Single InAs quantum dot arrays and directed self-organization on patterned GaAs (311)B substrates

Single InAs quantum dot arrays and directed self-organization on patterned GaAs (311)B substrates

E. Selçuk, A. Yu. Silov, and R. Nötzel[a)]
COBRA Research Institute, Eindhoven University of Technology, 5600MB Eindhoven, The Netherlands

(Received 8 April 2009; accepted 12 June 2009; published online 1 July 2009)

Formation of laterally ordered single InAs quantum dot (QD) arrays by self-organized anisotropic strain engineering of InGaAs/GaAs superlattice templates on GaAs (311)B by molecular beam epitaxy is achieved through optimization of growth temperature, InAs amount, and annealing. Directed self-organization of these QD arrays is accomplished by coarse substrate patterns providing absolute QD position control over large areas. Due to the absence of one-to-one pattern definition the site-controlled QD arrays exhibit excellent optical properties revealed by resolution limited (80 μeV) linewidth of the low-temperature photoluminescence from individual QDs. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3167813]

Laterally ordered semiconductor quantum dot (QD) arrays are highly demanded by the shrinking feature sizes in microelectonics[1,2] and especially the emerging quantum functional devices employing semiconductor QDs.[3,4] We have recently demonstrated the formation of laterally ordered InAs QD arrays on GaAs (311)B substrates by molecular beam epitaxy (MBE). The natural ordering of the QD arrays is created by self-organized anisotropic strain engineering of InGaAs/GaAs superlattice (SL) templates.[5,6] InGaAs QD growth, thin GaAs capping, anisotropic adatom surface migration during annealing, GaAs separation layer growth, and strain correlated stacking produces a two-dimensional (2D) lateral strain field modulation (of shallow 2D strain induced nodes) on the SL template surface governing InAs QD ordering on top in spotlike arrays of isolated QD molecules, down to single QDs in the center, due to local strain recognition. More complex architectures of QD arrays have been created on shallow- and deep-etched artificially patterned substrates. The self-organization, in this case, is guided by the stepped and faceted mesa sidewalls resulting in one-dimensional[7] and 2D (Ref. 8) QD arrays, and QD-free and dense regions, depending on the pattern design.

Here we first provide a detailed analysis of the formation of well ordered single InAs QD arrays. Next, we discuss that the substrate patterning will not only guide the self-organization with relative QD ordering but provoke also directed self-organization[9,10] with absolute QD position control. On coarse patterns of round holes and zigzag mesas with medium depth, the QD arrays are spatially locked to the mesa sidewalls leading to absolute QD position control over large areas. Earlier works to produce such ordered QD arrays required nanopatterning on the same length scales as the QD size and separation by sophisticated techniques such as electron beam or atomic force microscopy (AFM) lithography[11,12] which often degrade the structural and electronic quality of the QDs. The absence of one-to-one pattern definition in the directed self-organization process maintains high quality of the QDs which manifests itself in strong photoluminescence (PL) emission, comparable to unpatterned samples, and ultrasharp lines in low-temperature micro-PL from individual QDs with resolution limited linewidth of 80 μeV.

The samples were grown by solid source MBE on planar and patterned GaAs (311)B substrates. After oxide removal, the growth commenced with a 200 nm thick GaAs buffer layer at 580 °C, followed by a ten period InGaAs/GaAs SL template. Each SL period comprised, if not mentioned otherwise, 3.3 nm $In_{0.4}Ga_{0.6}As$ grown at 500 °C, 10 s growth interruption, thin capping by 0.5 nm GaAs at 500 °C, annealing for 2 min at 600 °C under As_4 flux, and growth of a 5.5 nm GaAs separation layer at 600 °C. The thickness of the last GaAs layer was 15 nm. On top of the SL template 0.6 nm InAs was deposited at 485 °C for formation of isolated InAs QD molecules. Single InAs QDs were formed at increased growth temperatures (~25 °C) of the SL template and InAs layer, and reduced InAs amount together with 30 s annealing. The QD formation was analyzed *in situ* by reflection high-energy electron diffraction (RHEED). The growth rates of GaAs and InGaAs were 0.073 and 0.132 nm/s, respectively, whereas that of InAs was 0.0013 5 nm/s. The substrate patterns of round holes and zigzag mesas were fabricated by optical lithography and wet-chemical etching in the $H_2SO_4:H_2O_2:H_2O$ (1:8:1000) solution.[13] The diameter of the holes was 6 μm, while the periodic zigzags had 10 μm width and separation with the mesa sidewalls alternately rotated $\pm 30°$ off $[0\bar{1}1]$. The etched depth was 100 nm. The structural properties of the samples were characterized by tapping-mode AFM in air. For PL the single QDs were capped with 100 nm GaAs. The micro-PL measurements were performed by exciting the samples, placed in a He-flow cryostat, with a He–Ne laser operating at 632.8 nm. The excitation power density was 35 $\mu W/cm^2$. Excitation and detection of the PL were through a microscope objective with spatial resolution of ~2 μm. The PL was dispersed by a triple monochromator and detected by a cooled InGaAs linear photodiode array with spectral resolution of 80 μeV.

