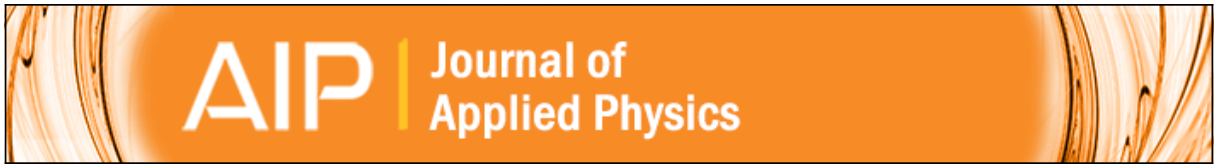


Dislocation-induced spatial ordering of InAs quantum dots: Effects on optical properties

R. Leon, S. Chaparro, S. R. Johnson, C. Navarro, X. Jin, Y. H. Zhang, J. Siegert, S. Marcinkevičius, X. Z. Liao, and J. Zou

Citation: Journal of Applied Physics **91**, 5826 (2002); doi: 10.1063/1.1467963
View online: http://dx.doi.org/10.1063/1.1467963
View Table of Contents: http://scitation.aip.org/content/aip/journal/jap/91/9?ver=pdfcov
Published by the AIP Publishing

Articles you may be interested in

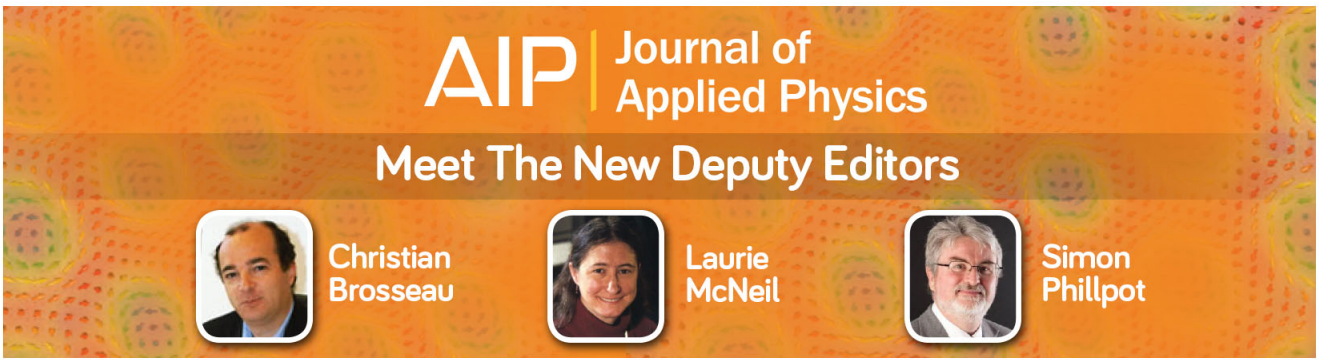
In islands and their conversion to InAs quantum dots on GaAs (100): Structural and optical properties
J. Appl. Phys. **107**, 014312 (2010); 10.1063/1.3269700

Improved optical properties of InAs quantum dots grown with an As 2 source using molecular beam epitaxy
J. Appl. Phys. **100**, 063107 (2006); 10.1063/1.2352809

Planar InAs growth on GaAs(001) and subsequent quantum dot formation by a surface induced morphological instability
J. Vac. Sci. Technol. B **22**, 2261 (2004); 10.1116/1.1771671

Anomalous temperature dependence of photoluminescence from InAs quantum dots
J. Appl. Phys. **88**, 2529 (2000); 10.1063/1.1288231

Shape transition of InAs quantum dots by growth at high temperature
Appl. Phys. Lett. **74**, 1224 (1999); 10.1063/1.123506



[This article is copyrighted as indicated in the article. Reuse of AIP content is subject to the terms at: http://scitation.aip.org/termsconditions. Downloaded to] IP: 209.147.144.21 On: Mon, 09 Feb 2015 22:28:37

Dislocation-induced spatial ordering of InAs quantum dots: Effects on optical properties

