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InAs/GaP semiconductor quantum dots (QDs) were spontaneously formed using direct island growth (Volmer–Weber) rather than Stranski–Krastanow (S-K) growth. Structural investigations of InAs/GaP QDs suggest kinetically limited growth and show a broad size distribution. Photoluminescence and cathodoluminescence spectroscopy reveal large inhomogeneous broadening with the emission peak centering at 1.7 eV. Device applications exploiting broad optical emission in QDs are discussed. © *1998 American Institute of Physics.* [S0003-6951(98)02911-8]

Semiconductor structures of reduced dimensionality are of considerable interest since lateral quantum confinement leads to new optoelectronic properties. Device implementation using these zero-dimensional (0D) structures is providing interesting challenges in quantum electronics, and many of these structures are already proving advantageous for technological applications.[1]

In S–K growth of quantum dots (QDs), initial deposition starts with formation of a stable two-dimensional wetting layer (WL). After this WL reaches a strain dependent critical thickness, islands begin to form and their concentration increases exponentially with further deposition. Effectively a very thin quantum well with a bound state near the emission for the barrier material, this WL can produce a separate photoluminescence (PL) peak which is intense for low densities of S–K QDs.

Here we report on a new type of quantum dot, based not on S-K growth, but on direct island formation or Volmer–Weber (V-W) growth.[2] V-W growth is observed for large lattice mismatches (11.4% here) and/or dissimilar crystal structures. V-W growth occurs by spontaneous three-dimensional island formation without prior 2D growth. This eliminates the effects of the WL layer, which accounts for many of the optical properties in S-K QDs. The extreme differences in band gap (1.85 eV) in between InAs and GaP also creates a large confining potential, despite the band edge shift which has the effect of making the confining potential shallower due to high strain. A deeper confining potential provides advantages in optoelectronic performance at elevated temperatures.[3]

A recent analysis of the suitability[4] of self-organized QDs for the application of persistent spectral-hole burning in frequency domain optical storage shows that QD structures are ideally suited for such applications. Due to the greater fluctuation in sizes, the InAs/GaP quantum dot structures presented here show even larger inhomogeneous broadening, making the ratio of inhomogeneous to homogeneous broadening larger than in S-K QDs. The observation of room temperature luminescence from these structures makes this application uniquely attractive. Incorporating these and other broadly emitting QDs in transmitters for Wavelength division multiplexing (WDM) would have advantages over quantum well lasers, which have emission at closely spaced wavelengths. Using light emitting diodes (LEDs) provides a wider optical emission band,[5] but coupling to fibers is inefficient. The QD structures described here could combine the higher coupling efficiency of semiconductor lasers with the broad band optical emission of LEDs.

InAs islands were grown on (100)-oriented GaP by solid-source molecular beam epitaxy (MBE). Phosphorus molecular beams were generated by a valved solid cracker source.[6] Reflection high-energy electron diffraction (RHEED) monitored surface morphology and growth rates for both GaP and InAs. After growth of a 100 nm GaP buffer layer, a twenty period superlattice composed of 5 nm alternating layers of $Al_{0.2}Ga_{0.8}P$ and GaP were grown to prevent sulfur out diffusion from the GaP substrates. After growth of a second 100 nm GaP buffer layer, InAs islands were formed by depositing 1 monolayer (ML) of InAs in five 1 s pulses (followed by a 3 s interruption) at a growth rate of 0.2 ML/s. When capped structures were used, 30 nm of GaP was grown above the InAs islands. InGaAs/GaAs and InAlAs/AlGaAs QD were grown by metalorganic chemical vapor deposition. Nominal ternary compositions were $In_{0.6}Ga_{0.4}As$, $In_{0.57}Al_{0.43}As$, and $Al_{0.34}Ga_{0.66}As$. Growth conditions for these are described elsewhere.[7] Low (77 K) temperature PL spectra were obtained using the 488 nm line of an argon ion laser, or the 532 nm continuous-wave output of a diode-pumped Nd:YVO4 for excitation, the signal was dispersed with a single grating 0.67 m monochromator, and

