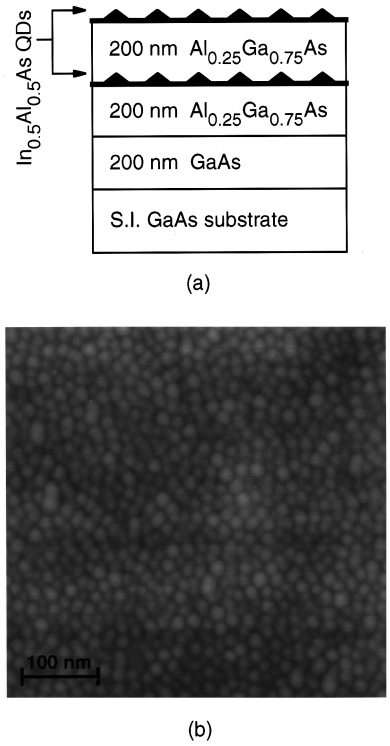

FIG. 1. (a) Schematic of the self-organized $In_{0.5}Al_{0.5}As/Al_{0.25}Ga_{0.75}As$ quantum dot heterostructure under study and (b) atomic force microscope image of the surface quantum dots showing a typical dot density of $\sim 10^{11}$ dots/cm^2.

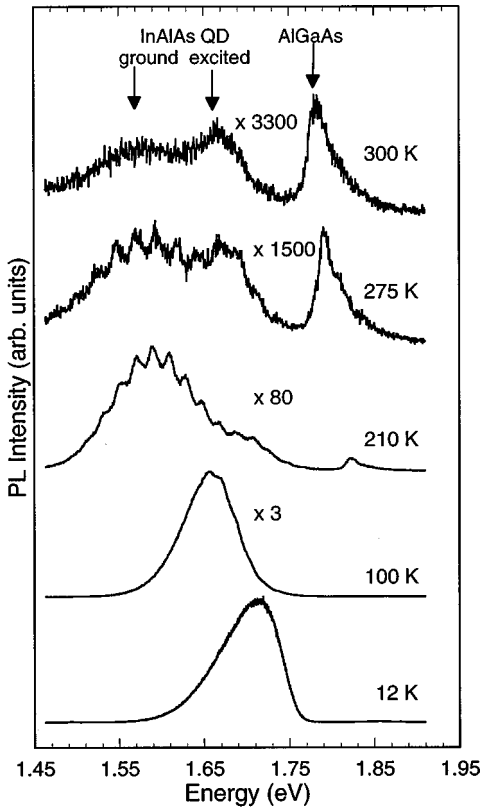

FIG. 2. Photoluminescence spectra of self-organized $In_{0.5}Al_{0.5}As/Al_{0.25}Ga_{0.75}As$ quantum dots for several temperatures in the range of 10–300 K. Notice sharp modulations in the spectra for temperatures above 150 K.

tensity of the higher energy luminescence increases with temperature. At $T=300$ K, the sharp peaks are not observed any more and the broad peak at higher energy is more prominent. At this temperature, the PL signal is weak and it is likely that the sharp peaks may be present, but the signal-to-noise ratio is small. We believe that the broad peak at higher energy is due to excited state transitions in the quantum dots, which has similarly been observed in In(Ga)As/GaAs quantum dots.[7–9]

At $T=300$ K, the two broad luminescence peaks at 1.582 and 1.672 eV have full width at half maximum (FWHM) values of 120 and 61 meV, respectively. The separation between the two peaks is 90 meV, which is similar to the 60–100 meV separation for In(Ga)As quantum dots measured by photoluminescence[7–9] and far-infrared absorption[10] measurements. At temperatures between $T=150$ and 275 K, the apparent sharp peaks in the PL spectra allow good Gaussian fits to be obtained. The fitted peaks have FWHM values between 18 and 22 meV and a separation of 19–24 meV, with no superimposed baseline or broad continuum in the fitting. Both the FWHM and energy separation of the peaks decrease with the lowering of temperature, but only by 4 meV for the FWHM, and 5 meV for the peak energy separation. This is significantly less than the change in kT in this temperature range (8 meV), indicating that the sharp peaks do have some associated inhomogeneous broadening. Series of sharp PL peaks, such as those shown in Fig. 2, are usually attributed to fringes in a Fabry–Pérot cavity or phonon replicas. In the present case, however, both of these possibilities are ruled out for the following reasons. The observed energy separation of 19–24 meV corresponds to a cavity thickness between 3.7 and 4.7 μm, which is too large for the layer thicknesses in our sample. Likewise, the longitudinal optimal (LO) phonon energies in AlAs, GaAs, and InAs (50, 35, and 29.6, respectively) are too large to account for the energy separation of the observed sharp peaks. If the sharp peaks were due to fluctuations in dot size or alloy composition, they should be more pronounced at low temperatures which contradicts the observed temperature dependence shown in Fig. 2.

