

Controlled InAs quantum dot nucleation on faceted nanopatterned pyramids

P. S. Wong, G. Balakrishnan, N. Nuntawong, J. Tatebayashi, and D. L. Huffaker^{a)}
Center for High Technology Materials, University of New Mexico, 1313 Goddard SE, Albuquerque, New Mexico 87106

(Received 19 December 2006; accepted 30 March 2007; published online 30 April 2007)

The selective quantum dot (QD) nucleation on nanofaceted GaAs pyramidal facets is explored. The GaAs pyramids, formed on a SiO₂ masked (001) GaAs substrate, are characterized by well-defined equilibrium crystal shapes (ECSs) defined by three crystal plane families including $\{11n\}$, $\{10n\}$, and (001). Subsequent patterned QD (PQD) nucleation on the GaAs pyramidal facets is highly preferential towards the (11n) planes due to superior energy minimization. The GaAs pyramid ECS and PQDs are examined using high-resolution scanning electron microscopy and room temperature photoluminescence. © 2007 American Institute of Physics. [DOI: 10.1063/1.2732825]

Quantum dots (QDs) formed by the Stranski-Krastanow (SK) growth mode are characterized by random nucleation, which results in nonuniform size and shape distribution. In the SK growth process, QD size, shape, and density are strongly linked by surface kinetics as determined by growth temperature, V/III ratio, and strain energy. Independent control over these QD parameters may be attractive for some device applications, especially those involving single carrier or intraband processes. However, it is difficult to alter the specific QD shape, for example, without affecting size or density. Arbitrary QD placement may enable better control of QD shape and size as surface kinetics can then be designed to produce a desired QD characteristic. A wide range of methods has been pursued to introduce arbitrary QD site placement including strain engineering,^{1,2} locally strain-enhanced etching,³ and lithographic patterning.⁴⁻⁷ These methods have produced very unique and promising results including the demonstration of coupled QD molecules,³ in-plane lasers,⁶ and single QD spectroscopy.⁴ In all of these categories, however, size/shape variation along with nonradiative recombination associated with patterning are unresolved and limit high performance device realization.

The nucleation of the PQD atop a GaAs pyramidal buffer has also been explored as an attractive approach to address radiative recombination issues and QD uniformity. The pyramid both separates the PQD from the processed interface and provides a nucleation platform of sufficiently small dimension to realize quantum size effects.⁷⁻¹⁰ While high quality PQD formation and room-temperature photoluminescence (RTPL) have been previously demonstrated, details of the pyramidal shape and the affect on PQD nucleation have not been observed or reported.

In the work described here, we have carefully characterized the GaAs pyramid equilibrium crystal shape (ECS) and correlated subsequent PQD nucleation trends to specific pyramidal facets. Clear observation and identification of the crystal faceting in both GaAs pyramids and surface PQDs are enabled using both plan-view and cross-section high resolution scanning electron microscopy (HRSEM). The PQD optical quality and simple band structure are characterized by RTPL and 77 K PL. The PL spectra are measured

with a 5 mW continuous wave He-Ne laser (spot size ~1.5 mm) and an InGaAs detector.

The sample growth is carried out using a low-pressure (60 torr) vertical Thomas-Swan metal-organic chemical-vapor deposition reactor with trimethylgallium, trimethylindium, and tertiarybutylarsine. The samples are grown on (001) GaAs substrates covered with a SiO₂ mask (25 nm thick) patterned using interferometric lithography and dry etching.¹¹ The patterning process results in circular openings of 230 nm (± 10 nm) in diameter with a pitch of 330 nm. The GaAs pyramids are grown at 700 °C to form three unique sets of limiting crystal planes described as pyramids A, B, and C. Specific details of GaAs pyramidal formation and faceting are described elsewhere.¹² The temperature is then reduced to 510 °C for the In(Ga)As PQD growth. For PL analysis, the PQDs are capped with InGaAs and GaAs also at 510 °C.