R. Leon
Jet Propulsion Laboratory, California Institute of Technology, Pasadena, California 91109

S. Chaparro, S. R. Johnson, C. Navarro, X. Jin, and Y. H. Zhang
Center for Solid State Electronics Research and Department of Electrical Engineering, Arizona State University, Tempe, Arizona 85287-6206

J. Siegert and S. Marcinkevičius
Department of Microelectronics and Information Technology, Section Optics, Royal Institute of Technology, 100 44 Stockholm, Sweden

X. Z. Liao and J. Zou
Australian Key Centre for Microscopy and Microanalysis, The University of Sydney, Sydney, New South Wales 2006, Australia

(Received 8 October 2001; accepted for publication 13 February 2002)

Misfit dislocations were used to modify the surface morphology and to attain spatial ordering of quantum dots (QDs) by molecular beam epitaxy. Effects of anneal time and temperature on strain-relaxed $In_xGa_{1-x}As/GaAs$ layers and subsequent spatial ordering of InAs QDs were investigated. Photoluminescence (PL) and time-resolved PL was used to study the effects of increased QD positional ordering, increased QD uniformity, and their proximity to dislocation arrays on their optical properties. Narrower inhomogeneous PL broadening from the QDs ordered on dislocation arrays were observed, and differences in PL dynamics were found. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1467963]

INTRODUCTION

Quantum dots (QDs) have been the focus of extensive research due to their electronic and optical properties. Also, their radiation hardness makes them ideal for devices used in space applications.[1,2] Several emerging device applications, such as lasers, detectors, and memories, using QDs have encouraged these efforts.[3–5] During the self-assembled growth on planar substrates, quantum dots arrange themselves in a random manner. While this is acceptable for some QD applications, control of the position and ordered QD configurations will enable other types of applications and offer new functions for QD devices, like new highly parallel computing architectures[6] and single electron transistors.[7]

Recent research on the self-organization of nanostructures has shown that ordering can be achieved through several mechanisms. Step alignment in bunched steps has produced long-range step alignment in InAs and InGaAs QDs.[8,9] Such step bunching-induced alignment has different effects in Ge/Si dots and results in increased lateral ordering[10] without the formation of strings of island on the step edges, due to the elastic repulsion between islands found in this system.[11] In contrast to the Ge/Si and InAs/GaAs systems where strain induced ordering is limited by their small elastic anisotropy,[12] very good long-range order has been achieved in lead salt crystals, in which strong elastic anisotropy leads to the formation of three-dimensional dot lattices, and the dot positioning shows anticorrelation with the underlying dot layers.[13] Good vertical alignment has been shown in InAs and InP QDs, in this case a positive correlation aligns the dots directly over existing islands when the capping layer is sufficiently thin to maintain a large enough residual strain field on the growth surface.

The interplay between surface morphology and misfit dislocations can also help in the spatial ordering of QDs.[14] Here we report a technique that takes advantage of the introduction of misfit dislocations to form highly ordered patterns of isolated InAs QDs on strain-relaxed $In_xGa_{1-x}As$ layers grown on GaAs substrates. One of the ways to form highly ordered patterns of QDs is to produce them on misfit dislocations deliberately introduced into the structure. During such growth, a rectangular pattern of misfit dislocations is transferred into well separated rows of sharply aligned quantum dots. However, dislocations may also have a negative effect and act as nonradiative recombination centers, thus reducing carrier lifetimes. In the present work we examine ordered quantum dot structures by studying the effects of growth conditions, surface morphology, and surface strain on the growth of three-dimensional (3D) InAs islands, i.e., QDs. Time-resolved photoluminescence (PL) was then carried out to study how the alignment and the introduction of dislocations affect the optical properties and carrier dynamics in these QDs.

