# Photoluminescence linewidths from multiple layers of laterally self-ordered InGaAs quantum dots

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Laterally ordered multilayered arrays of InGaAs quantum dots are investigated by photoluminescence as a function of high index GaAs substrates. Different laser wavelengths are used to investigate the photoluminescence from quantum dots layer-by-layer. High optical quality is demonstrated for laterally ordered quantum dot arrays. GaAs(511)B is identified as the optimum high index substrate for growth of InGaAs/GaAs multilayered quantum dots, demonstrating strong photoluminescence with a narrow full width at half maximum linewidth of 23 meV in spite of the potential for misfit dislocations. © *2005 American Institute of Physics*. [DOI: 10.1063/1.2131198]

Devices based on semiconductor quantum dots (QDs) are under development to utilize the unique optical and electronic properties that results from three-dimensional (3D) quantum confinement. In many cases, ordered QD arrays are part of the design of such devices. As a result, there has been a growing interest in lateral control of the QD position as evidenced by the report of a variety of clever organizational techniques developed most recently.[1–7] One interesting example among them is the observation of *laterally self-ordering* during the growth of InGaAs/GaAs QD multilayers.[4,5] Remarkably, the ordered pattern can be adjusted by selecting different high index substrates. For example, one-dimensional (1D) ordering, (QD chains up to several microns in length), has been achieved on GaAs(100) (Ref. 4) while two-dimensional (2D) ordering (a QD checkerboard) was observed on GaAs high index surfaces.[5] Although there have been many experimental and theoretical studies on the behaviors of QDs on high index GaAs surfaces,[5,8–11] the choice of the optimum indexed substrate for QD ordering, size uniformity, and importantly, the corresponding optical properties of the QDs, still remains an interesting question. Indeed, since the formation of InGaAs QDs on GaAs surfaces is strain driven, multiple QD layers may introduce misfit defects which deteriorate their optical properties. For this reason, a systematical investigation of the optical behavior of ordered QD structures as a function of high index can be of value for optimum size uniformity and optical performance of QD multilayers. In this letter, we demonstrate strong photoluminescence (PL) emission with a narrow linewidth from multiple QD layers having a high degree of lateral ordering. In particular, we systematically demonstrate GaAs(511)B to be the optimum growth substrate orientation for the growth of lateral ordered, high optical quality, InGaAs QD multilayers.

Samples for this study were grown by molecular-beam epitaxy (MBE) on GaAs(100) and GaAs($n$11)B (where $n$ is 9, 7, 5, 4, and 3). Details of the MBE system and the growth conditions have been reported elsewhere.[4,5] All of the substrates with different indexes were indium soldered side by side on a molybdenum block. The block was rotated during growth to ensure a uniform distribution of materials. The grown layered structure is shown in Fig. 1. The thickness of the GaAs spacer between QD layers is 120 monolayers, nearly double that used in previous reports.[5] This thickness was intentionally selected to insure that vertical tunneling of carriers between InGaAs QD layers is negligible[12] and that we are able to study the optical properties of a QD multilayered stack as optically independent layers.

Figure 2 shows atomic force microscopy (AFM) images of the InGaAs QDs in the last exposed layer. As expected, "long chains" of QDs and QD "checkerboards" are observed on GaAs(100) and GaAs high index surfaces. The autocorrelation plot, which are insets in Figs. 2(a) and 2(d), give quantitative measure to the nature of the 1D and 2D lateral ordering. For example, the 2D ordering achieved on high index surfaces in this experiment is much better than noted in previous reports.[5]

The PL measurement was performed at a temperature of 10 K using the 325 nm line of an HeCd laser, 514.5 nm line of an $Ar^+$ laser, 632 nm line of a HeNe laser, and 797 nm line of a Ti-sapphire laser, as the excitation wavelengths. By using different excitation wavelengths, the optical beam penetrated into the sample at different corresponding depths due