Figure 1 shows AFM images of the transition of InAs QD molecules to single QDs. The shallow 2D strain induced nodes on the SL template surface before QD formation, discussed in Refs. 6 and 8 in detail, are visible in Fig. 1 as the rhombuslike network of ridges with the QDs centered at the crossing points. Well-isolated ordered InAs QD molecules, shown in Fig. 1(a), are formed at 485 °C with InAs amount

a)Author to whom correspondence should be addressed. Electronic mail: r.noetzel@tue.nl. Tel.: +31 40 247 2047. FAX: +31 40 246 1339.

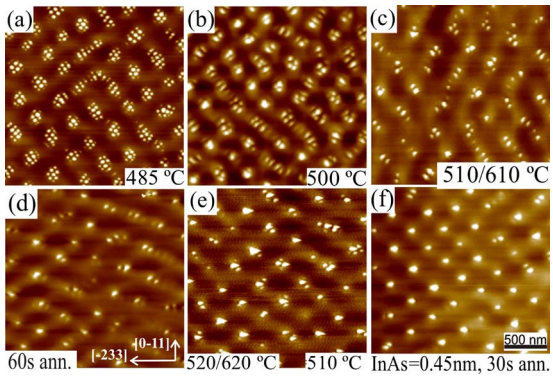

FIG. 1. (Color online) AFM images of the coalescence of InAs QD molecules into single QD arrays. (a) QD molecules, [(b)–(e)] transition stages as function of QD and SL growth temperatures, InAs amount, and annealing conditions as indicated in the text and figures, and (f) optimized single InAs QD arrays. The full height contrast is 15–20 nm.

of 0.6 nm, and InGaAs/GaAs growth and annealing temperatures of 500/600 °C for the SL template. When the QD growth temperature is increased to 500 °C [Fig. 1(b)] and the InGaAs/GaAs growth and annealing temperatures of the SL template to 510/610 °C [Fig. 1(c)] the InAs QDs begin to coalesce due to larger In adatom migration length during SL template and QD formation. Further QD coalescence occurs by 60 s annealing following the InAs QD formation [Fig. 1(d)]; however, on the 2D SL template nodes, multiple QDs, and a high percentage of missing QDs are observed, attributed to In desorption. Therefore, the annealing time is reduced to 30 s while the SL and InAs QD growth temperatures are further increased to 520/620 and 510 °C, and the InAs amount is reduced to 0.51 nm, improving single InAs QD formation [Fig. 1(e)]. Further increase in the growth and annealing temperatures results in thermal roughening leading to degradation of the structural quality of the QDs. Almost perfectly arranged single InAs QD arrays are created, finally, by a further reduction in the InAs amount to 0.45 nm as shown in Fig. 1(f). The average lateral periodicity of the single QDs is ~350 in the directions ±45° off $[0\bar{1}1]$, the base diameter is 80–100 nm and the areal density ~8.5 μm^{-2}.

