InGaAs/GaAs three-dimensionally-ordered array of quantum dots

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(Received 6 November 2002; accepted 30 May 2003)

We report on the first fabrication of InGaAs/GaAs quantum dots with both vertical and lateral ordering forming a three-dimensional array. An investigation of the photoluminescence spectra from the ordered array of quantum dots, as a function of both temperature and optical excitation intensity, reveals both a lateral and vertical transfer of excitation.

Recently, electronic and optoelectronic devices based on quantum wires and quantum dots (QDs) have been the subject of interest due to their potential applications as lasers, detectors, or photonic crystals.1 As a result, there has been an extensive effort to manipulate and control the position, size, shape, and density of QDs.2–5

In this letter, we report on the fabrication of a vertically and laterally ordered InGaAs/GaAs QD stack forming a three-dimensional QD array. The fabrication is achieved by utilizing multilayer vertical stacking grown at elevated temperature.6–11 An investigation of the photoluminescence (PL) spectra from these ordered arrays of QDs, as a function of both temperature and optical excitation intensity, reveals both a lateral and a vertical transfer of excitation.

The samples used in the experiments were grown on semi-insulating GaAs [001] substrates, with a miscut angle smaller than 0.05°, using solid source molecular beam epitaxy. After the native oxide was desorbed, a 150-nm-thick GaAs buffer layer was grown at 580 °C. The substrate was then cooled down to 540 °C for the growth of the multilayer dot structure. After a 2-nm-thick In0.36Ga0.64As layer was grown, three monolayers of GaAs were deposited without growth interruption to suppress In segregation. Then, after an 8 s interruption, 16 nm of GaAs was grown. The period of the superlattice was 15.

The samples were investigated using plane-view and cross-sectional transmission electron microscopy (XTEM) in order to examine vertical and lateral ordering was performed using a JEOL JEM2000FX microscope operated at 200 kV. PL studies were performed in a variable-temperature helium cryostat (4–300 K), under the excitation of the 514.5 nm line of a continuous-wave argon-ion laser. The PL signal was analyzed using a 0.5 m single-grating spectrometer, and detected using a photomultiplier tube.

Figures 1(a) and 1(b) show dark-field plan-view TEM and dark-field XTEM images of a typical multilayered QD sample, respectively. The XTEM image [Fig. 1(b)] shows an almost perfect vertical island correlation, and the plan-view TEM image [Fig. 1(a)] indicates that within each layer, the QDs were slightly elongated along [T10] direction and densely packed in long lateral chains (~1–2 μm in length). The effective 2D wetting layer (2DWL) thickness (about 0.7 nm) is the same for all layers. The average diameter of the QDs is about 45 nm while the average height is about 5 nm.

The average distance between QDs within a chain is about 20 nm. The separation between a QD in one chain and the nearest QD in a neighboring lateral chain is about 70–80 nm. The QDs in each chain appear to sit on a common InGaAs base with an estimated height of about 1.5–2 nm. The vertical correlation of the QDs is due to the vertical strain field between the buried and the subsequent QDs, while the lateral ordering of the QDs is related to the strain-field-modulated surface along the [T10] direction and the enhanced adatom migration length at the elevated growth temperature.

According to the XTEM, the tunnel barrier thickness between the vertically aligned QDs (defined as the average distance between the island tip in one layer and the island base in the second InGaAs layer) is about 10 nm and is approximately two times smaller than the barrier thickness between neighbor dots within a lateral chain. Based on calculations using the Wentzel–Kramers–Brillouin approximation and treating the dots as thin quantum wells,12,13 the electron tunneling time is estimated to be $\tau_e \sim 0.8$ ns for square barriers of about 10 nm wide. In comparison, the typical radiative recombination time $\tau_r$ for InGaAs quantum dots is about 0.5–2 ns.3,14 So, at low temperature, $\tau_r$ is comparable

![FIG. 1. (a) Dark-field plane-view TEM image of the sample obtained with g = 200. (b) [T10] XTEM image of the sample.](/appliedphysicsletters/83/5/987/fig1.jpg)