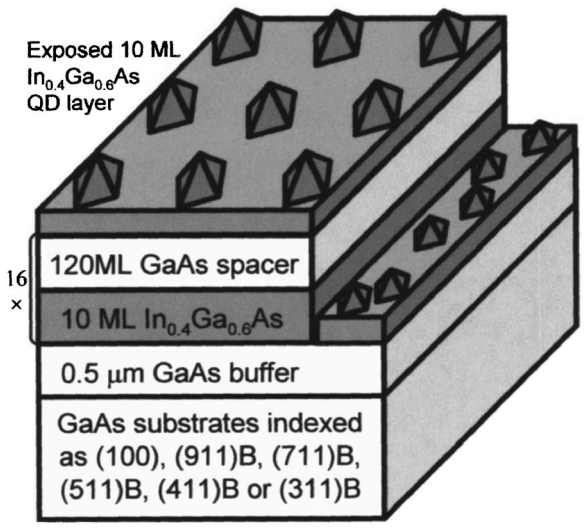


FIG. 1. Schematic of the grown layer sequence. The difference between QDs in the first layer and in the last layer is illustrated.

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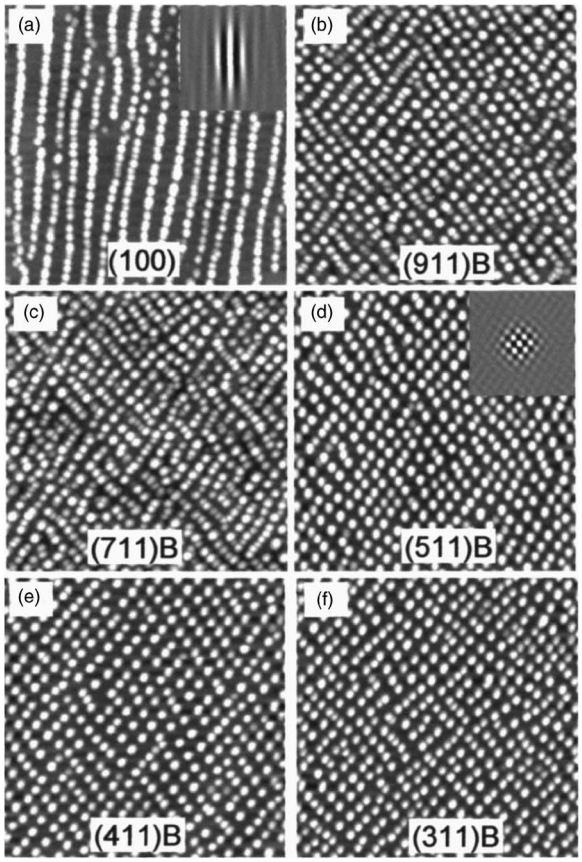


FIG. 2. AFM images (2 $\mu$m$\times$2 $\mu$m) of the exposed QD layers grown on (a) GaAs(100), (b) GaAs(911)B, (c) GaAs(711)B, (d) GaAs(511)B, (e) GaAs(411)B, and (f) GaAs(311)B. The insets in (a) and (d) show their corresponding autocorrelation.

to a change in the absorption coefficient with wavelength. It was assumed that the penetration depth $d$ of excitation light was $\sim 2/\alpha$, where $\alpha$ is the absorption coefficient. Given the strong spectral dependence of $\alpha$,[13] by changing the excitation wavelength, we vary the number of QD layers exposed to the laser light. For example, using the 325 nm excitation wavelength of the HeCd laser we only generated carriers in the top buried QD layer. Meanwhile, by using the 514.5 nm wavelength of the $Ar^+$ laser, approximately the top six buried layers of QDs were excited, etc. (see Fig. 3).

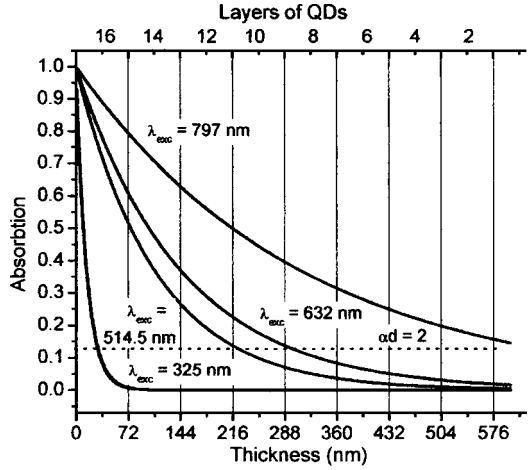


FIG. 3. Absorption through multiple layers of QDs as a function of thickness for different laser wavelengths.