The *in situ* RHEED analysis of the distinct growth stages is presented in Fig. 2. The relatively streaky pattern recorded in the $[\bar{2}33]$ azimuth of the SL template surface is shown in Fig. 2(a). The onset of QD formation which appears after about 5 min. of InAs deposition results in pronounced short-

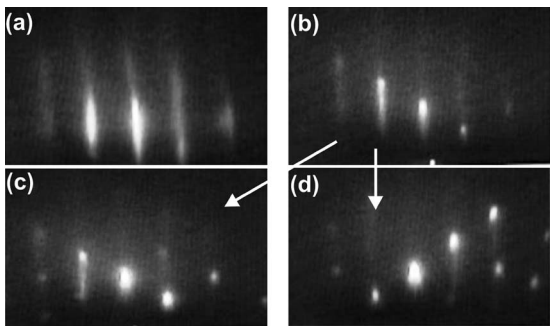

FIG. 2. RHEED patterns taken along $[\bar{2}33]$ during InAs QD formation. (a) SL template surface, (b) onset of QD formation, (c) single QD arrays, and (d) QD molecules.

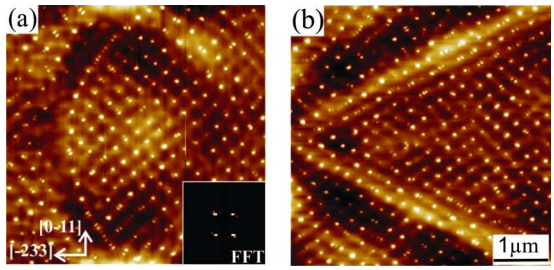

FIG. 3. (Color online) AFM images of single InAs QD arrays on (a) round hole and (b) zigzag mesa patterns. The inset in (a) shows the FFT image of the QD arrays in the center of the round hole pattern. The full height contrast is 50 nm.

ening of the streaks [Fig. 2(b)]. At this stage, InAs single QDs and/or groups develop which contain three or less QDs on average. After an additional growth time of ~30 s followed by 30 s annealing, a more spotty pattern is obtained with faint tails [Fig. 2(c)] corresponding to well isolated, periodically ordered single QDs for the growth conditions of the sample in Fig. 1(f). By further annealing of these QDs a streaky pattern evolves due to In desorption, flattening, and dissolving the QDs. On the other hand, a clear chevron pattern is observed [Fig. 2(d)] for InAs QD molecules by an additional growth time of ~2 min. beyond the onset of QD formation for the growth conditions of the sample in Fig. 1(a), due to denser coverage of the surface with QDs.

Figure 3 shows the AFM images of the single InAs QDs on the patterned substrates. On the hole pattern (the round hole shape changes to more triangular during GaAs buffer layer growth) the QDs arrange along the sidewalls in single rows following the contour of the holes [Fig. 3(a)]. This is attributed to preferential adatom migration from the slow-growing pattern sidewalls[14] to the bottom and top areas and enhanced strain relaxation at concave sidewall corners. The QD arrays inside the holes are spatially locked to the single rows of QDs, hence sidewalls, without change in the natural ordering in the areas away from the sidewalls. This is quantified by the fast Fourier transform (FFT) image shown in the inset of the Fig. 3(a) as well as the QD density and size in the center of the mesa holes which are unchanged compared to those in unpatterned areas. The QD arrays grown on the zigzag patterns shown in Fig. 3(b) (as well as on deep-etched zigzag and round hole patterns presented in Ref. 8) most clearly confirm this behavior (i.e., naturally ordered QD arrays which are spatially locked to the pattern sidewalls and corners) leading to absolute QD position control over large areas without one-to-one pattern definition.

Figure 4 depicts the PL properties of the single QDs on the patterned substrates. The temperature dependent micro-PL overview spectra of the GaAs capped single InAs QD arrays and SL template are shown in Fig. 4(a). At 5 K the PL energy of the SL template is centered at 1.34 eV and that of the QDs at ~1.28 eV. For increasing measurement temperature up to 60 K, the QD PL intensity strongly increases due to thermally activated carrier transfer from the SL template to the QDs. Distinct sharp emission lines from individual QDs are observed at elevated temperature together with sharp emission lines from localized excitons in the SL template. The temperature dependent high-resolution micro-PL spectra in the QD spectral region are shown in Fig. 4(b). Intense sharp lines are reobserved at elevated tempera-

