InAs/GaAs nanostructures grown on patterned Si(001) by molecular beam epitaxy

This content has been downloaded from IOPscience. Please scroll down to see the full text.

2008 Nanotechnology 19 455607
(http://iopscience.iop.org/0957-4484/19/45/455607)

View the table of contents for this issue, or go to the journal homepage for more

Download details:

IP Address: 124.16.156.249
This content was downloaded on 24/12/2013 at 03:25

Please note that terms and conditions apply.
InAs/GaAs nanostructures grown on patterned Si(001) by molecular beam epitaxy

Jun He\textsuperscript{1,3}, Kameshwar Yadavalli\textsuperscript{1}, Zuoming Zhao\textsuperscript{1}, Ning Li\textsuperscript{1}, Zhibiao Hao\textsuperscript{1}, Kang L Wang\textsuperscript{1} and Ajey P Jacob\textsuperscript{2}

\textsuperscript{1} Device Research Laboratory, Electrical Engineering, University of California, Los Angeles, CA 90095, USA
\textsuperscript{2} TMG External Programs, Intel Corporation, Santa Clara, CA 95052, USA

E-mail: hejun@ee.ucla.edu

Received 29 May 2008, in final form 15 August 2008
Published 9 October 2008
Online at stacks.iop.org/Nano/19/455607

Abstract

The potential benefit from the combination of the optoelectronic and electronic functionality of III–V semiconductors with silicon technology is one of the most desired outcomes to date. Here we have systematically investigated the optical properties of InAs quantum structure embedded in GaAs grown on patterned sub-micron and nanosize holes on Si(001). III–V material tends to accumulate in the patterned sub-micron holes and a material depletion region is observed around holes when GaAs/InAs/GaAs is deposited directly on patterned Si(001). By use of a 60 nm SiO\textsubscript{2} layer and patterning sub-micron and nanosize holes through the oxide layer to the substrate, we demonstrate that high optical quality InAs nanostructures, both quantum dots and quantum wells, formed by a two-monolayer InAs layer embedded in GaAs can be epitaxially grown on Si(001). We also report the power-dependent and temperature-dependent photoluminescence spectra of these structures. The results show that hole diameter (sub-micron versus nanosize) has a strong effect on the structural and optical properties of GaAs/InAs/GaAs nanostructures.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Low dimensional quantum structures such as quantum wells (QWs), quantum wires (QRs) and quantum dots (QDs) have been the focus of vast research efforts and the improvement of their electrical and optical properties have enabled the realization of many devices through novel physical effects [1–5]. One of the most widely investigated methods for QD fabrication is self-assembly in the Stranski–Krastanov (SK) growth mode in epitaxy of lattice mismatched systems [6–10]. Among these, self-assembled In(Ga)As/GaAs QDs have been most extensively studied for the last decade and a good understanding of their electronic and optical properties has been achieved both experimentally [1, 2] and theoretically [3, 4]. Their quantum confinement properties have been exploited for device applications such as quantum well infrared detectors [5], single photon quantum dot sources [6] and ultra-low-threshold quantum dot lasers [8].

On the other hand, silicon is currently the main material for microelectronics and nanoelectronics, accounting for roughly 90\% of the market for semiconductor devices. However, the indirect nature of the Si band structure prevents the realization of efficient light emitting devices as a result of the correspondingly low dipole transition probabilities. The potential benefit from combination of optoelectronic and electronic functionality of III–V semiconductors with silicon technology is one of the most desired outcomes. The possibility of III–V QDs (such as InAs QDs) grown on Si with good optical properties will greatly enable the integration of Si microelectronics and nanoelectronics with optoelectronic devices for interconnect and other optical communication applications. Up to now, researchers have