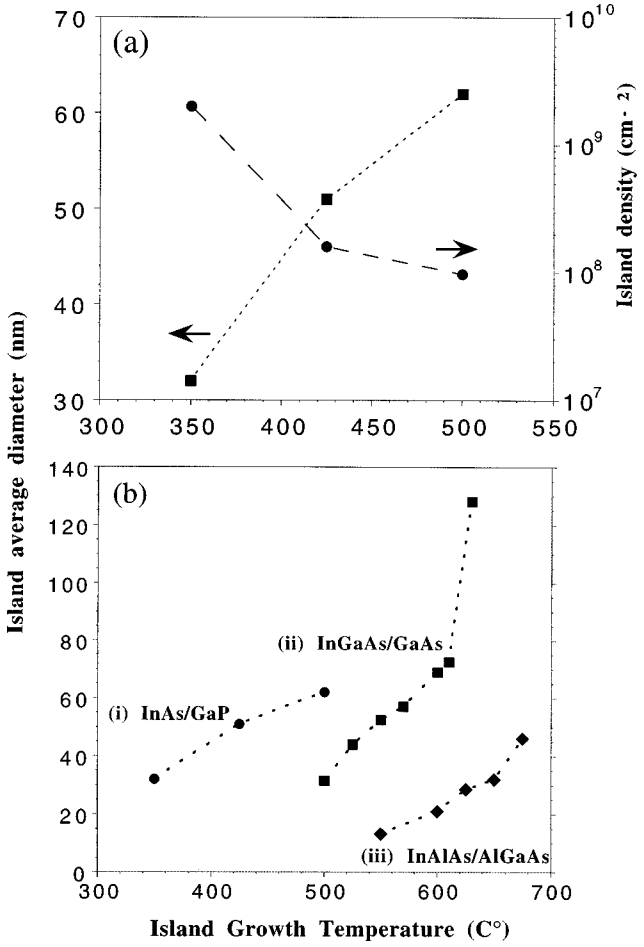

FIG. 1. (a) Temperature dependence of average sizes and concentrations in InAs islands self-formed on GaP surfaces. Values were obtained from statistical analysis of FM images. (b) Comparison of island diameters obtained for (i) InAs/GaP V-W QDs and for S-K type QDs, (ii) InGaAs/GaAs, and (iii) AlInAs/AlGaAs.