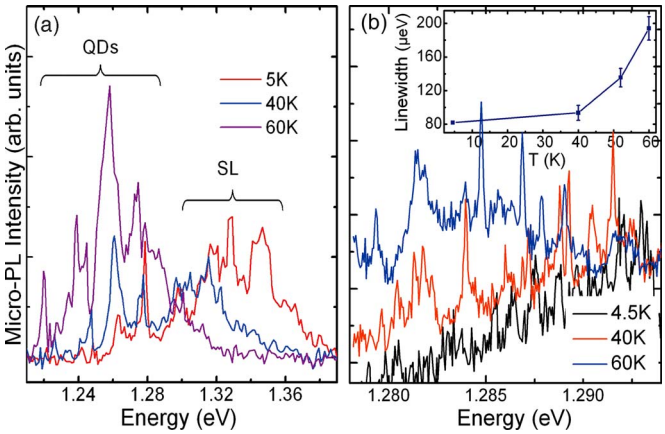

FIG. 4. (Color online) Temperature dependent micro-PL spectra of the capped single InAs QD arrays on patterned substrates. (a) Overview in the QDs and SL template spectral regions, and (b) in the QDs spectral region with 80 μeV resolution. The inset in (b) shows the linewidth broadening with increasing temperature measured for various lines indicated by the error bars.

ture. The linewidth slowly increases with temperature up to 40 K due to acoustic phonon scattering followed by a much steeper increase for higher temperatures due to optical phonon scattering,[15] shown in the inset. At 4.5 K, though the efficiency is low, sharp lines from individual QDs can clearly be identified from the temperature dependence with a resolution limited linewidth of 80 μeV.

In conclusion, formation of laterally ordered single InAs QD arrays by self-organized anisotropic strain engineering of InGaAs/GaAs SL templates on GaAs (311)B by MBE was achieved through optimization of growth temperature, InAs amount, and annealing. Directed self-organization of these QD arrays was shown on coarse substrate patterns of round holes and zigzag mesas. The QD arrays were spatially locked to the mesa sidewalls providing absolute QD position control over large areas. Due to the absence of one-to-one pattern definition the site-controlled QD arrays exhibit excellent optical properties revealed by resolution limited (80 μeV) linewidth of the low-temperature PL from individual QDs, which is required for the realization of future quantum functional devices.

[1]Semiconductor Industry Association, *International Technology Roadmap for Semiconductor* (Semiconductor Industry Association, San Jose, CA, 2005).
[2]D. J. C. Herr, Future Fab. Int. **20**, 82 (2006).
[3]D. Loss and D. P. DiVincenzo, Phys. Rev. A **57**, 120 (1998).
[4]M. H. Huang, S. Mao, H. Feick, H. Yan, Y. Wu, H. Kind, E. Weber, R. Russo, and P. Yang, Science **292**, 1897 (2001).
[5]T. Mano, R. Nötzel, G. J. Hamhuis, T. J. Eijkemans, and J. H. Wolter, Appl. Phys. Lett. **81**, 1705 (2002).
[6]T. v. Lippen, R. Nötzel, G. J. Hamhuis, and J. H. Wolter, Appl. Phys. Lett. **85**, 118 (2004).
[7]T. Mano, R. Nötzel, D. Zhou, G. J. Hamhuis, T. J. Eijkemans, and J. H. Wolter, J. Appl. Phys. **97**, 014304 (2005).
[8]E. Selçuk, G. J. Hamhuis, and R. Nötzel, J. Appl. Phys. **102**, 094301 (2007).
[9]R. Ruiz, H. Kang, F. A. Detcheverry, E. Dobisz, D. S. Kercher, T. R. Albrecht, J. J. de Pablo, and P. F. Nealey, Science **321**, 936 (2008).
[10]I. Bita, J. K. W. Yang, Y. S. Jung, C. A. Ross, E. L. Thomas, and K. K. Berggren, Science **321**, 939 (2008).
[11]S. Kiravittaya, A. Rastelli, and O. G. Schmidt, Appl. Phys. Lett. **87**, 243112 (2005).
[12]S. Ohkouchi, Y. Nakamura, H. Nakamura, and K. Asakawa, Jpn. J. Appl. Phys., Part 1 **44**, 5777 (2005).
[13]D. W. Shaw, J. Electrochem. Soc. **128**, 874 (1981).
[14]R. Nötzel, M. Ramsteiner, J. Menniger, A. Trampert, H.-P. Schönherr, L. Däweritz, and K. H. Ploog, J. Appl. Phys. **80**, 4108 (1996).
[15]N. I. Cade, H. Gotoh, H. Kamada, H. Nakano, S. Anantathanasarn, and R. Nötzel, Appl. Phys. Lett. **89**, 181113 (2006).