3 Author to whom any correspondence should be addressed.
focused on the growth of a continuous layer of III–V materials on Si [11–13]. However, the growth of electronic and optical quality material on Si remains a practically unsolved problem due to the large mismatch between Si and III–V semiconductor (e.g. InAs and GaAs). For example, Zakharov et al. have reported MBE growth of InAs directly on Si [11] and HRTEM revealed a high density of InAs clusters with a diameter of 3 nm. Such extremely small clusters prevent sufficient carrier localization and exhibit a very broad photoluminescence (PL) at 10 K. In this paper, we systematically investigate the effect of SiO2 thin film and the pattern size on optical properties of GaAs/InAs/GaAs nanostructure and demonstrate an effective route to obtain III–V nanostructure with good optical properties grown by MBE on patterned Si(001) masked with SiO2. We show that with the introduction of 60 nm patterned SiO2 on Si(001), the optical properties of GaAs/InAs/GaAs nanostructure formed by 2 MLs InAs layer embedded in GaAs grown on the patterned SiO2/Si are greatly improved as compared with those grown directly on patterned Si. We also report the power-dependent and temperature-dependent PL spectra of these nanostructures grown on holes of different size which show that hole size has a strong effect on the structural and optical properties of GaAs/InAs/GaAs nanostructure grown by MBE.

2. Experimental details

All the three samples studied are e-beam patterned hole arrays, approximately 360,000 holes are patterned over an area of 1 mm × 1 mm on each sample, the patterned size is sufficiently large to be able to position the laser beam spot of the PL measurement system. After developing the resist, the hole patterns were transferred by using CHF3 based reactive-ion etching (RIE). For sample A, the patterned holes with diameter of about 230 nm were etched down 60–90 nm into Si(001) substrate. For samples B and C, hole arrays were patterned and etched down to 60–90 nm depth through a 60 nm SiO2 mask layer which was first grown by thermal oxidation of Si substrates (in the holes, fresh Si surface is exposed). The hole diameter is 230 nm for sample B and 80 nm for sample C, respectively. Figure 1(a) shows the schematic structure of samples A (top schematic, prior to MBE growth of GaAs/InAs/GaAs layers), B and C (bottom schematic, after MBE growth of GaAs/InAs/GaAs layers). All patterned substrates were chemically cleaned before growth. For samples A, B and C, the same amount of GaAs/InAs/GaAs layers were grown by solid source MBE under exactly the same growth conditions. After the native oxide desorption at 800 °C (via pyrometer measured temperature), a nominal 200 nm GaAs buffer layer was grown at a substrate temperature of 670 °C. The temperature was then lowered to 530 °C to deposit 2 MLs of InAs with a growth rate of 0.075 Å s\(^{-1}\). The structure of the 2 MLs InAs depends strongly on the patterned hole size. After the InAs growth, the sample is capped with a 5 nm GaAs layer grown at 530 °C and then further capped with a 12 nm thick GaAs layer grown at 610 °C. During growth, the deposition of GaAs and InAs layers was monitored by reflection high-energy electron diffraction (RHEED). The morphologies of the GaAs/InAs/GaAs structures were investigated by scanning electron microscopy (SEM) and PL measurements were performed under the excitation of 488 nm line of an Ar-ion laser and the luminescence spectra were detected by liquid nitrogen cooled InGaAs photodetector array.

![Image](image-url)
GaAs/InAs/GaAs layers. (broad peak) from the patterned area is related to the MBE grown transverse optical (TO)-phonon-assisted excitons from Si sample. The PL from the unpatterned region is attributed to the outer curve) and unpatterned (sharp inner curve) regions of the GaAs/InAs/GaAs layers. The InAs layer is selectively grown buffer layer) in sample B and sample C indicate high quality 400 nm. The clear facets of GaAs pillar (which serves as a mask layer. Only the hole array regions with exposed Si surface are filled by GaAs with a net thickness of around 3. Results and discussion