EXPERIMENTAL DETAILS

The QD structures were grown by molecular beam epitaxy (MBE) on epi-ready n^+ GaAs $(100)\pm0.1°$ and semi-insulating GaAs $(100)\pm0.1°$ substrates that were rotated during deposition. Figure 1 shows a cross section of the structure grown. The basic aim is to grow a metastable struc-

 © 2002 American Institute of Physics
[This article is copyrighted as indicated in the article. Reuse of AIP content is subject to the terms at: http://scitation.aip.org/termsconditions. Downloaded to] IP: 209.147.144.21 On: Mon, 09 Feb 2015 22:28:37

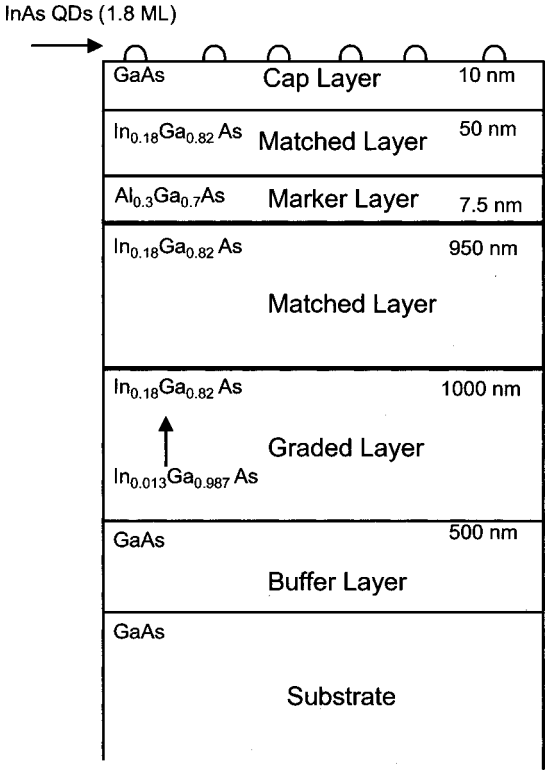


FIG. 1. Cross-sectional representation of the structure of a typical sample. All structures were annealed prior to InAs QD growth.

ture which relaxes during postgrowth annealing through a self-organized process and leads to the formation of gradual surface undulations [also known as cross-hatched patterns (CHPs)], which act as preferential sites for the nucleation of QDs. First a 500 nm buffer layer was deposited at 580 °C. The substrate temperature was then reduced to 510 °C and a 1000 nm compositionally graded $In_xGa_{1-x}As$ layer ($0.013 \leqslant x \leqslant 0.18$) was grown, followed by the deposition of a 950 nm $In_{0.18}Ga_{0.82}As$ layer, a 7.5 nm $Al_{0.3}Ga_{0.7}As$ marker layer, and another 50 nm of $In_{0.18}Ga_{0.82}As$. Finally, a 10 nm GaAs cap layer was deposited. From this point on the various samples were treated differently. The structures grown on n^+ GaAs were annealed, before QDs growth, at 620 °C for 15, 45, 60, 80, and 95 min, respectively. The structure grown on semi-insulating GaAs was annealed for 60 min at 650 °C. After annealing, the QDs were formed by depositing 1.8 monolayers (ML) of InAs at a growth temperature of 525 °C. GaAs caps of 20 nm were deposited on all samples used in the optical measurements. Control InAs QDs were grown on top of a dislocation-free GaAs layer that was used to compare the QD surface morphology and PL spectroscopy results.

Structural information was obtained from atomic force microscopy (AFM) (in contact mode) and transmission electron microscopy (TEM), both in plan view and in cross section. These show that the QD surface densities diminish when QDs are formed on a strain-relaxed $In_xGa_{1-x}As$ layer: the QD surface densities are only $3-4\times10^9/cm^2$ in the ordered structures and $3-4\times10^{10}/cm^2$ in the control or defect-free structures.

Time-resolved PL was carried out at 80 K after excitation at 800 nm by a short laser pulse from a self mode-locking Ti:saphire laser (pulse duration 80 fs, repetition frequency 95 MHz). A synchroscan streak camera with an infrared enhanced photocathode, combined with a 0.25 m spectrometer (temporal resolution 3 ps), was used for PL detection. The average excitation power of 2 mW used in the experiments corresponds to about 4×10^{11} electron-hole pairs per cm^2 per pulse.