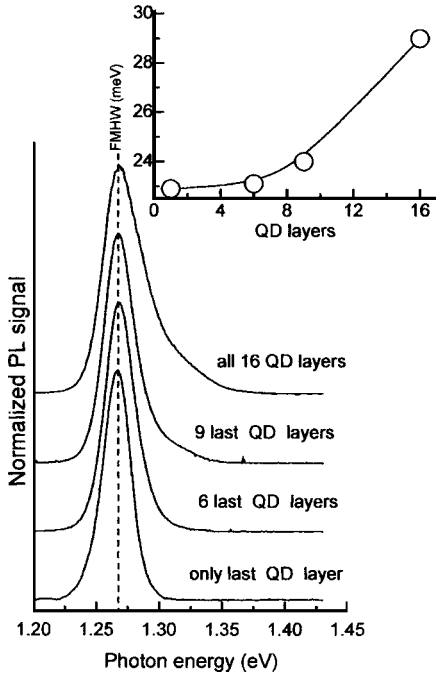


FIG. 4. PL spectra from quantum dots grown on GaAs(511)B excited by different laser wavelengths to access different layers of QDs. The inset shows the PL linewidth broadening with more QD layers excited.

Figure 4 shows the PL spectra from InGaAs QDs grown on GaAs(511)B with four different laser excitation wavelengths. With more layers of QDs excited, the PL spectra broadened toward the high-energy side. The inset shows the full width at half maximum of the spectra as a function of the number of QD layers probed. The main cause of the linewidth broadening is a high-energy tail originating from several of the bottom layers of QDs. The structural origin of the high-energy tail is illustrated in Fig. 1 by the size difference between QDs in the bottom buried layer and the top exposed layer. The QD size of the bottom layer is relatively small. Its strain-relaxation transmitted through the GaAs spacer induces the earlier formation of QDs in the second layer. Therefore, the InGaAs wetting layer is thinner and more InGaAs material contributes to the formation of QDs even though the total InGaAs deposited is same. In addition, the transmitted strain field from the first layer of QDs has many overlapping functions (for closely spaced QDs in the bottom layer) and consequently results in a lower density of QDs in the second deposited layer. In summary, the development of lateral ordering is accompanied by a gradual increase in QD size during the first several QD layers of growth until a stable size of QDs with lateral ordering is reached after several QD layers (8–10 QD layers).[4,5,14] The high-energy tail of PL is from the smaller QDs in the size transition region. After 16 buried layers of QDs, the linewidth of the PL reaches 29 meV, which is still narrower than the typical value reported for InGaAs QDs.[15]

The PL spectra from our samples, excited by the laser wavelength of 325 nm, are shown in Fig. 5. All samples investigated show high quality optical behavior. The PL linewidth is shown in Fig. 6 as a function of the geometrical angle between the crystallographic plane (100) and the surface of the substrate. The PL linewidth is 37 meV for the dotchains on GaAs(100) and increases slightly to 42 meV for the QDs on GaAs(911)B. Short QD chains are often ob-

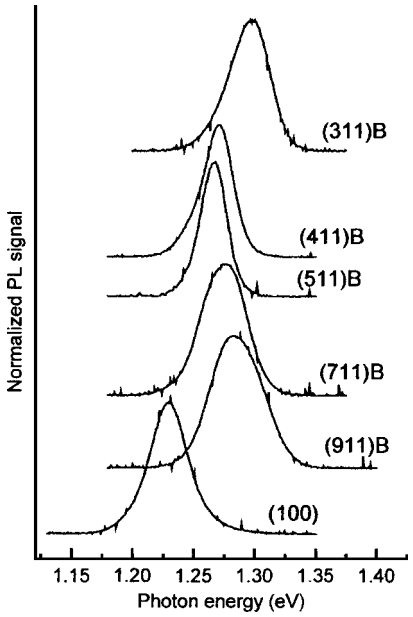


FIG. 5. Comparison of PL spectra from quantum dots grown on different substrates.

