Electronic and optical properties of GaN/AlN quantum dots on Si(111) subject to in-plane uniaxial stresses and variable excitation


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We have studied the excitation- and polarization-dependent optical properties of GaN/AlN self-assembled quantum dots (QDs) grown on Si(111) substrates. Ensembles of QDs were subject to various external stress configurations that resulted from the thermal expansion coefficient mismatch between the GaN/AlN layers and the Si(111) substrate and ranged from in-plane uniaxial stress, primarily along the (1120) directions, to in-plane biaxial stress, having magnitudes ranging from 20–30 kbar. Limited regions of uniaxial stress were obtained by exploiting naturally occurring microcracks that form during the postgrowth cooling. These microcracks act as stressors in order to create the highly localized regions of uniaxial stress. The local strain tensors for such QDs, which are subject to an interfacial stress perturbation, have been determined by modeling the dependence of the QD excitonic transition energy on the interfacial stress. Cathodoluminescence (CL) measurements of the excitonic transitions exhibit an in-plane linear polarization anisotropy in close proximity to microcracks. The polarization anisotropy is strongly dependent on the sample temperature and the electron beam excitation conditions used to excite the QD ensemble. Localized CL spectroscopy of the QDs exhibits emissions from both the ground and excited states, whose relative contributions depend on the level of excitation and temperature. Experimental results indicate that the polarization anisotropy vanishes at high temperatures (~300 K) with an increasing excitation of the QDs, while the anisotropy decreases more slowly with excitation at low temperatures (~60 K). A theoretical modeling of the effect 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 $6 \times 6 \mathbf{k} \cdot \mathbf{p}$ and effective mass methods for calculations of the $e-h$ wave functions and electron and hole quasi-Fermi levels for varying levels of state filling. We attribute carrier filling and a thermal excitation of holes into higher energy QD hole states during excitation to account for the observed gradual decrease in the polarization anisotropy with an increasing electron-hole pair excitation density at $T=300$ K.


I. INTRODUCTION

In the past decade, much attention has been directed toward GaN/AlN-based wurtzite self-assembled quantum dots (QDs) as a result of potential solid-state light emitting applications in the visible wavelength range. An important characteristic of group III-nitride compounds is the existence of a large polarization field, originating from both piezoelectric and pyroelectric polarizations. The charge polarization will create electric fields mainly along the [0001] QD growth axis that will be screened in a complex manner when electrons and holes begin to fill the excited energy levels of the QD during sufficient levels of excitation. In this work, vertically stacked layers of GaN/AlN self-assembled QDs were grown by the Stranski–Krastanov method on a Si(111) substrate using molecular beam epitaxy (MBE). During the subsequent cooling from growth temperatures, the thermal expansion coefficient mismatch between the Si substrate and GaN/AlN film containing multiple layers of QDs leads to an additional tensile stress at the Si/III-nitride interface, which is partially relaxed by the formation of microcracks that propagate parallel to the interface and along the (1120) directions. We have previously demonstrated that these defects serve as excellent stressors which can modify the strain tensor of an ensemble of QDs in close proximity (i.e., within a few microns) of the microcracks and lead to limited regions of in-plane uniaxial stress. The excitonic luminescence of QDs with a uniaxial stress perturbation exhibits an in-plane linear polarization anisotropy that depends on temperature.

The development of light emitting diodes (LEDs) with linear polarization is a topic of current interest for backlit liquid-crystal displays (LCDs). In LCDs, linearly polarized light is generated within an LCD pixel by passing unpolar-
ized light through a polarization filter, followed by a red, green, or blue color filter to create a color pixel source with a desired intensity. During the polarization filtering process $\sim 50\%$ of the light power is absorbed and dissipated as heat. Therefore, by utilizing highly polarized LED sources, a substantial savings in energy may be realized by eliminating need for the polarization filter. Polarized monochromatic light can be generated by InGaN-based LEDs grown on non-polar orientations.\textsuperscript{13,14} Optical intensity modulation of non-polar InGaN LEDs by an LCD system has been demonstrated.\textsuperscript{15} Therefore, a tailoring of the polarization properties of GaN/AlN QD-based light emitters could enable similar applications in LCDs.