RESULTS AND DISCUSSIONS

Structural analysis shows several interesting features in the resulting morphology. Lines of ordered QDs appear along [011] and [0-11]. The higher density of QDs nucleating along [011] and [0-11] also creates large areas that are denuded of QDs in between rows of aligned QDs. The surfaces of the samples are no longer flat but instead exhibit undulations parallel to the ⟨011⟩ directions. It was also observed for longer anneal times and/or higher anneal temperatures that the thin dislocation slip lines where the QDs align evolve into wider and deeper trenches. The annealing process provides enough thermal energy to cause the layers to relax by creating misfit dislocations and thus CHPs in the $In_xGa_{1-x}As$ layer, which is metastable due to its gradual change in composition. As the anneal proceeds, dislocations glide to the top surface of the structure where favorable nucleation sites are formed, resulting in the observed InAs QD alignment. Figure 2 shows that the InAs QDs form highly ordered lines along ⟨011⟩ for a sample that was annealed for 60 min at 620 °C prior to the deposition of InAs. As was predicted by Samonji *et al.*[15] the surface of the sample has undulations that are parallel to [0-11] as well as less noticeable ones that are parallel to [011]. It has been reported that, for InGaAs/GaAs heteroepitaxy, the incorporation rate of group III atoms to the *B*-type step, parallel to [011], is higher than that to the *A*-type step, parallel to [0-11].[15] This asymmetry in adatom diffusion, implying that the surface diffusion length along [011] is longer than that along the [0-11] direction, could explain the undulations observed. These undulations provide nucleation sites for the QDs, thus facilitating alignment of the QDs.

To study the wider dislocation areas (trenches) in more detail, smaller scan images of these regions were taken. AFM analysis showed that, as the anneal time increases, the dislocation lines (where the QDs form) develop into wide trenches were the QDs agglomerate. AFM also showed the QDs to be very uniform. Figures 3(a)–3(d) show height mode images, where the brightest areas represent the tallest features and the darker areas represent lower features, of a 1 μm region of the sample annealed at 650 °C for 60 min. The QDs are seen to nucleate along both side walls and the bottom of the trenches. This can be clearly seen in Fig. 3(b), which is a 3D representation of Fig. 3(a). TEM also shows QD nucleation in trenches, seen in Fig. 3(e). TEM examination did not show dislocations within the quantum dots. Figures 3(c) and 3(d) are cross-sectional views of Fig. 3(a) along [011] and [0-11], respectively. Here the surface undulations parallel to the [0-11] direction are seen to be more

[This article is copyrighted as indicated in the article. Reuse of AIP content is subject to the terms at: http://scitation.aip.org/termsconditions. Downloaded to] IP: 209.147.144.21 On: Mon, 09 Feb 2015 22:28:37