We analyze PQD nucleation on the three unique GaAs pyramid shapes A, B, and C shown in plan-view SEM images of Figs. 1(a)–1(c), respectively, along with schematic illustrations of limiting crystal facets. Pyramid A in Fig. 1(a) has six hexagonal facets representing $\{115\}$ and $\{105\}$ groups, and a (001) apex. Pyramid B, in Fig. 1(b), is defined by the $\{115\}$, $\{105\}$, $\{113\}$, and $\{103\}$ facet groups, and a (001) apex. Pyramid C, in Fig. 1(c), is defined by $\{111\}$, $\{011\}$, and $\{113\}$ facet groups with a (001) apex. Corresponding cross-sectional profiles of the GaAs pyramids are examined by HRSEM in Fig. 2 to understand pyramid geometry. The im-

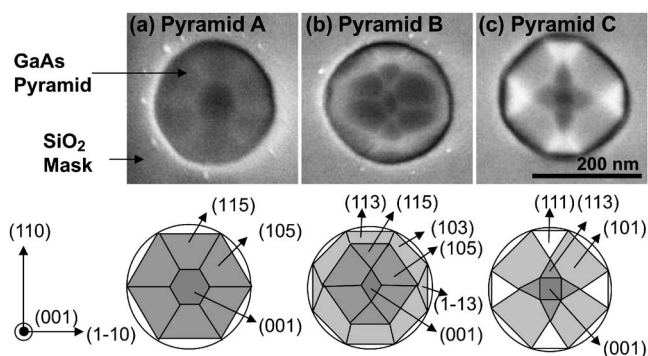


FIG. 1. Top-view SEM images and the corresponding structural schematics of three unique types of GaAs pyramids.

^{a)}Electronic mail: huffaker@chtm.unm.edu

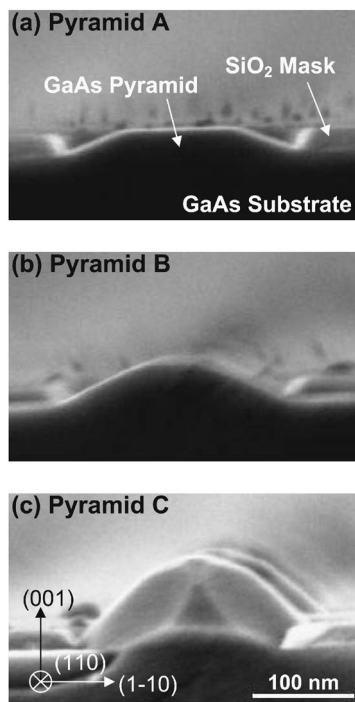


FIG. 2. Cross-sectional SEM images of the three types of GaAs pyramids.

ages indicate a pyramidal height ranging from ~ 30 to 90 nm in pyramids A, B, and C, respectively. To ensure that the observed cleave plane intersects the pyramid center, special care is taken to scan along the cleaved wafer edge to identify the individual structure with maximum height.

Figures 3(a)–3(f) show plan view HRSEM images of surface PQDs formed on pyramids A, B, and C. The figures

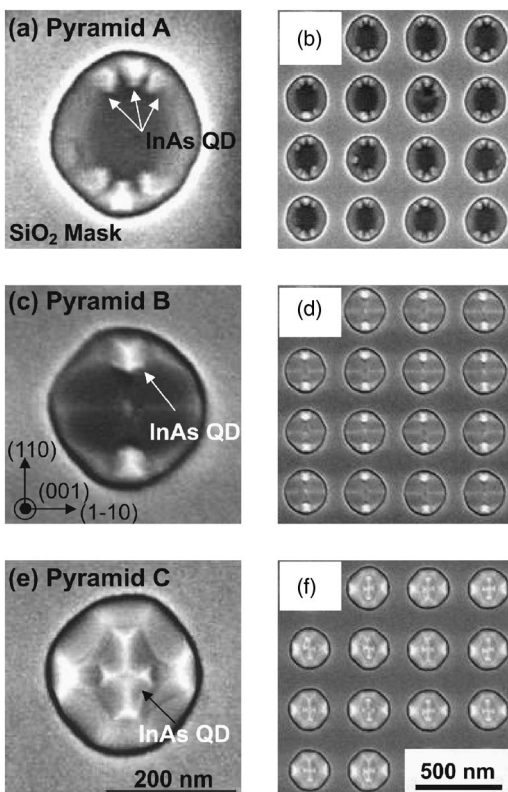


FIG. 3. Top-view SEM images of surface InAs PQDs on three types of GaAs pyramids.

show images of both a single pyramid and an array of pyramids. Figures 3(a), 3(c), and 3(e) are higher resolution images that feature only one single pyramid and elucidate PQD faceting. The PQDs, indicated by arrows, appear as lighter features in contrast to the surrounding GaAs pyramid surface. Figures 3(b), 3(d), and 3(f) are lower resolution, higher contrast images that indicate the statistical distribution of the preferential nucleation.