In this study, we explore in detail the effects of localized stress perturbations in the GaN/AlN QD system. Using the method of cathodoluminescence (CL) spectroscopy, we excite locally groups or ensembles of vertically stacked QDs. The effects of screening of the polarization field in the QD, state-filling, and changes in the polarization anisotropy with varying excitation were studied both experimentally and theoretically by calculating changes in the electron and hole quasi-Fermi levels. In the calculations, we obtained three-dimensional (3D) self-consistent solutions 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 $e$-$h$ wave functions. While previous self-consistent calculations of nitride-based QDs have been reported, primarily theoretical results without extensive experimental measurements were presented in these works.\textsuperscript{5,16–18} In this paper, we present and compare our experimental results and theoretical calculations of the QD polarized emission for varying excitation densities and temperatures.

II. EXPERIMENT

The samples were grown by MBE using the two-dimensional (2D) to 3D Stranski–Krastanov growth mode transition.\textsuperscript{2,19,20} Two samples were grown on Si(111) substrates and consist of AlN (30 nm)/GaN (400 nm)/AlN (700 nm) buffer layers followed by either 40 or 85 layers of GaN QDs, labeled as samples S40 and S85, respectively. A schematic illustration of the sample structures for S40 and S85 is shown in Fig. 1(a). The growth of sample S40 (S85) involved 18 nm (6.7 nm) thick AlN barrier layers with 2.6 nm (1.6 nm) thick GaN QD layers, resulting in an average dot height of $\sim 5$ nm ($\sim 3.7$ nm), as determined previously by transmission electron microscopy (TEM) measurements for samples possessing very similar structures and growth conditions. The average volumes are also known from TEM and are $\sim 370$ nm$^3$ and $\sim 230$ nm$^3$, respectively, for samples S40 and S85. Both samples were terminated with a 40 nm thick AlN capping layer. The average dot density per QD plane is $\sim 5 \times 10^{11}$ cm$^{-2}$.

Our CL detection system is mounted on a JEOL 5910 scanning electron microscope (SEM). The samples were mounted on a variable temperature stage that is connected via a copper braid to a closed-cycle He cryorefrigerator. An ellipsoidal mirror with variable three-axis positioning collects luminescence emitted from the sample. The emitted luminescence is collected by the mirror and focused onto a coherent optical fiber bundle with a vacuum rotatable linear polarizer positioned before the fiber optics, as schematically illustrated in Fig. 1(b).\textsuperscript{10} Two polarization directions for the polarizer will be denoted with the subscripts $\perp$ and $\parallel$ to indicate detection orientations with $E$ perpendicular and parallel to a microcrack that is oriented along the [1120] crystallographic direction. The polarization anisotropy ratio, $R_p = I_{\parallel}/I_{\perp}$, is defined by the ratio of the integrated CL intensities, $I$, under the two orthogonal polarizer orientations and is given by $R_p = I_{\parallel}/I_{\perp}$. The light from the flexible fiber bundle was transferred to a 1/4 m monochromator outside the SEM vacuum system. The spectral resolution of the monochromator was 2 nm ($\sim 15$ meV) at $\lambda = 400$ nm (3,100 eV). The dispersed light was detected with a multialkali photomultiplier tube which enabled photon counting. Time-resolved CL experiments were performed with the method of delayed coincidence in an inverted single photon counting mode.\textsuperscript{21} Electron beam pulses of 50 ns width with a 1 MHz repetition rate were used to excite the sample.

III. THEORETICAL CALCULATIONS USING A 3D $\mathbf{k} \cdot \mathbf{p}$ METHOD

The electron and hole wave functions and energies were obtained with 3D $6 \times 6 \mathbf{k} \cdot \mathbf{p}$ calculations using the NEXTNANO\textsuperscript{5} quantum nanostructure simulation code.\textsuperscript{9,10,22}
The calculations utilized a 3D Schrödinger equation for wurtzite materials including strain, deformation potentials, spin orbit coupling, and piezoelectric and pyroelectric charges, the latter of which accounts for the large polarization field in the QD along the [0001] growth direction. In order to calculate the wave functions, a single band model for the electrons and a six-band \( k \cdot p \) Hamiltonian for the holes were employed. The coupling between the conduction and valence bands was neglected, owing to the size of the GaN and AlN bandgaps.23

We employed the material parameters used previously for calculations of the polarization field and eigenstates in GaN/AlN QDs.24–26 The fully strained GaN/AlN QDs were reported previously.10 External tensile stresses ranging from purely biaxial to uniaxial were employed to simulate the thermal stress in the AlN barrier layers in varying proximity to the microcracks.