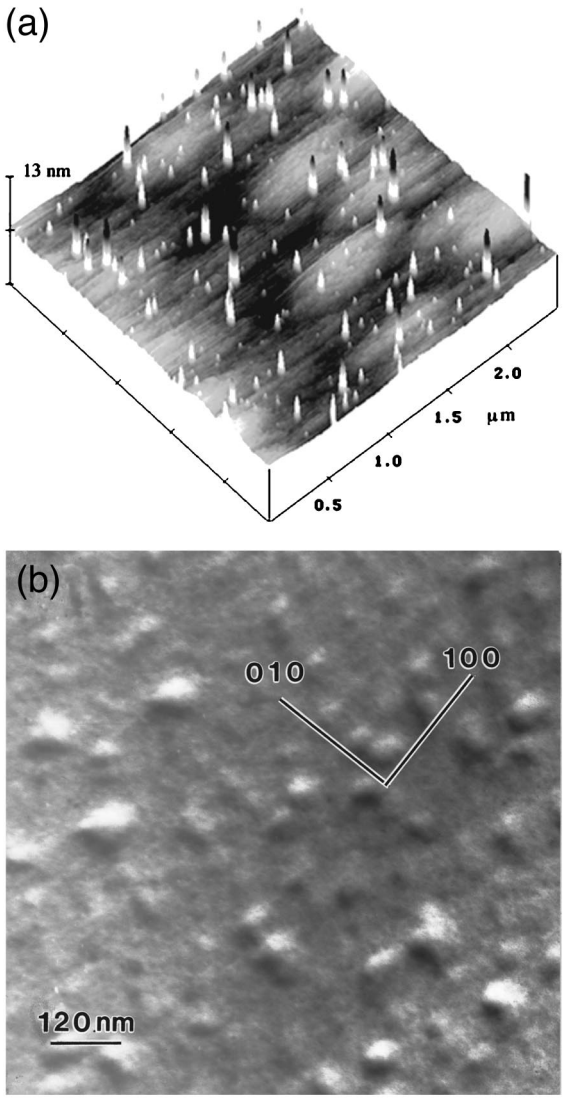

FIG. 2. (a) Surface FM plot of InAs islands on GaP showing broad size distribution. Vertical enhancement: $Z=28(X,Y)$. Micron scale undulations are seen on the surface of GaP for many of the growths independently of InAs island formation. (b) Plan view TEM micrograph of InAs dots in a GaP matrix.

collected using a photomultiplier tube or a cooled Ge detector and lock-in techniques. Cathodoluminescence (CL) spectroscopy and imaging was carried out in a JEOL JSM35C scanning electron microscope with a liquid He cold stage using an Oxford Instruments MonoCL2 system. InAs/GaP island formation was examined for various growth temperatures using force microscopy (FM). Plan view transmission electron microscopy (TEM) (JEOL 200CX operated at 200 keV) was also used to obtain structural information from capped samples. FM and TEM gave statistical information on island sizes, shapes, size uniformity, and areal densities.

InAs island formation was obtained with 1 ML deposition. FM and TEM analysis of the islands show that all of the deposited InAs was converted to 3D islands, with no WL. Therefore, we attribute the growth mode to direct island formation or V-W growth.

Analysis of the surface islands show that as in S-K island formation, smaller and more numerous quantum dots are formed at lower growth temperatures.[7] Plots of temperature dependence of island sizes and surface densities are shown in Fig. 1. Comparisons of island sizes for InAs/GaP QDs and S-K type QDs (InGaAs/GaAs and AlInAs/AlGaAs)[1,8–10] are presented in Fig. 1(b). While both types of island formation show kinetically limited growth, larger diameters for a given growth temperature are obtained in InAs/GaP QDs than in the S-K QDs mentioned above.

Statistical analysis of these InAs/GaP islands shows another important difference with respect to S-K islands,[11] and this is a much larger size dispersion. Figure 2 shows an FM surface plot and a plan view TEM image illustrating island densities and morphologies. Large size inhomogeneities in these islands can be seen with either imaging technique. The island diameters are comparable from FM or TEM measurements, but the island surface densities appear higher when this is measured with plan view TEM. This discrepancy could be due to ripening during cooling in uncovered islands. It is also possible that some of the smaller islands are missed with the FM plane-fit due to GaP surface roughening.

The islands are lens shaped or hemispherical when small. For the larger islands, some facetting is seen to occur in FM, and TEM examination also shows that some of the

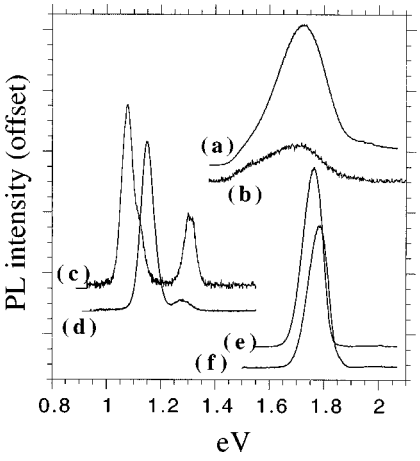

FIG. 3. PL at 77 K (a) from InAs/GaP QDs at 77 K and (b) at 300 K. (c), (d) From low surface densities of InGaAs/GaAs quantum dots. Wetting layer luminescence is shown on both spectra. (e) and (f) PL intensity from AlInAs/AlGaAs quantum dots shown for comparison (77 K).