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to $\tau_r$, or the tunneling rate is on the order of or may even exceed the carrier recombination rate. Consequently, carriers captured by a dot in a given column are predicted to migrate only along the column at low temperature and become localized in the dot with the lowest ground state energy before recombining radiatively. Therefore, for low excitation intensity $I_{ex}$, optical emission will come from the ground states of the larger dots in each vertically correlated stack. With increasing $I_{ex}$, however, the ground states of these dots will saturate and carriers will recombine also from the ground states of smaller dots in the stack. As a result, we should expect the PL peak to broaden asymmetrically towards higher energy as $I_{ex}$ increases, which is confirmed by the experimental data shown in Fig. 2. Under low $I_{ex}$, the QD emission is described by a single Gaussian profile with a full width at half maximum (FWHM) of 55 meV. As $I_{ex}$ increases, the PL peak exhibits a blueshift with a simultaneous asymmetric broadening towards the high-energy direction as predicted. At $I_{ex}=200$ W/cm$^2$, the FWHM of the PL spectrum (70 meV) is 1.3 times of that for $I_{ex}=0.1$ W/cm$^2$ (see inset in Fig. 2).

At even higher intensities (>600 W/cm$^2$) a well-defined second peak appears in the spectra at about 1.385 eV, which becomes stronger in intensity with increasing $I_{ex}$. At the $I_{ex}$ above 2500 W/cm$^2$ a third peak at 1.445 eV emerges. We do not believe that these new peaks are associated with emission from the QDs. Theoretical estimates on the electronic structure of our QDs based on Ref. 15 show clearly that ground state transition energies greater than 1.25 eV posses only a single bound electron state. Therefore, to be associated with the QDs, these new emission lines would have to be transitions from the single electron state ($e_0$), to excited hole states ($h_i$). But such $e_0\rightarrow h_i$ transitions are forbidden optically, and would be expected to be quite weak. A more likely explanation, and the one we take here, is that the origin of the additional PL peak at 1.385 eV at a high $I_{ex}$ is the existence of a common InGaAs base for all QDs sitting on one chain.16 For the QDs within one chain, the common base may be considered as a one-dimensional wetting layer (1DWL), analogous to the 2DWL generally existing in Stranski–Krusznev growth mode (see inset in Fig. 4). A calculation for the transition from ground-electron to ground-hole states ($E_{1DWL}$) for a wire-like In$_{0.36}$Ga$_{0.64}$As structure (with a lateral width of about 40 nm and a height of about 1.5 nm) predicts a PL peak at 1.390 eV$^{15}$ that is very close to the position of the second peak in PL spectra with high $I_{ex}$, (Fig. 2). Meanwhile, we attribute the small third peak observed at an even higher $I_{ex}$ at 1.445 eV to the manifestation of the 2D In$_{0.36}$Ga$_{0.64}$As WL.

In addition to the behavior of PL versus excitation intensity, we also explored PL as a function of temperature. At low $I_{ex}$ intensity, with increasing temperature ($T$) we may expect the process of thermal carrier transfer between the QDs within each chain to become important. Indeed, according to a simple thermionic model, at low $T$, carriers are frozen randomly into the dot states (after initial partial redistribution due to the tunneling between QDs within each vertical column). With increasing $T$, carriers are expected to be thermally activated outside the dots into the 2DWL and/or the GaAs barrier and then relax into different dots. Carriers hopping between dots will favor a drift of carriers toward the dots having lower excited state energies, resulting in a narrowing of the PL spectrum and a redshift for the PL peak that is larger than expected from the thermal shrinkage of the InGaAs band gap with elevating $T$. Figure 3(a) plots the temperature dependence of the PL emission from the sample with $I_{ex}=0.4$ W/cm$^2$. The PL spectra show a significant redshift with elevating $T$. At the same time, the PL spectrum first narrows (from 60 to 40 meV) in the temperature range between 20 and 100 K, then monotonously broadens for $T>100$ K. The temperature dependences of the FWHM and PL peak position are plotted in Figs. 3(b) and 3(c), respectively. The low intensity PL redshift shown in Fig. 3(c) is essentially greater than that of the In$_{0.36}$Ga$_{0.64}$As free exciton emission (dotted line) at $T>60$K.

