Control of polarized emission from selectively etched GaN/AlN quantum dot ensembles on Si(111)

Daniel H. Rich*,1, Ofer Moshe1, Benjamin Damilano2, and Jean Massies2

1 Department of Physics, The Ilse Katz Institute for Nanoscale Science and Technology, Ben-Gurion University of the Negev, P.O.B 653, Beer-Sheva 84105, Israel
2 Centre de Recherche sur l’Hétéro-Epitaxie et ses Applications, Centre National de la Recherche Scientifique, Rue B. Gregory, Sophia Antipolis, 06560 Valbonne, France

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Multiple layers of GaN/AlN quantum dot (QD) ensembles were grown by the Stranski-Krastanov method on Si(111) using molecular beam epitaxy. During the subsequent cooling from growth temperature, the thermal expansion coefficient mismatch between the Si substrate and GaN/AlN film containing the vertically stacked QDs leads to an additional biaxial tensile stress of 20–30 kbar in the III-nitride film. We have selectively modified the thermal stress in the QD layers by etching a cross-hatched pattern into the as-grown sample using inductively coupled Cl2/Ar plasma reactive ion etching. The results show that a suitable choice of stripe width from ~2 to 10 μm and orientation along [11-20] and [1-100] can create regions of in-plane uniaxial stress that enable a selective and local control of the polarized luminescence from ensembles of QDs which were probed with cathodoluminescence. A theoretical modeling of the effects of carrier filling on the polarization anisotropy and the excitonic transition energy was performed, as based on three dimensional self-consistent solutions of the Schrödinger and Poisson equations using the 6×6 k-p method.

1 Introduction

Potential solid-state light emitting applications in the visible wavelength range continue to supply the motivation for studies of GaN/AlN-based wurtzite self-assembled quantum dots (QDs) [1,2,3]. An important characteristic of group III-nitride compounds is the existence of a large polarization field, originating from both piezoelectric and spontaneous polarizations [1,2]. Recently, we demonstrated that thermal stress induced microcracks along <11-20> in GaN/AlN QD layers grown on Si(111) can serve as excellent stressors which create an in-plane uniaxial stress of 20–30 kbar in limited regions of the III-nitride film, owing to the large mismatch in thermal expansion coefficient between the III-N film and Si [1,2,3,4,5]. In close proximity (i.e., within a few μm’s) of the microcracks the excitonic luminescence from the GaN QDs is partially linearly polarized, as a result of the uniaxial stress [7-10]. Moreover, the selective etching of a pattern in these samples has recently demonstrated a tailoring of the stress and local control of the polarized emission [11]. The effect of an in-plane uniaxial stress on optical transitions in GaN/AlN QDs has been examined theoretically [9,12,13].

The development of III-nitride-based light emitting diodes (LEDs) with polarized light emission is a topic of current interest with immediate applications in backlit nematic-phase liquid-crystal displays (LCDs). Exploiting polarized III-nitride based LEDs in LCD systems could yield a substantial increase in power efficiency for the display system by reducing or eliminating the need for polarization filters. Consequently, an added motivation for the implementation of III-nitride QD polarized LEDs fabricated on standard Si(111) substrates is for potential applications in large area LCD systems, which could benefit economically from the integration of III-nitride QD films with Si microelectronics [14].
In this paper, we further demonstrate a method to selectively manipulate thermal stress in III-nitride films grown on Si and to control the linear polarization of the excitonic luminescence from nitride-based QDs. Such a post-growth control of strain in epitaxially grown nanostructures using an ex-situ patterning of the QD film is shown to provide a potentially useful method for the generation of polarized light emitters. We have selectively modified the thermal stress in multiple layers of GaN/AlN self-assembled QDs grown on Si(111) by etching a cross-hatched pattern into the as-grown sample using inductively coupled Cl₂/Ar plasma reactive ion etching (RIE). The results show that a suitable choice of stripe width from ~2 to 10 μm and orientation can create regions of in-plane uniaxial stress that enable a selective and local control of the polarized luminescence from ensembles of QDs. Using cathodoluminescence (CL) imaging and spectroscopy with linear polarization detection, we have analyzed the polarized emission from the selectively etched GaN QD/Si(111) cross-hatched pattern. Moreover, we have measured the excitation-dependence of the polarized luminescence emitted from QDs in order to better understand the effects of stress on the p-orbital character of holes in excited states and the resulting optical polarization anisotropy. We present results of our theoretical modelling of the effects of carrier filling on the anisotropy of the polarized emission, as based on three-dimensional (3D) self-consistent solutions of the Schrödinger and Poisson equations using the 6\(k\)-\(p\) method.