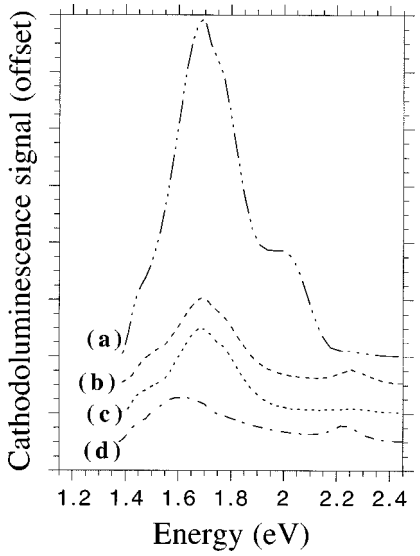

FIG. 4. CL spectra from capped InAs islands grown on GaP. (a) 15 keV electron beam accelerating voltage at a sample temperature of 8 K. (b), (c) 300 K and plan view excitation at 20 and 15 keV, respectively. (d) Edge excitation of a cleaved sample away from the surface. 20 keV and 300 K.

larger dots have a rectangular base with sides parallel to the ⟨100⟩ crystallographic directions. No dislocations associated with these islands were found in TEM or CL imaging.

PL spectra of capped InAs/GaP QDs are shown in Fig. 3(a) for low (77 K) and 300 K [Fig. 3(b)]. The FWHM is broad (215 meV), as expected[9] from the PL emission from QDs with a nonuniform size distribution. The peak is centered at 1.73 and extends from the visible red to the near infrared. Other differences between S-K and V-W QDs become evident when comparing PL spectra of InAs/GaP QDs with that obtained from InGaAs/GaAs QDs in similar concentrations. Figures 3(c) and 3(d) show PL spectra from InGaAs/GaAs QDs for two different values of QD surface densities. As seen in the figure, WL emission is still strong for InGaAs/GaAs QDs in densities of $7\times10^9/\mathrm{cm}^2$ and $8\times10^8/\mathrm{cm}^2$[curves 3(b) and 3(c), respectively]. The QD surface density for the InAs/GaP QDs shown in Fig. 3(a) is estimated to be $(3-4)\times10^9/\mathrm{cm}^2$ from plan view TEM. For comparison, Figs. 3(e) and 3(f) show emissions from S-K AlInAs/AlGaAs QDs of slightly different sizes (32 and 30 nm, respectively).

CL imaging from cleaved specimens reveals that emission around 1.73 eV originates near the surface. Figure 4(d) shows CL spectra collected from the central portion of a cleaved specimen, away from the surface. CL spectra collected near the surface shows a prominent peak that is much weaker away from the surface. The origin of the higher energy peak seen from the more surface sensitive CL at 8 K [Fig. 4(a)] could be from smaller QD that are quenched from thermionic emission at elevated temperatures due to shallower confining potentials. CL imaging thus narrows down the origin of the 1.7 eV broad luminescence to the surface region, minimizing the effects of bulk luminescence. In addition, the possibility of emission associated with dislocations is unlikely. However, it is possible that emission from deep interface states may also contribute to the PL and CL spectra presented here.

Device applications incorporating these structures are very promising. The broader PL peak from inhomogeneous broadening can be exploited in applications requiring wavelength independent optical properties, making InAs/GaP QDs promising for WDM and frequency-selective optical memories based on persistent hole burning.[4] Additional experimental work is needed to determine values for radiative lifetimes, measure inhomogeneous broadening from the individual QD PL emission and achieve greater QD surface densities. InAs/GaP has not been used toward any quantum well (QW) applications due to the large lattice mismatch. Many fundamental issues regarding interfacial and materials properties of the InAs/GaP system thus remain to be investigated.

To conclude, deep confinement QDs have been made by direct V-W growth of InAs islands of nanometer dimensions imbedded in GaP. A broad distribution in island sizes could be associated with a large inhomogeneously broadened PL and CL emission in a range that includes red luminescence. Temperature dependence of island size formation indicates that sizes and concentrations in these QDs can be tuned.

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