IV. EXPERIMENTAL RESULTS AND DISCUSSION

A. Excitation-dependent Polarized CL Spectroscopy

The presence of (1120) oriented microcracks in the GaN/AlN/Si samples has been shown to create limited regions of an in-plane uniaxial stress within \( \Delta x \) of \(-0.5\) to \(1.0\) \(\mu m\) from the cracks.9–12 We have previously examined in detail the dependence of \( R_p (R_p = I_{p}/I_{b}) \) as a function of distance (\( \Delta x \)) from a microcrack, whose experimental geometry is illustrated in Fig. 1(b). We have shown that \( R_p \) decreases monotonically toward \( R_p = 1 \) as the e-beam is positioned at distances sufficiently far from the microcracks (\( \Delta x > 3 \) \(\mu m\)), for various temperatures.10–12 Again, the maximum value of \( R_p \) occurs for the lowest temperature, and this value decreases as the temperature increases. An increased uniaxial tensile stress near the microcrack and an increased biaxial tensile stress far from the microcrack are expected to occur as the temperature is reduced leading to an increased value of the polarization anisotropy at lower temperatures. The polarization anisotropy ratio \( R_p \) is a direct probe of deviations from biaxial symmetry.10,12

We have performed excitation-dependent local CL spectroscopy measurements by positioning the e-beam a distance \( \Delta x \) on a line along a direction perpendicular to a [1120]-oriented microcrack, as shown in Fig. 1(b). Stack plots of local CL spectra for sample S85 and S40 are shown in Figs. 2 and 3 for temperatures (\( T \)) of 300 K and 60 K, for e-beam currents (\( I_{b} \)) ranging from 0.05 to 27 nA, and for positions, \( \Delta x \), of 0.5 \(\mu m\) and 5 \(\mu m\), which represent regions of nearly pure uniaxial and biaxial tensile stresses, respectively, as labeled in the panels of Figs. 2 and 3. For regions of uniaxial stress (\( \Delta x = 0.5 \) \(\mu m\)) in S85, Fig. 2 shows that the maximum value of \( R_p \) increases from \(-1.5\) to 2.1 as the temperature decreases from 300 to 60 K. For sample S40, the polarization anisotropy is reduced in comparison to S85, but the trend is similar in the region of pure uniaxial stress as the maximum value of \( R_p \) increases from \(-1.3\) to 1.7 as the temperature decreases from 300 to 60 K, as shown in Fig. 3.

For the region where biaxial stress is predominant (\( \Delta x \) \(\geq 5 \) \(\mu m\)), \( R_p \) does not deviate far from 1 for both samples. The excitation dependent results for the polarization anisotropy ratio are summarized in Fig. 4, which show \( R_p \) as a function of \( I_{b} \) for temperatures of 60 and 300 K. Both experiment and theory are shown for comparison in Figs. 4(a) and 4(b). Of particular relevance is the large excitation dependence of \( R_p \) that is observed for temperatures of 300 K and yet noticeably diminished in the measurements at \( T = 60 \) K for S85 and S40. For S85 at \( T = 300 \) K, \( R_p \) is observed to decrease from \(-1.5\) to 1.0 nearly linearly with log...
known effect attributed to carrier filling in the QD in which the quantum confined Stark effect leads to an electric field dependent $e\cdot h$ oscillator strength. As the excitation density of the pulsed $e$-beam increases for large $I_b$, the initial larger screening of the field leads to an increased $e\cdot h$ oscillator strength and smaller decay time $\tau$ during the beginning of the decay transient. As the decay proceeds within a transient, the average screening gradually decreases, leading to a larger field, smaller oscillator strength and larger $\tau$ near the end of the transient. Thus, in order to obtain the carrier lifetime for a given steady state $e$-beam injection current, we utilize a linear fit to the initial portion of the transient resulting from a pulsed excitation at the same $I_b$ used for steady-state excitation, as shown in Fig. 5. We have employed a standard approach that determines the relationship between the average $e\cdot h$ occupancy in the QD, $\langle n \rangle$, and the electron beam current, $I_b$. In Fig. 6, we show the measured lifetime and $\langle n \rangle$ as a function of $I_b$ for S85 and S40.