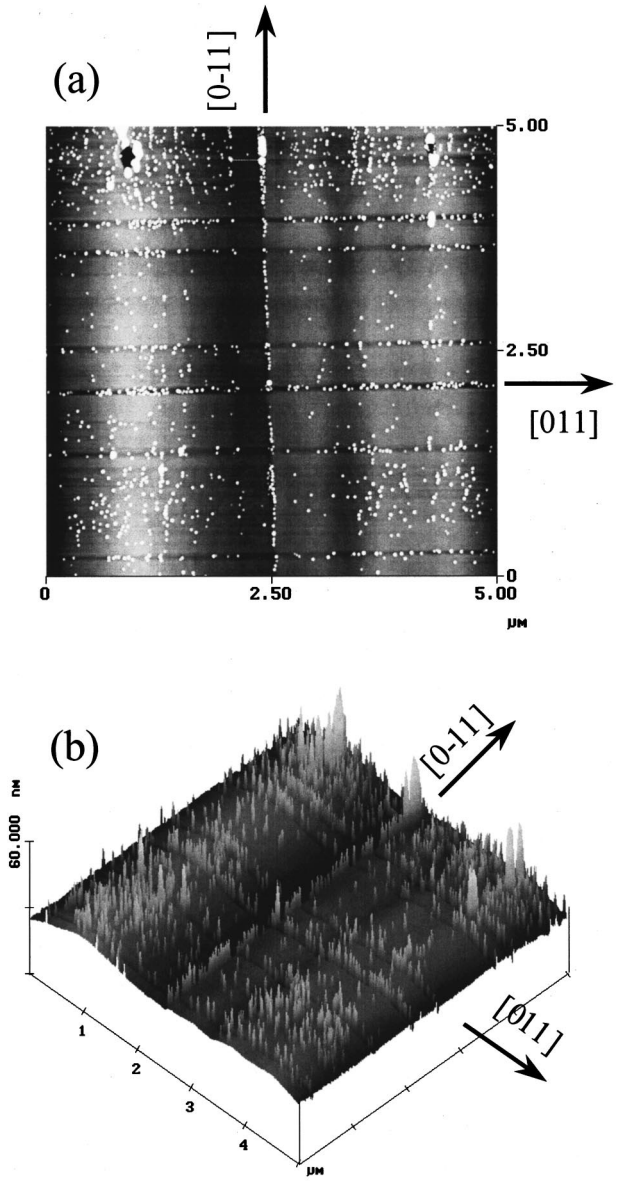


FIG. 2. (a) 5 μm AFM image of a sample that was annealed for 60 min at 620 °C prior to the deposition of InAs. The InAs islands (QDs) align along ⟨011⟩ and large denuded areas start to appear. (b) 3D representation of (a); here undulations are seen to form more prominently along [110].

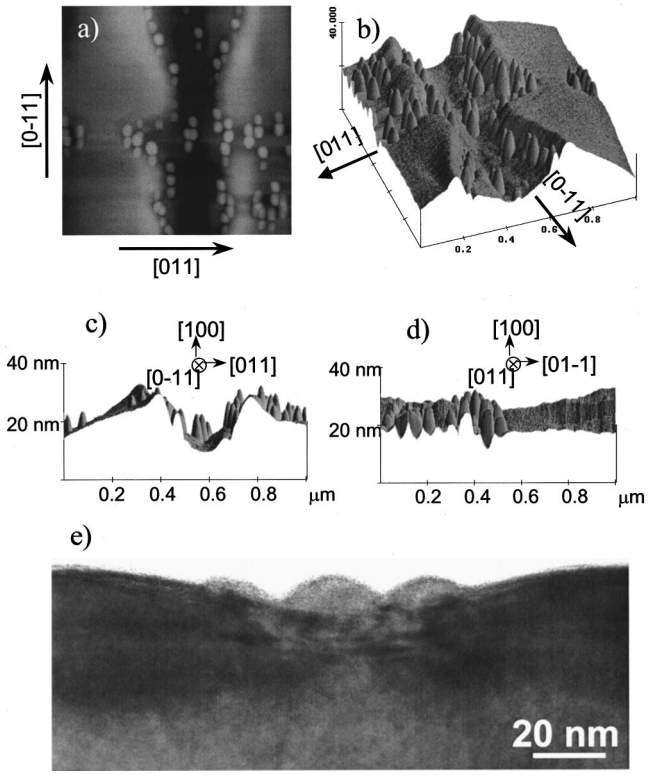


FIG. 3. (a) Top view and (b) 3D representation of a region where the dislocation slip line has become a wide trench in a sample grown on a semi-insulating GaAs (100) wafer and annealed for 60 min at 650 °C. (c), (d) View directly into the ⟨011⟩ directions where it can be seen that the trenches open along [0−11] and QDs do not show a preference of where in the trench the will nucleate. (e) TEM cross-sectional image of InAs QDs on a trench formed as a result of dislocation slip plane step bunching.

prominent. Longer anneal times and higher anneal temperatures mean increased diffusion kinetics, which allow the samples to achieve a more energetically favorable configuration. In the Si/Ge system it is known that atoms diffuse away from the highest strain regions and create trenches.[16]

In dislocation-induced ordering like that in this work and in Ge/Si quantum dots[14] the alignment observed is difficult to explain by the strain field from dislocations, since the dislocation core is far from the dot nucleation sites and the strain field of dislocations decreases abruptly with distance from their core.[17] Here alignment could be a result of preferential nucleation on the shear steps that form on the surface. The cross-hatched patterns have been explained by inhomogeneous strain fields associated with the underlying dislocations which lead to spatially nonuniform growth rates at the growth front. Since both of these are present, slip steps and anisotropic residual strain on the surface, it is difficult to separate the effects of these two mechanisms on QD nucleation.