2 Experiment

The GaN/AlN QD sample was grown by molecular beam epitaxy (MBE) using the Stranski-Krastanov growth mode transition. The sample was grown on Si(111) and consists of AlN (30 nm)/GaN (400 nm)/AlN (700 nm) buffer layers followed by 85 layers of GaN QDs, labeled as sample S85. The growth involved 6.7-nm thick AlN barrier layers with 1.6-nm thick GaN QD layers, resulting in an average dot density of ~5 \(\times\) \(10^{10}\) cm\(^{-2}\) [6-9]. SiO₂ and photo-resist mask patterns were formed on the sample, through which the GaN/AlN QD layers were selectively etched until the underlying Si substrate was exposed. Inductively coupled Cl₂/Ar plasma RIE was used to etch into the as-grown sample a cross-hatched pattern consisting of square trenches of area 20 × 20 μm\(^2\) and having stripes of varying widths along the orthogonal in-plane [11-20] and [1-100] directions. The main growth and processing steps are schematically illustrated in Fig. 1. The spacing between the edges of the square trenches was intentionally varied to create vertical and horizontal stripes having widths ranging from ~2 to 10 μm. The widths of adjacent parallel stripes decrease by ~1 μm in both left-to-right and top-to-bottom sequences of the cross-hatched pattern, as illustrated in Fig. 1(f) and observed in the scanning electron microscopy (SEM) and CL images in Fig. 2.

Our CL detection system is mounted on a JEOL 5910 SEM [6-9]. Two polarization directions for the polarizer will be denoted with the subscripts \(\perp\) and \(\parallel\) to indicate detection orientations with \(\mathbf{E}\) perpendicular and parallel to a stripe along [11-20]. The polarization anisotropy ratio, \(R_p\), is defined by the ratio of the integrated CL intensities, \(I\), under the two orthogonal polarizer orientations and is given by \(R_p = I_\perp/I_\parallel\). CL spectra and images were acquired with an e-beam energy (\(E_b\)) of 15 keV and beam currents (\(I_b\)) that varied between 50 pA and 30 nA. The spectral resolution of the monochromator was 2 nm (~15 meV) at \(\lambda = 400\) nm (3.100 eV).

3 Results and discussion

The patterned region that was probed with CL is shown in the SEM and polarization anisotropy ratio images of Figs. 2(a)-2(f). A magnified portion of the dashed rectangle in Fig. 2(a) is shown in Fig. 2(b). The horizontal and vertical dashed lines in Fig. 2(b) indicate paths on which the focused e-beam was positioned for localized CL spectroscopy measurements. Local CL spectra at \(T = 46\) K, acquired under \(\mathbf{E} \perp [11-20]\) and \(\mathbf{E} \parallel [11-20]\) detection orientations, are shown in Fig. 3 for various e-beam positions along the center of vertical and horizontal stripes that connect adjacent square trenches. The distances \(\Delta x\) and \(\Delta y\) indicate distances from the beginning of the dashed horizontal and vertical lines in Fig. 2(b) on which the e-beam was focused during the acquisition of the local CL spectra of Fig. 3. The polarization anisotropy is...
evident in Fig. 3(a) by the ratios, $R_p$, which vary from 1.53 to 1.75 for each set of CL spectra acquired along the vertical stripe (i.e., increments along the $\Delta y$ direction). A similar result is observed for CL spectra acquired for excitation on the orthogonal stripe with increments along the $\Delta x$ direction, as observed in Fig. 3(b).