C. Modeling of the excitation-dependent QD electronic states and optical transitions

In order to develop a theoretical model for our luminescence data, evaluate the screening of the electric field in the QDs, and calculate the excitation dependence of the polarized CL spectra and anisotropy ratio, $R_p$, we have expanded the multiband $\mathbf{k} \cdot \mathbf{p}$ and single band effective mass treatment for the occupation of multielectron and multi-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 $e\cdot h$ wave functions in the Hartree approximation were performed. The primary multicarrier correction to the energy levels results from the carrier-induced screening of the electric field in the QD which is caused by piezoelectric and pyroelectric polarization.

We calculate the electron and hole wave functions and energy levels while taking into account the modifications of the potential profile caused by the accumulation of carriers in the QDs. The modeling procedure is summarized with a flow chart in Fig. 7 which describes the procedure and order of
our self-consistent approach for solving the nonlinear Poisson-Schrödinger equation. The calculations for the wurtzite GaN QD system include the effects of strain, thermal stress, the microcrack-induced stress perturbation, deformation potentials, band offsets, and piezoelectric and spontaneous polarization fields to obtain the excitation-dependent eigenstates, electron and hole quasi-Fermi levels (\(\varphi_e\) and \(\varphi_h\)), and carrier occupation distribution among the QD electron and hole eigenstates. Solutions to the 6 \(\times\) 6 \(\mathbf{k}\cdot\mathbf{p}\) and effective mass Hamiltonians were obtained self-consistently. First, trial solutions of the \(e\)-\(h\) wave functions, \(\psi_e^0\) and \(\psi_h^0\), and energies, \(E_e^0\) and \(E_h^0\), for a given \(e\)-\(h\) occupation number \((n)\) were obtained in the usual manner by first obtaining solutions without the Hartree correction potential, \(V_p(\mathbf{r})\). Following the usual \(\mathbf{k}\cdot\mathbf{p}\) formalism, the electron and hole wave functions are expressed as

\[
\begin{align*}
|\psi_e^0\rangle = \sum_{k=1}^{2} F_{e,k}^0(\mathbf{r})|k\rangle \\
|\psi_h^0\rangle = \sum_{l=1}^{6} F_{h,l}^0(\mathbf{r})|l\rangle
\end{align*}
\]

in which the Bloch part of the electron wave function, \(|\psi_e^0\rangle\), is represented by two basis s-like states \(|S\rangle\) and \(|S\rangle\) and denoted by \(|k\rangle\). The Bloch parts of the hole wave functions, \(|\psi_h^0\rangle\), are represented by the six basis p-like functions, \(|l\rangle\), in the following representation: \(|x\rangle\), \(|y\rangle\), \(|z\rangle\), \(|x\rangle\), \(|y\rangle\), and \(|z\rangle\), where coordinates \(x\), \(y\), and \(z\) refer to the [1\bar{1}00], [11\bar{2}0], and [0001] crystallographic directions. For the hole state \(j\), \(\Phi_{ij}(\mathbf{r})\) are the six hole envelope functions that are obtained from the 3D 6 \(\times\) 6 \(\mathbf{k}\cdot\mathbf{p}\) method and \(\Phi_{eij}(\mathbf{r})\) are the two electron envelope functions for the electron state \(i\) obtained from the 3D single effective mass calculation. The next step is the calculation of the electron and hole quasi-Fermi levels, \(\varphi_e\) and \(\varphi_h\), for a given steady-state \(e\)-\(h\) pair occupation number, \((n)\). From the discrete delta-function nature of the zero-dimensional QD density of states (DOS) and by invoking charge neutrality requirements during \(e\)-\(h\) excitation at a temperature \(T\), \(\varphi_e\), and \(\varphi_h\) are obtained from solutions to

\[
(n) = \sum_{i=1}^{N} f(E_i^e - \varphi_e) = \sum_{j=1}^{N} f(\varphi_h - E_j^h),
\]

where \(f(E)\) is the Fermi–Dirac function for electrons and holes, \(N\) is the largest state number that was used in the calculations, and electron and hole state occupancy is given individually by \(n_i^e = f(E_i^e - \varphi_e)\) and \(n_j^h = f(\varphi_h - E_j^h)\), respectively. Electron and hole spin degeneracy was also taken into account.