Room temperature continuous wavelength (cw) and low temperature time-resolved PL were observed from QD states in some of these structures. Figure 4(a) shows PL spectra from two different samples in which the InAs QDs were grown on strain-relaxed InGaAs layers. This is compared to PL spectra obtained from InAs QDs grown on defect-free GaAs. The PL spectra from the ordered InAs QDs grown on the dislocation arrays are narrower that the one obtained from the ''control'' InAs QDs (grown on defect-free GaAs). The 80 K QD PL spectra show a single peak centered at 1.22 eV with a full width at half maximum (FWHM) of 36 meV for the sample with dislocations compared to 1.19 eV and 128 meV for the reference sample. The reference sample has large inhomogeneous broadening since the growth was not optimized to obtain good size uniformity. High QD surface densities also contribute to increased inhomogeneous broadening. A narrower PL peak of the ordered QD sample suggests smaller QD size variations and perhaps reduced ensemble interactions with neighboring QDs. This observation agrees with the better uniformity of the QDs on the arrays observed by AFM imaging, and also with the observed ''sparse'' regions without QDs, which would imply fewer differences from an altered strain environment due to effects from neighboring QDs.[18] The PL peak from the disordered

[This article is copyrighted as indicated in the article. Reuse of AIP content is subject to the terms at: http://scitation.aip.org/termsconditions. Downloaded to] IP: 209.147.144.21 On: Mon, 09 Feb 2015 22:28:37