We believe that the observed temperature-dependent characteristics of the luminescence spectra can be understood in terms of the distribution of carriers among the dots. Changes in the PL line shape of InAs/GaAs self-assembled quantum dots with varying temperature have similarly been observed and attributed to carrier transfer in the dots.[11,12] At very low temperatures, ($T \leqslant 100$ K), the photoinduced carriers are frozen in the dots in a nonequilibrium distribution. A single broad PL peak reflects ground state transitions from this group of dots. As the measurement temperature is increased ($100 < T < 280$ K), the photoinduced carriers are distributed more widely among the entire dot population. Since the latter vary in size, and perhaps slightly in shape depending on growth conditions, the ground state transition energies vary by small amounts. The origin of the observed sharp peaks may therefore be attributed to discrete groups of dot sizes. Evidence of this phenomenon has also been reported

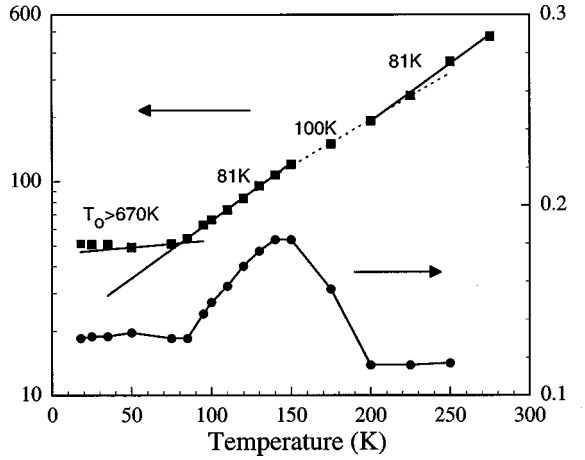

FIG. 3. Measured slope efficiency and characteristic temperature of a four-layer $In_{0.4}Ga_{0.6}As/GaAs$ quantum dot laser. The clear increase in characteristic temperature and slope efficiency near $T=100-150$ K suggest a change in carrier distribution in the dots, similar to the sharp modulations occurring near $T=150$ K in the photoluminescence spectra of the $In_{0.5}Al_{0.5}As/Al_{0.25}Ga_{0.75}As$ quantum dots.

for self-organized InAs/GaAs[12,13] and InAs/InP (Ref. 14) quantum dots, where modulations in PL spectra were observed with energy spacings of 20–30 and ~50 meV, respectively. At the same time, with the increase of temperature, a transition from the dot excited states is also observed. As the temperature is increased further ($T \geqslant 280$ K), the signal-to-noise ratio decreases and the narrow PL peaks are not observed. Instead, the dot luminescence is characterized by two broad peaks resulting from ground and excited state transitions from a large size distribution of dots. Our explanation of the observed temperature-dependent PL is also supported by a related experiment in which the slope efficiency of four-dot layer $In_{0.4}Ga_{0.6}As/GaAs$ quantum dot lasers has been measured as a function of temperature. The results are shown in Fig. 3. For $T \leqslant 100$ K the slope efficiency is low and maintains a constant value because only the carriers in a fraction of the dots participate in lasing. At higher temperatures, as the carriers equilibrate among the entire dot population and contribute to lasing, the slope efficiency rapidly increases. After going through a peak value, the slope efficiency decreases due to carrier heating and leakage effects, which is characteristic of any semiconductor laser. A similar behavior is also displayed by the threshold current J_{th}. The characteristic temperature T_0, given by $[J_{th}(T) = J_{th}(0)\exp(T/T_0)]$, remains at a constant high value ($\geqslant 600$ K) for $T \leqslant 100$ K, and drops to ~80–100 K for $100\,\text{K} < T < 300$ K, as shown in Fig. 3.

In summary, we have observed sharp luminescence features in a large ensemble of self-organized $In_{0.5}Al_{0.5}As/Al_{0.25}Ga_{0.75}As$ quantum dots. The sharp features are attributed to luminescence from discrete groups of dot sizes. These features also exhibit a temperature dependence which is due to the changing distribution of carriers in the dots at varying temperatures. With further understanding and control of self-organized growth, a narrow dot size distribution may be realized for large ensembles of dots leading to narrow luminescence linewidths useful for optoelectronic applications.

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