FIG. 6. (Color online) The measured lifetime, \(\tau\), and average occupation number, \((n)\), for various e-beam injection currents, \(I_b\), for samples S85 (a) and S40 (b) in regions of uniaxial stress. The relation described in Ref. 27 was used to connect \((n)\) with \(I_b\). Dashed lines represent extrapolated values for \(\tau\) and \((n)\).
account in the calculations by assigning separate state numbers, \( i \) and \( j \), to each set of degenerate time-reversal conjugate states. The next step is the calculation of the excess carrier charge density for the electrons and holes which depend on the quasi-Fermi levels and temperature according to

\[
\rho_e(\vec{r}) = -e \sum_{j=1}^{N} f(E_i^{(e)} - \varphi_e)|\psi_i^{(e)}(\vec{r})|^2
\]

and

\[
\rho_h(\vec{r}) = e \sum_{j=1}^{N} [f(\varphi_h - E_j^{(h)})|\psi_j^{(h)}(\vec{r})|^2.
\]

(3)

The Hartree contribution to the potential, \( V_H(\vec{r}) \), is then readily calculated from Poisson’s equation as follows:

\[
\vec{\nabla}[\varphi_e(\vec{r}) \vec{\nabla}V_H(\vec{r})] = -\frac{1}{\varepsilon_0} [\rho_e(\vec{r}) + \rho_h(\vec{r})].
\]

(4)

The Hartree contribution is then added to the potential for charge polarization and the GaN/AlN band edge profiles, thereby enabling a series of iterative cycles involving the calculation of \( \psi_i^{(e)} \), \( \psi_j^{(h)} \), \( E_i^{(e)} \), \( E_j^{(h)} \), \( \varphi_e \), and \( \varphi_h \) for a given temperature, \( T \), and steady-state \( e-h \) occupation number, \( \langle n \rangle \). We have set the convergence conditions such that successive iterations must differ by less than 0.5 meV in the electron and hole energies.

The integrated intensity of polarized luminescence is determined by the carrier occupation in the QD and the electron-hole wave function overlap according to

\[
I_{\perp,\parallel} = \sum_{i,j} n_i^{(e)} n_j^{(h)} |M^{(i,j)}(n_i^{(e)}, n_j^{(h)})|^2,
\]

where

\[
M^{(i,j)}(n_i^{(e)}, n_j^{(h)}) = \sum_{k=1}^{2} \sum_{l=1}^{6} \int d^3r F_{ek}(\vec{r}) F_{hl}(\vec{r}) (k|\hat{e}_{\perp,\parallel} \cdot \hat{r}) |l\rangle.
\]

(5)

The unit electric field polarization vectors, \( \hat{e}_{\perp,\parallel} \), represent polarizations that are perpendicular or parallel to the [1120] microcrack directions, which we have defined as the \( y \)-axis direction in our coordinate system. The polarization anisotropy ratio, \( R_p \), is then calculated from Eq. (5) as

\[
R_p = \frac{I_{\perp}}{I_{\parallel}} = \frac{\sum_{i,j} n_i^{(e)} n_j^{(h)} |M^{(i,j)}(n_i^{(e)}, n_j^{(h)})|^2}{\sum_{i,j} n_i^{(e)} n_j^{(h)} |M^{(i,j)}(n_i^{(e)}, n_j^{(h)})|^2}.
\]

(6)

It is apparent from Eqs. (1) and (5) that the relative weights of the \( p_z \) and \( p_y \) orbitals, as determined by the relative values of the six hole envelope functions, \( F_{hl}(\vec{r}) \), will have an appreciable effect on the polarization anisotropy ratio \( R_p \).

We have calculated self-consistently the spectrum of electron and hole states using our self-consistent \( 6 \times 6 \) \( \mathbf{k} \cdot \mathbf{p} \) method. The energies of the electron and hole states, \( E_i^{(e)} \) and \( E_j^{(h)} \), are shown as a function of the average occupation number \( \langle n \rangle \) in Fig. 8 for S85 subject to a uniaxial stress of 30 kbar. As expected, the spectrum of hole states shifts downwards in energy with an average rate of \( \sim 15 \) meV and 9 meV, respectively, for S85 and S40, per addition of an \( e-h \) pair due to the partial screening of the electric field in the QD. A similar upward shift of \( \sim 8 \) meV and 5 meV, per \( e-h \) pair for electron levels also occurs, respectively, for S85 and S40, resulting in a net shift in the transition energy \( E_x \) that is \( \sim 23 \) meV and 14 meV per \( e-h \) pair, which is similar to changes observed in previous CL measurements.\(^{28}\) These energy shifts are obtained by averaging over the values obtained for \( \langle n \rangle \) ranging from 0.5 to 4 and 0.5 to 2 for S85 and S40, respectively. We note that differences in the average energy shift between the electron and hole states are due to differences in confinement and localization for both particles. We also note that the single band effective mass nature of the Schrödinger equation for the electrons and an approximately in-plane radially symmetric potential leads to well-defined \( s- \), \( p- \), and \( d- \) orbital symmetries and near-degeneracies, which are denoted by the groups of state numbers \( \{ l \} \) that are \( \{ 1, 2 \} \), \( \{ 3-6 \} \), and \( \{ 7-12 \} \), as observed in Fig. 8(a).\(^{29,30}\)