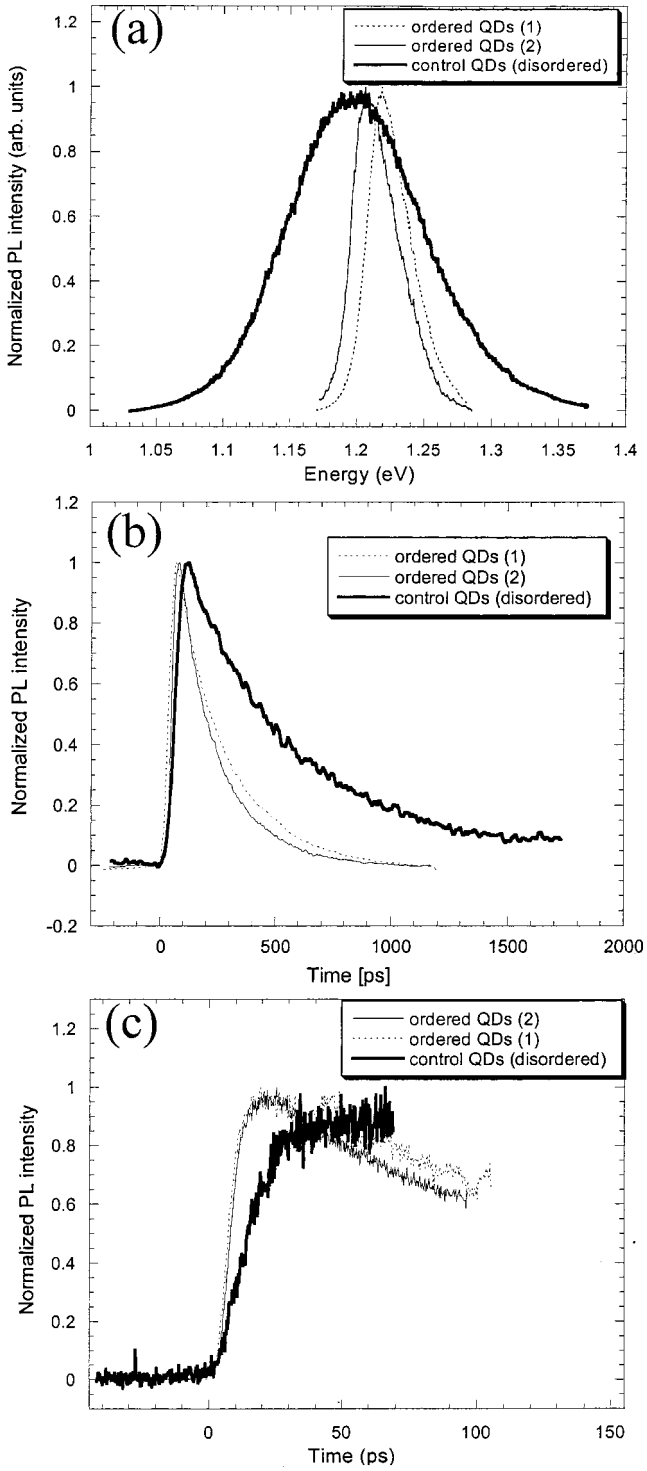


FIG. 4. Photoluminescence results for ordered InAs QDs grown on strain-relaxed InGaAs layers and annealed under different conditions [(1) 15 min at 620 °C and (2) 60 min at 650 °C] compared to a spectrum obtained from InAs QDs grown under similar conditions (525 °C and 1.8 ML of InAs) but on dislocation-free GaAs. (a) Low temperature (80 K) PL spectra of InAs QD structures formed on dislocation arrays and the control InAs randomly formed on defect-free GaAs buffer layers show much less inhomogeneous broadening for the two ordered QD structures; time-resolved PL transients measured on (b) longer and (c) shorter time scales that demonstrate full PL transients and PL rise times for QDs formed on dislocations and for QDs formed on a planar, defect-free GaAs buffer layer.

(control) QD ensemble is broader than average, but since the growth conditions were not changed during growth of the QDs other than deposition on strain-relaxed layers, we believe that this reduction in inhomogeneous PL broadening would also occur with optimized control samples.

Figures 4(b) and 4(c) show differences in PL transients from these different types of QD structures measured on long and short time scales. From these transients, the PL rise and decay times are extracted. The 80 K carrier lifetimes are 170 and 460 ps for the ordered and disordered QD samples, respectively. In both cases the carrier lifetimes are too short to be determined solely by radiative recombination. This observation implies the presence of defects in the dots or at their interfaces. The 80 K PL rise times are 5 ps for the ordered QDs and 14 ps for the control InAs QDs. A probable reason for the reduced PL rise time is reduced carrier transport time because of reduced transport distance. For above band gap excitation, the PL rise time accounts for carrier transport, capture, and relaxation. The absorption depth for the exciting radiation in GaAs is about 0.5 μm, and it is from this depth that the carriers are collected into the dots in the reference sample with uniform and defect-free GaAs barriers. In the ordered QD samples, carriers are collected basically from the 20 nm thick GaAs cap layer, because the 10 nm thick GaAs layer, on which the QDs are grown, acts as a barrier for carriers generated in the InGaAs layers deeper in the structures. In addition, dislocations may act as concurrent trapping centers, further diminishing the volume from which carriers are collected into the QDs.

CONCLUSIONS

In conclusion, we have used AFM imaging and cross-sectional analysis combined with photoluminescence to study the effects of dislocations on the morphology, spatial ordering, and optical properties of InAs QDs on strain-relaxed $In_xGa_{1-x}As$ layers grown by MBE. We observed that, by annealing the metastable $In_xGa_{1-x}As$/GaAs structure, the surfaces of the samples develop undulations along the $\langle 011 \rangle$ directions, which induce alignment of QDs. As this ordering develops, areas denuded of QDs begin to appear. By increasing the annealing time and/or temperature the dislocation lines widen and transform into trenches. QDs nucleation is concentrated in these trench regions with no apparent preference for the side walls or the bottom of the trenches. Narrower PL peaks were obtained from the ordered QD structures formed on strain relaxed $In_xGa_{1-x}As$ layers. Shorter PL rise and decay times were also observed at low temperatures. Shorter rise times are explained by decreased carrier transport times within the barriers surrounding the QDs.