As $I_{ex}$ is increased, the ground states of all QDs are expected to be occupied and the carrier transfer between QDs in the ensemble should practically vanish. In this case one can expect the absence of any narrowing of the PL spectrum and the same temperature dependence of the PL peak as the InGaAs band gap with increasing $T$. These behaviors are confirmed in the observed PL spectra [Figs. 3(b) and 3(c)]. For $I_{ex}=100$ W/cm$^2$, the FWHM of the QD PL peak is observed to be constant up to 200 K and then, in the range of 200–300 K, to increase monotonously [Fig. 3(b)]. At the same time, with increasing $I_{ex}$, the difference between the temperature dependence of the QD PL peak position and the In$_{0.36}$Ga$_{0.64}$As free exciton emission diminishes [Fig. 3(c)].

Consistent with the earlier discussion, the temperature at which both the narrowing and shifting of the QD PL spectra occur should be determined by the energy separation between the ground state of the smaller dots and the energy level of the 2DWL. In Ref. 11, for the PL spectra of InGaAs QDs, with approximately the same size distribution but lateral randomly distributed, the effective carrier transfer process (that produces a narrowing PL line and an acceleration of the PL peak redshift) begins at $T\geq 100$ K. In our sample the narrowing of the PL spectrum begins at about 20 K, a significantly lower $T$ [Fig. 3(b)]. This behavior supports the
existence of an additional energy level located between the 2DWL and ground state of the smallest QDs and is consistent with the proposed 1DWL with an energy level at about 1.385 eV. Indeed, as seen from the inset in Fig. 3(a), the dependence of integrated PL intensity for QDs as a function of temperature exhibits the Arrhenius behavior typically observed for quantum wells. The thermal activation energy extracted from this Arrhenius plot is found to be \( (130 \pm 10) \) meV. This value is significantly smaller than the energy difference between the ground state PL peak position and peak at 1.445 eV caused by the 2D In\(_{0.36}\)Ga\(_{0.64}\)As WL (180 meV) or at 1.517 eV caused by the GaAs barrier (257 meV) which observed for high \( I_{\text{ex}} \), but it is very close to the energy difference between the PL peak position and the \( E_{\text{1DWL}} \) (about 120 meV). Therefore, due to the relatively small energy separation at 20–30 K, carrier thermal emission begins out of the smaller QDs and is captured into the 1DWL energy level. The captured carriers can relax over a longer distance along the chain and find a lower energy minimum among the ensemble of QDs in the chain.

An intriguing result was observed in investigating the polarization anisotropy (PA) for the ground QD PL transition as a function of temperature. As shown in Fig. 4, the PA, defined as \( \delta = (I_- - I_+)/(I_+ + I_-) \), where \( I_+ \) and \( I_- \) denote the PL intensities with the polarization parallel and perpendicular to the chain direction ([\( 1\overline{1}0 \)]), respectively, increases with temperature. This temperature dependence of PA can be considered as an additional confirmation for active interdot carrier transfer with increasing \( T \). Indeed, due to the slightly elongated shape of QDs in such system along coalescence direction [\( 1\overline{1}0 \)], one can expect the existence of definite PA in ground optical transition in [\( 1\overline{1}0 \)] and [\( 1\overline{1}0 \)] directions. On the other hand, according to the Ref. 17 the larger dots in such ensemble must have a larger anisotropy due to increasing with size the shear strain and piezoelectric effects. So inasmuch at \( T > 50–70 \) K the ground state QD PL emission is caused by larger dots due to thermionic redistribution of carriers one can expect an increasing PA. At \( T > 120–150 \) K the growth of PA almost stops as thermal emission of carriers out of the larger QDs (and recapture by all dots) in the ensemble becomes significant.

In summary, we have fabricated a three-dimensional array of (In,Ga)As/GaAs QDs that offers the possibility of engineering interesting electric transport properties. The three dimensionality of the array is supported by planar and cross-sectional TEM and by PL spectral data.

The authors acknowledge financial support of NSF PHY-0099496 and DMR-0080054.

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16 As clearly seen from Fig. 1(a) the plane-view TEM image shows a darker contrast between QDs than that of the regions between chains. Generally such contrast difference could be a consequence of several possibilities, i.e., differences in strain field, chemical composition, as well as thickness. Following our PL data, we consider the main reason for darker contrast between QDs comparing with the regions between chains is due to thickness difference.