Figures 3(a) and 3(b) show preferential PQD nucleation on the $(11\bar{5})$ and $(\bar{1}15)$ facets of a single pyramid and on a pyramid array, respectively. Figure 3(b) indicates that an average of three QDs is formed on each $(11\bar{5})$ facet, though two to four QDs per facet are observed. No PQD nucleation is noted on the (001) apex, and rarely on $\{10\bar{5}\}$ planes. These QDs appear with an arrowheadlike tapered shape bound by the $\{10\bar{1}\}$ and $\{11\bar{1}\}$ planes with an approximate base dimension of 40×30 nm². On pyramid B, Figs. 3(c) and 3(d), two PQDs per pyramid are visible, one QD on each of the $(11\bar{3})$ and $(\bar{1}13)$ facets rather than surrounding $\{11\bar{5}\}$, $\{10n\}$, or (001) facets. Figure 3(d) shows two QDs per pyramid across the array indicating the strong preferential nucleation on the $(11\bar{3})$ and $(\bar{1}13)$ planes. Each PQD, bound by $\{11\bar{1}\}$ facets, has a base dimension of 40×40 nm². This shape is very similar to the kitelike InAs QDs that have been observed by Jacobi *et al.*¹³ Figures 3(e) and 3(f) show one highly faceted PQD formed on the apex of each pyramid C, which consists of $\{11\bar{3}\}$ and (001) planes. These PQDs are bound by $\{11\bar{1}\}$ and $\{10\bar{1}\}$ facets, with a small (001) apex and a base dimension of about 80×120 nm².

The trend in Fig. 3 indicates that the InAs PQD nucleation shows a strong affinity for the higher index GaAs facets such as $\{11\bar{5}\}$ and $\{11\bar{3}\}$ and avoids nucleation on the (001) , $\{11\bar{1}\}$, and the $\{10n\}$ surfaces. The only instance of InAs growth on a non- $\{11n\}$ facet is in Figs. 3(e) and 3(f) where the QD nucleations is on both the $\{11\bar{3}\}$ and (001) facets. This is believed to be a result of the smaller InAs QDs nucleating on the preferred $\{11\bar{3}\}$ surface and then coalescing over the (001) apex. This selectivity on the nucleation sites can be explained by the minimization of the QD energy on the ECS pyramid facets.

For SK QD formation on a planar growth surface, the QD assumes a shape associated with the lowest possible free energy. The net energy of the QD has been defined by Moll *et al.*¹⁴ as

$$E_{\text{QD}} = E_{\text{elastic}} + E_{\text{surface}} + E_{\text{edge}},$$

where E_{elastic} is the elastic energy relief due to partial strain relaxation inside the QD, E_{surface} is the surface energy associated with increased surface area of the QD, and E_{edge} is the energy associated with the various facets and the resulting edges of the QD. The growth of the InAs QDs on the GaAs ECS pyramids, however, involves adatom deposition atop the entire multifaceted pyramid surface. With the (001) , $\{11n\}$, and $\{10n\}$ planes existing side by side, the InAs QD nucleates on the facet for which the E_{QD} has the smallest value. While a variety of factors such as facet surface energy, surface kinetics, and strain contribute to the total QD energy, the dominant parameter is believed to be strain related. The GaAs $\{11n\}$ facets provide an optimal strain environment compared to other planes through optimal facet surface area, strain relief at the facet edge or perhaps growth plane tilt.¹⁵

Ongoing research including HR transmission electron microscopy will elucidate this trend.