ACKNOWLEDGMENTS

Part of this research was carried out at the Jet Propulsion Laboratory, California Institute of Technology, under a contract with the National Aeronautics and Space Administration. Financial support from the Swedish Foundation for International Cooperation in Research and Higher Education (STINT) is gratefully acknowledged.

[This article is copyrighted as indicated in the article. Reuse of AIP content is subject to the terms at: http://scitation.aip.org/termsconditions. Downloaded to] IP: 209.147.144.21 On: Mon, 09 Feb 2015 22:28:37

[1] P. G. Piva, R. D. Goldberg, I. V. Mitchell, D. Labrie, R. Leon, S. Charbonneau, Z. R. Wasilewski, and S. Fafard, Appl. Phys. Lett. **77**, 624 (2000).
[2] R. Leon, G. M. Swift, B. Magness, W. A. Taylor, Y. S. Tang, K. L. Wang, P. Dowd, and Y. H. Zhang, Appl. Phys. Lett. **76**, 2071 (2000).
[3] S. Fafard, K. Hinzer, S. Raymond, M. Dion, J. McCaffrey, Y. Feng, and S. Charbonneau, Science **274**, 1350 (1996).
[4] J. L. Jimenez, L. Fonseca, D. Brady, J. P. Leburton, D. E. Wohlert, and K. Y. Cheng, Appl. Phys. Lett. **71**, 3558 (1997); J. Phillips, K. Kamath, and P. Bhattacharya, *ibid.* **72**, 2020 (1998).
[5] S. Muto, Jpn. J. Appl. Phys., Part 2 **34**, L210 (1995).
[6] T. Lundstrom, W. Schoenfeld, H. Lee, and P. M. Petroff, Science **286**, 2312 (1999).
[7] I. Amlani, A. O. Orlov, G. L. Snider, C. S. Lent, and G. H. Bernstein, Appl. Phys. Lett. **72**, 2179 (1998).
[8] N. Ikoma and S. Ohkouchi, Jpn. J. Appl. Phys., Part 2 **34**, L724 (1995).
[9] R. Leon, T. J. Senden, Yong Kim, C. Jagadish, and A. Clark, Phys. Rev. Lett. **78**, 4942 (1997).
[10] J. Zhu, K. Brunner, and G. Abstreiter, Appl. Phys. Lett. **73**, 620 (1998).
[11] J. Floro, G. Lucadamo, E. Chason, L. Freund, M. Sinclair, R. Twesten, and R. Hwang, Phys. Rev. Lett. **80**, 4717 (1998).
[12] J. Stangl *et al.*, J. Vac. Sci. Technol. B **18**, 2187 (2000).
[13] V. Shchukin, D. Bimberg, V. Malyshkin, and N. Ledentsov, Phys. Rev. B **57**, 12262 (1998).
[14] S. Y. Shiryaev, F. Jensen, J. L. Hansen, J. W. Petersen, and A. Larsen, Phys. Rev. Lett. **78**, 503 (1997).
[15] K. Samonji, H. Yonezu, Y Takagi, and N. Ohshima, J. Appl. Phys. **86**, 1331 (1999).
[16] S. A. Chaparro, Y. Zhang, J. Drucker, D. Chandrasekhar, and D. J. Smith, Appl. Phys. Lett. **76**, 3534 (2000).
[17] J. W. Matthews and A. E. Blakeslee, J. Cryst. Growth **27**, 118 (1974).
[18] R. Leon, S. Marcinkevicius, X. Z. Liao, J. Zou, D. J. H. Cockayne, and S. Fafard, Phys. Rev. B **60**, R8517 (1999).

[This article is copyrighted as indicated in the article. Reuse of AIP content is subject to the terms at: http://scitation.aip.org/termsconditions. Downloaded to] IP: 209.147.144.21 On: Mon, 09 Feb 2015 22:28:37