Figures 1(b) and (c) show the SEM images of surface morphologies of sample A and C after-growth of GaAs/InAs/GaAs layers. In figure 1(b), an SEM image taken in the patterned area of sample A is shown. From the SEM image, we can identify the patterned area and can clearly see that III–V materials accumulate in the patterned holes and a material depletion region forms around the sub-micron holes. This material depletion region around the patterned holes looks very similar to the result obtained for InAs grown on patterned GaAs(001) substrates [14]. This might indicate the same diffusion mechanism for the growth on finite patterned areas in both material systems (In(Ga)As/Si and InAs/GaAs). The accumulation of III–V materials in the holes can be attributed to the fact that the subsequent gallium or indium adatoms will directionally diffuse toward the holes since the strain field from the GaAs initially deposited in the holes lowers the chemical potential of the gallium or indium adatoms inside the patterned area. Figure 1(c) shows the typical selective growth of III–V materials (200 nm GaAs buffer layer, subsequent 2 MLs InAs and immediate GaAs capping layer) in the holes of sample C. GaAs is selectively filled in the nanoholes. The inset in figure 1(c) shows the SEM image of sample B with GaAs selectively filled in the sub-micron holes. The growth conditions are optimized for achieving selective epitaxy [15]. Hence, there is no nucleation observed on top of the SiO2 mask layer. Only the hole array regions with exposed Si surface are filled by GaAs with a net thickness of around 400 nm. The clear facets of GaAs pillar (which serves as a buffer layer) in sample B and sample C indicate high quality of GaAs crystallinity which implies better optical properties of GaAs/InAs/GaAs layers. The InAs layer is selectively grown on top of these pillars and capped immediately by a thin GaAs layer for PL measurements.

Figure 2 depicts low temperature PL spectra of sample A under the excitation power density of 5.6 W cm$^{-2}$. When the laser beam is shining on the patterned area the PL spectrum reveals a broad peak (outer curve) and when the laser beam is moved to the unpatterned area, the broad peak disappears and a sharp peak (inner curve) centered at 1127 nm comes up due to the transverse optical (TO)-phonon-assisted excitons from Si:P [16, 17]. This clearly shows that the broad peak is related to the MBE grown GaAs/InAs/GaAs layers. However, the PL efficiency from this structure is weak probably due to the formation of dislocations in the III–V materials grown on the patterned Si substrate as well as due to misfit dislocations at the interface. These results indicate that on patterned Si substrate, under the applied growth conditions, MBE grown III–V material tends to accumulate in the holes, however, the optical properties are not greatly improved as compared with the growth of continuous layer of III–V materials and III–V QDs directly on unpatterned Si substrate [11–13].

The PL spectra of sample B obtained with excitation power density from 0.7 to 56 W cm$^{-2}$ at 65 K are shown in figure 3(a). All the PL spectra exhibit a three-peak feature at 966 nm (peak 1), 1033 nm (peak 2), and 1127 nm (peak 3) even at very low excitation power density. Peak 3 is attributed to TO-phonon-assisted excitons from Si:P [16]. This is confirmed by the observation that as the laser beam moves out of the pattern area, no PL peak is observed except peak 3. Thus, we assign peak 1 and peak 2 with full-width at half-maximum (FWHM) of 50 meV and 83 meV, respectively, to the 2 MLs InAs embedded in GaAs layers. The PL integrated intensity from GaAs/InAs/GaAs of sample B is strongly improved in comparison with that of sample A. As the integrated intensity reflects the influence of the nonradiative recombination channels in the structure, the above results indicate that the crystallinity of the III–V material grown on sample B is much higher compared to that on sample A. Thus, optical quality of GaAs/InAs/GaAs layers selectively grown on patterned SiO2/Si (sample B) is greatly improved compared with that grown directly on patterned Si substrate (sample A), under these growth conditions. For the applied growth conditions (as above) of InAs on GaAs buffer layer, InAs QDs typically emit around 1 µm with a linewidth of several tens of milli-electron-volts [18, 19]. The PL results obtained here imply the formation of InAs QDs on the GaAs buffer layer grown in 230 nm diameter holes, which is also confirmed by the following temperature-dependent PL measurement. As discussed previously, all the PL spectra, in the range which is related to the 2 MLs InAs, can be decomposed into two Gaussian peaks (peak 1 and peak 2) with an energy interval of 65 meV shown in dash lines in figure 3(a). Figure 3(b) shows the integrated PL intensity of QDs (peak 1 + peak 2) and Si peak (peak 3) obtained from figure 3(a) as a function of the excitation laser power density. The integrated PL intensity of QDs and the Si peak increases linearly with increasing excitation power density (from 0.7 to 56 W cm$^{-2}$) at 65 K. The PL peak positions and the intensity ratio of peak 2 to peak 1 do not change significantly with the increase in excitation power.
Figure 3. (a) PL spectra of sample B obtained at 65 K by varying excitation power density from 0.7 to 56 W cm\(^{-2}\). Each of the spectra can be decomposed into two Gaussian peaks, shown with the dashed lines (bottom). (b) Dependence of integrated PL intensity as a function of excitation power. Dash-dot lines are a linear fitting to experimental data. The PL intensity ratio of peak 2 to peak 1 is also plotted as a function of power density. The dotted line is a linear fit to experimental data and shows that the PL intensity ratio is constant as power density is changed.