As the primary cause of the energy shifts is the Hartree term in the electrostatic potential, it is worth examining aspects of the calculation that lead to a partial screening of the [0001]-oriented polarization field of the QD, as shown in Fig. 9 for S85. Calculations were performed for the structures of S40 and S85 subject to uniaxial stresses of 20 and 30 kbar, respectively, along the direction of the [1120]-oriented microcrack. One-dimensional (1D) representations of the charge densities, \( \rho_{e,h}(z) \), in which the charge densities are averaged over the \( x-y \) plane, are shown in Fig. 9 for S85. The charge density for the holes is shown inverted in Fig. 9(b).
The charge densities are further shown overlayed on the 1D cross-section of the excitation-dependent band profiles for the conduction and valence band edges. As the occupation number \( n \) increases, the calculations show the expected screening of the band-edge potential and reduction in the average electric field along the [0001] QD axis. The reduced field leads to a broadening and shift in the centers of gravity for \( \rho_{e,h}(z) \), which are both toward the center of the QD, consistent again with the expected increase in the \( e\hbar \) oscillator strengths for increasing excitation conditions. The Hartree term in the potential, \( V_{B}(z) \), for a line along [0001] through the QD center, is also shown in Fig. 9(d) for various \( n \). The calculated average screening fields, \( F_s \), along [0001] for both the S85 and S40 structures are shown in Fig. 9(d) as a function of \( n \). These values are consistent with previous studies showing carrier induced screening in GaN/AlN QDs.\(^5\)\(^\text{28}\) The screening fields increase approximately linearly with \( n \), but exhibit small differences in slopes between the \( n \) values.

We have further calculated the excitation dependence of the CL spectra in order to compare with the measured energy shifts and polarization anisotropy. The CL spectra are modeled according to

\[
I(h\nu) = \sum_{i,j} |M_{ij}|^2 f\left(E_i^{(e)} - \phi_e\right)f\left(E_i^{(h)} - \phi_h\right)\delta(h\nu - E_i^{(e)} - E_i^{(h)})
\]

\[
= \sum_{i,j} \left|M_{ij}\right|^2 f\left(E_i^{(e)} - \phi_e\right)f\left(E_i^{(h)} - \phi_h\right)\delta(h\nu - E_i^{(e)} - E_i^{(h)}),
\]

where \( M_{ij} \) is the momentum matrix element, the Dirac \( \delta \)-function represents the electron and hole joint DOS, and \( h\nu \) is the emitted photon energy. The excitation-dependent momentum matrix element, \( M_{ij} \), is of the form expressed in Eq. (5) for polarized spectra with the electric field parallel or perpendicular to the microcracks or its square can be calculated by averaging over all three Cartesian directions, i.e.,

\[
|M_{ij}|^2 = \frac{\left(\langle |M_{ij}^{(x,x)}|^2 + |M_{ij}^{(y,y)}|^2 + |M_{ij}^{(z,z)}|^2\rangle\right)^{1/3}}{3},
\]

for nonpolarized CL spectra. In either case, \( M_{ij} \) is calculated from the wave functions resulting from the self-consistent calculations. We approximate the broadening of the Dirac \( \delta \)-functions with a Lorentzian function with homogenous width of 1 meV. The calculated nonpolarized CL spectra for a single QD subject to the uniaxial stress configuration at \( T=300 \) K are shown in Figs. 10(a) and 10(b) for the structures of S85 and S40 with various \( e\hbar \) occupation numbers \( \langle n \rangle \). Gradual shifts in the various excitonic transitions toward higher energy due to screening of the field along with an increased participation of higher energy transitions are observed with increasing \( \langle n \rangle \). Figures 10(c) and 10(d) show cases for calculated polarized
CL spectra of a single QD under occupations of $\langle n \rangle = 2$ and $\langle n \rangle = 4$ for S40 and S85, respectively. Although the ground state transition is polarized as expected, a reversal in the calculated energies by $\sim 200$ meV relative to the experimental peak energies for S40, these results represent a reasonable agreement between experiment and theory when considering the absence of the Coulomb correction and the large full-width-at-half-maxima of $\sim 650$ meV and $\sim 490$ meV for the CL spectral line shapes of the broadened QD ensembles for S85 and S40, respectively, as observed in Figs. 2 and 3.