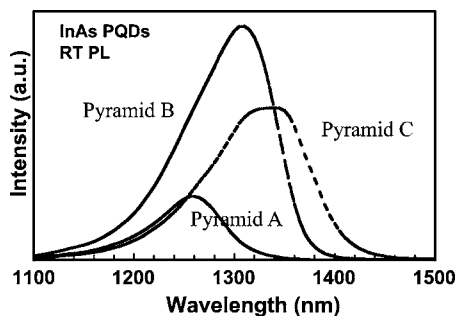


FIG. 4. RTPL spectra measured for PQRs formed on the three types of GaAs pyramids.

Figure 4 shows the RTPL spectra of capped InAs PQRs similar to Figs. 3(a)–3(f). The ground state emission wavelength ranges from 1.25 to 1.35 μm for PQRs on pyramids A–C, respectively. The wavelength is consistently longer for the larger PQRs formed on pyramid C compared to pyramid A or B. The full widths at half maximum of the spectra are 59, 72, and 79 meV, respectively. The inhomogeneous broadening comes from the variation of the pattern size/shape, the size of the QD-forming GaAs facets, and the resulting PQR size/shape. Low-temperature PL spectra (77 K, data not shown) indicate three-dimensional quantization through the saturation of the ground state and observed excited states of PQRs on all three kinds of pyramids. Separation between intraband energy levels ranges from 42 to 64 meV.

In conclusion, we have demonstrated selective InAs PQR nucleation on GaAs ECS pyramid facets. The PQR

nucleation preferentially occurs on the $\{11n\}$ family rather than $\{10n\}$ family or (001). This phenomenon is likely due to minimized total QD energy dominated by strain effects. This demonstration represents initial steps towards improved understanding of nanostructure formation on nonplanar, nano-defined surfaces.

- ¹H. Lee, J. A. Johnson, M. Y. He, J. S. Speck, and P. M. Petroff, *Appl. Phys. Lett.* **78**, 105 (2001).
- ²T. van Lippen, R. Notzel, G. J. Harnhuis, and J. H. Wolter, *J. Appl. Phys.* **97**, 044301 (2005).
- ³Rudeesun Songmuang, Suwit Kiravittaya, and Oliver G. Schmidt, *Appl. Phys. Lett.* **82**, 2892 (2003).
- ⁴M. H. Baier, S. Watanabe, E. Pelucchi, and E. Kapon, *Appl. Phys. Lett.* **84**, 1943 (2004).
- ⁵M. H. Baier, C. Constantin, E. Pelucchi, and E. Kapon, *Appl. Phys. Lett.* **84**, 1967 (2004).
- ⁶V. C. Elarde, R. Rangarajan, J. J. Borchardt, and J. J. Coleman, *IEEE Photonics Technol. Lett.* **17**, 935 (2005).
- ⁷S. Birudavolu, N. Nuntawong, G. Balakrishnan, Y. C. Xin, S. Huang, S. C. Lee, S. R. J. Brueck, C. P. Hains, and D. L. Huffaker, *Appl. Phys. Lett.* **85**, 2337 (2004).
- ⁸D. L. Huffaker, C. P. Hains, N. Nuntawong, Y. C. Xin, P. S. Wong, L. Xue, S. R. J. Brueck, and L. Lester, *J. Appl. Phys.* **99**, 33503 (2006).
- ⁹T. Umeda, K. Kumakura, J. Motohisa, and T. Fukui, *Physica E (Amsterdam)* **2**, 714 (1998).
- ¹⁰H. An and J. Motohisa, *Appl. Phys. Lett.* **77**, 385 (2000).
- ¹¹S. C. Lee, K. J. Malloy, and S. R. J. Brueck, *J. Appl. Phys.* **90**, 4163 (2001).
- ¹²G. Balakrishnan, R. Molecke, P. S. Wong, N. Nuntawong, and D. L. Huffaker (unpublished).
- ¹³K. Jacobi, *Prog. Surf. Sci.* **71**, 185 (2003).
- ¹⁴N. Moll, M. Scheffler, and E. Pehlke, *Phys. Rev. B* **58**, 4566 (1998).
- ¹⁵B. W. Dodson, D. R. Myers, A. K. Datye, V. S. Kaushik, and B. Martinez-Tovar, *Phys. Rev. Lett.* **61**, 2681 (1988).