To further identify the nature of the two peaks, the temperature dependence of the PL emission of sample B was investigated under the excitation power density of 5.6 W cm\(^{-2}\) from 65 to 135 K, as shown in figure 4(a). In figure 4(a) the top-most curve is the PL emission obtained at 65 K with each subsequent curve below obtained at a higher temperature (temperature increment 5 K). As expected, the two PL peaks shift to longer wavelengths as the temperature increases, which is mainly due to thermal expansion of the lattice constant and electron–phonon scattering, as in bulk semiconductors. Meanwhile, as the temperature is increased, the intensity of peak 1 (high-energy peak) decreases. The intensity ratio of peak 2 to peak 1 shown in figure 4(b) increases initially as the temperature is increased to 85 K reaching a maximum around 85 K, and then the intensity ratio decreases as the temperature is increased to 120 K, with the intensity ratio at 120 K remaining higher than at 65 K. The observed behavior can be attributed to the carrier repopulation process with increasing temperature. As the temperature is increased up to 85 K, carriers are thermally activated from the small dots...
Figure 5. (a) Excitation power dependence of the PL spectra from sample C. A single peak centered at 869 nm is seen relating to the InAs layers embedded in GaAs. The wavelength of this emission peak is very close to that from InAs wetting layer (870 nm) in self-assembled InAs/GaAs QDs, indicating that in sample C, deposition of 2 MLs of InAs results in the formation of GaAs/InAs/GaAs quantum well. (b) Temperature dependence of the PL for sample C. The top-most curve is the PL emission obtained at 65 K with each subsequent curve below obtained at a higher temperature (temperature increment of 10 K). A red-shifted peak from the InAs/GaAs layers can be seen as shown by the dashed line. For temperature greater than 85 K, part of the carriers thermally excited to the WL and/or the GaAs barrier layer from the quantum dots recombine in the WL and/or the GaAs barrier layer while the remaining carriers are captured by the larger QDs leading to the observed intensity ratio behavior between 85 and 120 K. In sample B, there might be several tens of InAs QDs on the GaAs buffer layer grown in a given 230 nm diameter hole and the InAs QDs in each hole are separated from the InAs QDs grown in neighboring holes. Hence, we do not see a change in FWHM with temperature as also shown in figure 4(b), unlike in the case of InAs QDs grown on GaAs [9, 20]. Earlier, carrier repopulation process has been reported in InAs QDs grown on GaAs substrate [20, 21]. To our knowledge, this is first time such carrier redistribution was observed in the InAs/GaAs QDs grown on Si(001). The PL emission from sample B was found to persist up to 260 K. The persistence of this excitonic emission at high temperature is evidence suggesting the formation of dislocation-free InAs QDs grown on patterned SiO$_2$/Si due to additional lateral confinement. These results suggest that selectivity grown GaAs in the sub-micron holes provides a high quality GaAs buffer layer for subsequent growth of InAs self-assembled QDs to achieve strong optical activity with proper carrier confinement.