D. Analysis of the excitation-dependence of the polarization anisotropy

Spatial variations in the optical polarization properties are attributed to the stress-dependent variations in the $p_x$ and $p_y$ characters of the valence band edges and the ground state hole wave function. In Fig. 12, the effect of a variable external uniaxial stress ($\sigma_{xx} = 0$) on the $e/h$ isosurfaces are shown for the structure of S85 is shown with $\sigma_{yy} = 0$ kbar, 2.5 kbar, 10 kbar, and 30 kbar in panels (a)–(d), respectively, for the non-self-consistent case of $\langle n \rangle = 0$. Isosurfaces of the ground-state hole $e/h$ wave function (with $|\psi_{e,h}(\vec{r})|^2 = a_{e,h}|\theta_{\text{max}}|^2$, where $|\theta_{\text{max}}|^2$ is the maximum probability density of the wave function) for the S85 structure are shown with $a_e = 0.01$ and $a_h = 0.1$ so as to distinguish more easily between the two states. It is apparent that the s-like electron state retains its near cylindrical symmetry for all uniaxial stress values shown. However, the shape and direction of elongation of the hole isosurface changes in Fig. 12 as the value of $\sigma_{yy}$ is increased toward its maximum value of 30 kbar which was used in our calculations. Similar results were observed for calculations for the structure of S40. Figures 12(e) and 12(f) show 3D plots of the $e/h$ isosurfaces for S85 with a uniaxial stress ($\sigma_{xx} = 30$ kbar and $\sigma_{yy} = 0$) and a biaxial stress ($\sigma_{yy} = \sigma_{zz} = 30$ kbar), respectively, illustrating the shape of the QD and spatial separation of the $e/h$ wave functions along [0001]. Previously, we showed that a change from biaxial to uniaxial stress alters the admixture of $p_x$ and $p_y$ characters of the band edges and ground state hole wave function, changes the shape and direction of elongation of the hole isosurfaces, and accounts well for the subsequent
anisotropy in the ground state polarization dependent optical transitions and its spatial variation in the vicinity of the microcracks. Thus, this same interpretation ought to apply for the case of a variable uniaxial stress. Indeed, a calculation of \( R_p \) versus \( \alpha_{yy} \) for the present case is shown for both S40 and S85 structures in Fig. 12(g), illustrating the connection between hole isosurface orientation, admixture of \( p_x \) and \( p_y \) characters, and the resulting \( R_p \).

The striking temperature dependent behavior of \( R_p \) in Figs. 2–4 suggest thermal excitation to higher lying hole states plays an important role in reducing \( R_p \) for higher excitation conditions at room temperature. Since the energy spacing between confined electron states is \( \sim 4–5 \) times greater than the spacing for hole states, which have spacings typically ranging from 10–20 meV, we hypothesize that the thermal excitation for holes near \( T=300 \) K will have an appreciable effect on the distribution of hole occupancy, particularly for high excitation conditions (i.e., \( \langle n \rangle > 1 \)).

It is possible to observe the position and stress dependent character of the wave function by projecting the two Bloch states with like spin, \( |x\rangle \) and \( |y\rangle \) onto the hole wave function by calculating \( f_{WF} = |\langle x|\psi|y\rangle|^2 \) where \( i=x \) or \( y \). We note that \( y \) is referenced as the \([1120]\) microcrack direction in the calculations. We show \( f_{WF} \) for the lowest four hole states as a function of distance, \( \Delta x \), from a microcrack in Fig. 13. The relative \( p_x \) orbital character of the Bloch states, \( f_{WF} \) reduces significantly for higher energy hole states (i.e., for \( j \geq 2 \)) for uniaxial stress (\( \Delta x=0 \)) and approaches \( \sim 0.5 \) for biaxial stress (\( \Delta x=3 \) \( \mu \)m). The present case in Fig. 13 is illustrated for \( \langle n \rangle = 0.5 \), but also shows similar results for larger \( \langle n \rangle \). The case of \( \langle n \rangle = 0.5 \) is important since it is a self-consistent calculation for \( E_{h} \) that reflects the lowest energy excitonic transition in Slater’s ground state transition and should therefore most closely represent the experimental ground state \( e-h \) transition for low excitation conditions. The inset of Fig. 13 further shows the calculation of \( R_p \) versus \( \Delta x \), confirming that \( R_p \) reduces from \( \sim 1.7 \) to \( \sim 1 \) in transitioning from uniaxial to biaxial stress.