served mixing in the QD checkerboard on GaAs(911)B as shown in Fig. 2(b). As the geometrical angle is further increased, the PL linewidth first reduces and then increases. The linewidth minimizes at 23 meV for the 2D organized QDs on GaAs(511)B. Although GaAs(311)B has been reported to be a better choice than GaAs(100) as a substrate for the growth of InGaAs QDs,[16] our investigation demonstrates the GaAs(511)B is the optimum surface orientation for multiple layers of InGaAs/GaAs QDs.

Clearly, we observe a strong correlation between the PL linewidth and the QD lateral ordering. The greater the 2D ordering, the narrower the PL linewidth. For example, as we vary the index of the substrate, 2D ordering peaks for GaAs(511)B while the PL linewidth minimizes. Physically, this correlation is expected. Tersoff *et al.* proposed a simple model to explain the vertical and lateral ordering observed in the growth of multiple layers of QDs.[17] When two islands in a lower QD layer are close to each other they give rise to a single minimum in the strain field in the QD layer just above. Thus from layer-to-layer, island sizes eventually reach a "stable size" beyond which they no longer merge with one another. The assumption here, however, is that there is a stain field in all lateral directions that leads to a stable island size in all directions. In fact, this is not the case in our study. For example, for GaAs(001) or GaAs(911)B, high diffusion along the dot chain direction (dimer row direction) acts to eliminate the strain field and prevent a stable size from developing along that direction in layer-to-layer growth. On the other hand, by increasing the step density along the dot-chain direction, growth on a GaAs(511)B substrate experiences uniformly low lateral diffusion, both along the dot-chain direction and perpendicular to it. Consequently a more uniform stable lateral QD size can develop in both directions. Given a typically fixed aspect ratio or shape, a more uniform lateral size will in-turn yield a more uniform vertical size. Since lateral ordering will result in a more uniform stable size distribution, we may expect the observed correlation between lateral ordering and PL linewidth.

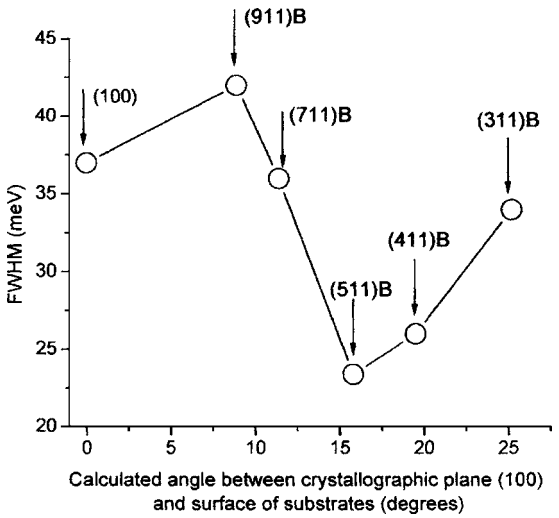


FIG. 6. Change in PL linewidth as a function of the calculated angle between crystallographic plane (100) and surface of substrates.

In conclusion, lateral self-ordering of QDs resulting from the growth of InGaAs/GaAs multiple QD layers is investigated by PL measurements. The achievement of a high degree of lateral ordering is observed to be accompanied by strong PL emission and a narrow linewidth in spite of the potential for misfit dislocations. The GaAs substrate index for multilayering InGaAs QDs is optimized for (511)B. A photoluminescence linewidth as narrow as 23 meV is observed from the QDs grown on GaAs(511)B with optimum 2D lateral ordering. As a result, lateral self-ordered QD arrays with high optical quality have an excellent opportunity to find application as detector and emitter arrays.

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