In order to investigate the effect of pattern size on the structural and optical properties of InAs layers embedded in GaAs selectively grown in the patterned holes, we performed excitation-power-dependent and temperature-dependent PL measurement on sample C which has patterned nanoholes of 80 nm diameter. Figure 5(a) depicts the PL from sample C for excitation power density from 0.35 to 56 W cm$^{-2}$. In contrast to the PL from sample B showing two clearly defined peaks at the energies 1.282 eV (966 nm), and 1.199 eV (1033 nm) from two group of QDs, the PL from sample C exhibits a single peak centered at 1.426 eV (869 nm) at 65 K, originating from the InAs layers embedded in GaAs. For self-assembled InAs/GaAs QDs, the peak from InAs wetting layer is normally centered around 870 nm [2, 22] which indicates that, for sample C, deposition of the same amount (2 MLs) of InAs results in the formation of GaAs/InAs/GaAs quantum well instead of InAs/GaAs QDs as in sample B. This is further confirmed by temperature-dependent PL measurement shown in figure 5(b). Figure 5(b) depicts PL from sample C measured from 65 to 135 K at an excitation power density of 5.6 W cm$^{-2}$ with a temperature increment of 10 K, and as mentioned above we see one peak (wavelength 869 nm at 65 K) associated with the InAs layers embedded in GaAs. The temperature-dependent measurement clearly shows the variation of the peak position with temperature (a red shift can be seen indicated by the dashed line). Also, FWHM as a function of temperature (not shown) deduced from figure 5(b) agrees well with the characteristic WL (quantum well) behavior in InAs/GaAs QDs [2, 21]. This result implies that the critical thickness of 2D to 3D transition can be extended when the pattern size is scaled down, in our case, from 230 nm down to 80 nm, leading to different InAs/GaAs nanostructure and optical properties under the same deposition of 2 MLs InAs. This is due to the fact that, as shown in figure 1(a), InAs was deposited on the top of selectively grown GaAs buffer layer which forms pillars in the holes. Figure 1(c) clearly shows that the mesa area of...
selectively grown GaAs pillar in sample B is greater than that in sample C. Hence, the InAs epitaxial layer grown on the GaAs mesa of sample B can mostly deform vertically to relieve the mismatch stress on a larger mesa. In contrast, for sample C, the mismatch stress can be relieved not only vertically but also laterally on a smaller mesa area. In addition, the strain partitioning between the epitaxially grown InAs layer and the selectively grown GaAs mesa in the 80 nm hole of sample C occurs more strongly compared to that of sample B. Huang [4] demonstrated a theoretical model for strain and dislocations in epitaxial layers grown on substrates of a finite dimension (mesa), which showed that the strain energy in epitaxial layers can be significantly reduced and thus, the critical thickness can be significantly extended when the layer is nucleated on a patterned substrate mesa. Our results are consistent with this theoretical model. The intensity of the peak centered at 869 nm from InAs/GaAs quantum well (in sample C) is not as strong as that from InAs/GaAs QDs in sample B, this is probably due to the low responsivity of our InGaAs photodetector below 900 nm and also due to less lateral carrier confinement in the InAs/GaAs quantum well structure.

4. Conclusion

We have studied the effect of thin SiO$_2$ and the pattern size on the structural and optical properties of GaAs/InAs/GaAs layers grown by MBE on Si(001). We have demonstrated that GaAs/InAs/GaAs layers with high structural quality and strong optical activity, both QDs and QWs, can be epitaxially incorporated on Si by MBE growth. Selectively grown GaAs in the patterned holes provides a high quality buffer layer for InAs self-assembled growth to achieve strong optical activity. Furthermore, we also have investigated the effect of pattern size on the optical and structural properties of GaAs/InAs/GaAs layers. This work demonstrates the possibility of using nanostructures for integration of III–V materials on Si and of obtaining strong optical activity in these lattice mismatched systems on Si, which may lead to efficient Si-based LEDs and lasers in the near future.

Acknowledgments

This work was partially supported by an Intel grant (Dr Wilman Tsai) and Focus Center Research Program (FCRP), Center on Functional Engineered Nano Architectonics (FENA) monitored by Dr Betsy Weitzman. We thank the staff of UCLA Nanolab for their support.

References