Thus, we expect that the polarization anisotropy ratio \( R_p \) will decrease for optical transitions that involve increasingly higher energy hole states, as obtained by increasing \( e-h \) occupation numbers, \( \langle n \rangle \), or an enhanced thermal excitation to higher energy hole states for \( T=300 \) K. Using the spectrum of hole wave functions in S40 and S85 that were calculated self-consistently, we have calculated \( R_p \) at high and low temperatures (300 and 5 K) using Eq. (6) by taking into account the occupation of the excited holes using Fermi–Dirac statistics in a fully self-consistent fashion involving separate electron and hole occupation numbers, \( n^e \) and \( n^h \). Using this method, the calculated \( R_p \) versus \( \langle n \rangle \) is shown for both structures in Fig. 4(b) at \( T=5 \) and 300 K, in order to consider the extreme temperature limits involved with thermal excitation of the hole states. The results agree qualitatively with the experimental polarized CL results for \( R_p \), as shown in Fig. 4(a). For low temperatures, it is apparent that a minimal thermal excitation of the hole states 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 (\( I_{ex} \) \( \sim 12 \) nA), a sudden decrease in \( R_p \) is observed at low temperatures where a step is observed (downward ar-
rows) for both samples in Fig. 4(a). Such a decrease in $R_p$ at higher excitations is consistent with the participation of higher energy hole states whose relative $p_z$-orbital character of the Bloch states, $f_{WFi}$, also decreases, as observed in Fig. 13. In comparison, $R_p$ decreases gradually and nearly linearly as a function of $\langle n \rangle$ at $T=300$ K, as observed in Fig. 4 for both the experiment and calculations for $S40$ and $S85$. Thus, 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 $R_p$ with $\langle n \rangle$ at $T=300$ K, while almost no thermal excitation of holes occurs at the lowest temperatures in the calculations ($T=\pm 5$ K).

V. CONCLUSION

In this work, GaN/AlN self-assembled QDs were grown by the Stranski–Krastanov method on a Si(111) substrate using MBE. During the subsequent cooling from growth temperatures, microcracks formed parallel to the interface and mainly along the $\langle 11\overline{2}0 \rangle$ directions. We demonstrated that these defects serve as excellent stressors, providing limited regions of in-plane uniaxial stress which can be exploited to perturb the electronic states of the QDs. The excitonic luminescence of QDs with a uniaxial stress perturbation exhibits an in-plane linear polarization anisotropy. Localized CL spectroscopy of the QDs exhibits emissions from both the ground and excited states, whose relative contributions depend on the level of excitation and temperature. We have studied these emissions using polarization-resolved CL for ensembles of QDs. The effects of screening of the polarization field in the QD and state-filling were studied experimentally with CL and theoretically with a 3D $6 \times 6 \mathbf{k} \cdot \mathbf{p}$ self-consistent calculation method for solving the Schrödinger and the Poisson equations. In the modeling, we examined occupation of the excited QD electron and hole states, determined the quasi-Fermi levels, and calculated the excitation and uniaxial stress dependence of the CL polarization anisotropy. Experimental results indicate that 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. 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 ($T=\pm 5$ K). These results demonstrate the complex interplay between external stress, temperature, and excitation conditions on the polarization dependence of the excitonic luminescence from an ensemble of GaN/AlN self-assembled QDs.

27The average carrier density is represented by $\langle n \rangle = (E_F + E_D)/3(\epsilon E_V / \epsilon A_d)\rho$, where $\rho$ is the density of state CL of the quantum dot and $V_e$ is the excitation volume of the $E_e = 15$ keV electron beam. We have used $V_e = 2.1 \mu$m³ for the present GaN/AlN material system.