The spatial distribution of the polarization anisotropy ratio, $R_p$, was further examined by acquiring monochromatic CL images at $h\nu = 3.00$ eV under $E \perp [11-20]$ and $E \parallel [11-20]$ detection orientations, as shown in Figs. 2(c) and 2(d), respectively, at $T = 46$ K. Spatial variations in CL emission can be observed, as the intensity is modulated with the appearance of bright and dark bands parallel to the edges of certain stripes in the monochromatic images. Since these bands appear to correlate with undulations in the surface texture observed in the same corresponding regions of the SEM image of Fig. 2(a), we attribute their presence as due to an inhomogeneity during mask formation and subsequent RIE, leading to a partial etching of some regions under the patterned mask. The images of Figs. 2(e) and 2(f), maintained at temperatures of 300 K and 46 K, respectively, show ratios $R_p = I_E(x,y)/I_{\perp}(x,y)$ for each $640 \times 480$ pixel image. The false color bar is used to map $R_p$ and shows variations in $R_p$ between adjacent vertical and horizontal stripes exhibiting varying widths for the two sample temperatures. A clear reversal in the polarization anisotropy is seen near the centers of horizontal and vertical stripes, consistent with the polarized CL spectra shown in Fig. 3(a). Moreover, the largest values of $R_p$ ($R_p \approx 2$) are observed for the narrowest stripes approaching $\sim 4 \mu$m towards the right in Fig 2(d), owing to a more complete stress relief (i.e., $\sigma_x \approx 0$ and $\sigma_y \approx 30$ kbar) along the stripe’s orthogonal direction near its center. An increase in $R_p$ is observed near the centers of the stripes as the temperature is reduced from 300 to 46 K. An increase in uniaxial tensile stress at low temperatures is expected to occur due to the further contraction of the GaN/AlN film relative to the Si substrate and leads to an increase in $R_p$ near the centers of the stripes. A similar temperature dependence of $R_p$ was observed in close proximity to microcracks in sample S85, again as a result of an increase in uniaxial stress during sample cooling [7,8]. In addition, a reduced thermal excitation of holes into higher energy hole states occurs at lower temperatures and contributes to a larger $R_p$, as will be shown in the results of the self-consistent $k-p$ calculations. In most cases, the intersections of horizontal and vertical stripes yield an $R_p \approx 1$ which is consistent with the presence of biaxial stress (i.e., $\sigma_x \approx \sigma_y$). Some exceptions can be found, particularly at $T = 46$ K, owing to a possible anisotropic contraction of the QD layers in the cross-hatched pattern during cooling.

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We have measured $R_p$ and the shift in the QD excitonic transition energy, $\Delta E_{2s}$, as a function of the $e$-beam excitation density for temperatures of 46 and 300 K. The $e$-beam was focused on a position near the center of a stripe and the $e$-beam current, $I_e$, was varied during the $R_p$ and $E_r$ measurements, the results of which are shown in Fig. 4. We express the carrier population in the QDs using the average $e-h$ carrier occupation number, $<n>$, which we de-
formed. The Hartree potential, $e^{-}\hbar$ model with remote band coupling for wurtzite semiconductors can be simplified to the 6 $\times$ 6 excitation dependence of Pikus (RSP) Hamiltonian [15,16]. In order to model the valence-band structure, i.e., the so-called Rashba-Sheka-tions were performed self-consistently for a uniaxial stress of 30 kbar. The downward arrows indicate the locations of steps that occurred in $R_p$ upon an increasing excitation at low temperatures for both the experimental data and calculations. Since the mixing of conduction and valence bands near the direct band edges is negligible, due to the large energy gap, the exact 8 $\times$ 8 $\mathbf{k} \cdot \mathbf{p}$ Hamiltonian obtained from Kane’s model with remote band coupling for wurtzite semiconductors can be simplified to the 6 $\times$ 6 $\mathbf{k} \cdot \mathbf{p}$ Hamiltonian for the valence-band structure, i.e., the so-called Rashba-Sheka-Pikus (RSP) Hamiltonian [15,16]. In order to model the excitation dependence of $R_p$ and $\Delta E_r$, in the GaN/AlN QDs, we have employed three dimensional 6 $\times$ 6 $\mathbf{k} \cdot \mathbf{p}$ calculations using the nextnano$^3$ quantum nanostructure simulation code [6-9]. We have extended the multi-band $\mathbf{k} \cdot \mathbf{p}$ and single band effective mass treatment for the occupation of many electron and hole states. Self-consistent calculations of the Schrödinger and Poisson equations using the 6 $\times$ 6 $\mathbf{k} \cdot \mathbf{p}$ and effective mass methods for the calculation of the $e^{-}\hbar$ wavefunctions in the Hartree approximation were performed. The Hartree potential, $V_{H}(r)$, was calculated from the total charge density associated with the occupation of the electron and hole ground and excited energy states in a manner consistent with a determination of the electron and hole quasi-Fermi levels, $\varphi_n$ and $\varphi_h$. The Hartree potential and the potential due to both piezoelectric and spontaneous charge polarizations, $V_{p}(r)$, were added to the 6 $\times$ 6 $\mathbf{k} \cdot \mathbf{p}$ RSP Hamiltonian in an iterative manner until convergence of the eigenstate energies was obtained. A detailed description of the model used for the calculation of the polarization dependent momentum matrix elements is presented in Ref. [9] and is summarized by the flow chart in Fig. 5. In the calculations, the $x$-, $y$-, and $z$-axes directions refer to the [1-100], [11-20] and [0001] crystallographic directions in our coordinate system. Various stresses ranging from pure uniaxial tensile stress (i.e., $\sigma_x = 0$ and $\sigma_y = 30$ kbar) to pure biaxial tensile stress (i.e., $\sigma_x = \sigma_y = 30$ kbar) were applied to the GaN/AlN QD model structure.

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The calculated $R_p$ and $\Delta E_r$ from the model are shown in Fig. 4(b) for a pure uniaxial stress of 30 kbar along [11-20] (i.e., $\sigma_x = 0$ and $\sigma_y = 30$ kbar). The results agree qualitatively with the experimental polarized CL results for $R_p$ and $\Delta E_r$ vs $<n>$ in Fig. 4 in which the $e^{-}\hbar$ beam was positioned near the center of a vertical stripe. For low temperatures, it is apparent that a minimal thermal excitation of holes results in an $R_p$ that is roughly independent of excitation until the first electron and hole levels are doubly occupied. Upon increasing the excitation ($I_D > 12$ nA and $<n> > 2$), a sudden decrease in $R_p$ is observed at low temperatures where a step is observed (downward arrows) for both the experiment and calculations in Figs. 4(a) and 4(b), consistent with the participation of excited hole states whose relative $p_z$-orbital character of the Bloch states also decreases [8,9]. In comparison, $R_p$ decreases gradually and nearly linearly as a function of $e^{-}\hbar$ occupation number, $<n>$, for $<n> \leq 2$ at $T = 300$ K, as observed in both the experi-
ment and calculations, owing to carrier filling and a thermal excitation of holes into higher energy QD hole states. Therefore, these results for the excitation dependence of the polarization anisotropy for GaN QDs in patterned stripes are consistent with similar results for GaN QDs excited in closed proximity (~0.5 μm) to microcracks [7-9], again confirming the uniaxial character and magnitude of the stress (~30 kbar) in the patterned stripe regions.

4 Conclusion In this work, we have selectively modified the thermal stress in GaN/AlN QD layers grown on Si(111) by etching a cross-hatched pattern into the as-grown sample using inductively coupled Cl2/Ar plasma reactive ion etching. The results show that a suitable choice of stripe width from ~2 to 10 μm and orientation can create regions of in-plane uniaxial stress that enable a selective and local control of the polarized luminescence from ensembles of QDs. With an e-beam excitation near the center of a stripe under a uniaxial tensile stress of ~30 kbar, the CL polarization anisotropy ratio \( R_p \) vanishes at high temperatures with an increasing excitation of the QDs, while the anisotropy decreases more slowly with excitation at low temperatures. The effects of screening of the polarization field in the QD and state-filling were studied as a function of e-beam current and the average carrier occupation number \( \langle n \rangle \) with CL and with a 3D 6×6 \( k \cdot p \) self-consistent calculation method. We attribute carrier filling and a thermal excitation of holes into higher energy hole states during excitation to account for a nearly linear decrease in the polarization anisotropy ratio, \( R_p \), with \( \langle n \rangle \) at T = 300 K, while almost no thermal excitation of holes occurs at the lowest temperatures in the calculations (T = 46 K). These results demonstrate the possibility of utilizing growth of III-nitride QDs on thermally mismatched Si as a method in strain engineering to create polarized solid-state light sources that could be incorporated into LCD display technologies.

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