Efficient Fourier space quantum dot k p for wurtzite systems including smooth alloy profile and spatially varying elastic and dielectric constants

Cite as: J. Appl. Phys. **129**, 224301 (2021); https://doi.org/10.1063/5.0051133 Submitted: 22 March 2021 . Accepted: 16 May 2021 . Published Online: 08 June 2021

🔟 Luc Robichaud, and ២ Jacob J. Krich



ARTICLES YOU MAY BE INTERESTED IN

Multiscale simulations of the electronic structure of III-nitride quantum wells with varied indium content: Connecting atomistic and continuum-based models Journal of Applied Physics **129**, 073104 (2021); https://doi.org/10.1063/5.0031514

Rational construction of staggered InGaN quantum wells for efficient yellow light-emitting diodes

Applied Physics Letters 118, 182102 (2021); https://doi.org/10.1063/5.0043240

Perspective on the future of silicon photonics and electronics Applied Physics Letters **118**, 220501 (2021); https://doi.org/10.1063/5.0050117





J. Appl. Phys. **129**, 224301 (2021); https://doi.org/10.1063/5.0051133 © 2021 Author(s).

scitation.org/journal/jap

and nanostructures. Both approaches rely on elastic, dielectric, and electronic parameters, which vary spatially in QD structures due to

the changing alloy content. Essentially, all methods calculate the

continuously varying local strain field and its impact on the elec-

tronic structure through deformation potential and piezoelectric

couplings. Frequently, calculations assume sharp material interfaces

and uniform alloy content in the dot and host regions; they addi-

tionally assume that the elastic and dielectric constants are spatially

uniform.^{7,8} Continuous spatial dependence of these material param-

eters is readily included in real-space methods, including both TB

and $\mathbf{k} \cdot \mathbf{p}$. Strain, even with spatially varying elastic constants, can

be calculated using real-space finite element methods for continuum

elastic models^{7,9,10} and with valence force fields in atomistic

models.^{7,11,12} Smooth variation of the alloy fraction can also be

ARTICLE

Efficient Fourier space quantum dot k · p for wurtzite systems including smooth alloy profile and spatially varying elastic and dielectric constants

Cite as: J. Appl. Phys. 129, 224301 (2021); doi: 10.1063/5.0051133 Submitted: 22 March 2021 · Accepted: 16 May 2021 · Published Online: 8 June 2021

Luc Robichaud^{1,a)} 🕩 and Jacob J. Krich^{1,2} 🕩

AFFILIATIONS

¹Department of Physics, University of Ottawa, Ottawa K1N 6N5, Canada ²School of Electrical Engineering and Computer Science, University of Ottawa, Ottawa KIN 6N5, Canada

^{a)}Author to whom correspondence should be addressed: Irobi019@uottawa.ca

ABSTRACT

We present methods to calculate the electronic structure of wurtzite quantum dot systems with continuous alloy profiles within Fourierspace-based $\mathbf{k} \cdot \mathbf{p}$ theory. We incorporate spatially varying elastic and dielectric constants in strain and piezoelectric potential calculations. A method to incorporate smooth alloy profiles in all aspects of the calculations is presented. We demonstrate our methodology for the case of a 1-dimensional InGaN quantum dot array and show the importance of including these spatially varying parameters in the modeling of devices. We demonstrate that the convergence of the lowest bound state energies is for good approximation determined by the largest wave vector used in constructing the states. We also present a novel approach of coupling strain into the $\mathbf{k} \cdot \mathbf{p}$ Hamiltonian, greatly reducing the computational cost of generating the Hamiltonian.

Published under an exclusive license by AIP Publishing. https://doi.org/10.1063/5.0051133

I. INTRODUCTION

Given their large range of bandgaps, from 0.78 to 3.51 eV, InGaN materials have attracted attention in applications such as LEDs, single-photon emitters, water splitting, and solar cells.^{1–5} For any application, device performance depends on having an electronic structure well tuned to its target application. Since the electronic structure of quantum dots (QDs) can be dramatically changed by varying their size and composition, they can be quite attractive for applications. InGaN QD structures grown by molecular beam epitaxy (MBE) have significant In diffusion,¹ leading to a smoothly varying alloy profile without a sharp material interface. This indium diffusion results in a material whose elastic and electrical parameters vary with the local alloying fraction. Quantum dots with intentionally graded indium fractions have also been of interest.⁶ Given the electronic structure's sensitivity to composition, accurate alloy profiles are crucial for accurate modeling.

Tight binding (TB) and $\mathbf{k} \cdot \mathbf{p}$ theory are standard approaches for calculating single-particle electronic structures for bulk materials

included. 10 However, both TB and real-space $\boldsymbol{k}\cdot\boldsymbol{p}$ have important limitations. Tight binding methods can be accurate but become expensive when considering systems with a large number of atoms,¹³ though linearly scaling methods extend their reach for an increasing number of systems.¹⁴ The $\mathbf{k} \cdot \mathbf{p}$ method is not atomistic





and gives a good balance between accuracy and computational requirements for mid-to-large dots.¹⁵ Its real-space implementations using the finite difference method^{9,10} can be successful, but they can suffer from spurious midgap states arising from finite difference formulae effectively sampling bulk states at large wave vectors where $\mathbf{k} \cdot \mathbf{p}$ breaks down, especially at high alloy fractions.^{16,17}

Fourier space approaches to $\mathbf{k} \cdot \mathbf{p}$ have been widely used with good success in modeling the electronic structure of QDs.^{8,18–24} They provide an excellent compromise between computational cost and accuracy, allowing consideration of nanoscale structures beyond the scale of TB. Fourier space $\mathbf{k} \cdot \mathbf{p}$ methods give the direct control of wave-vector sampling, making it easy to reject the spurious states.^{8,19}

A hybrid approach using real-space parametrization and fast-Fourier-transform-based strain and Hamiltonian calculations allows rapid determination of band-edge energies in nanostructures, including piezoelectric potentials and smoothly varying alloy profiles.^{6,23,24} Each eigenstate must be found independently, making this approach particularly good when interested in the lowest electron and hole states or in small dots that only contain a few bound states. The traditional Fourier space approach, as taken from Refs. 8 and 18-22, produces a non-sparse Hamiltonian, which is diagonalized to obtain a full eigenvalue spectrum, making it useful when interested in properties such as optical absorption. References 18-22 reduced the computational cost of this approach by using symmetry adapted bases that block diagonalize the Hamiltonian.^{18,20,25} This latter Fourier space-based $\mathbf{k} \cdot \mathbf{p}$ method has only been used with sharp material interfaces, spatially uniform elastic and dielectric constants, and spatially uniform dot and host alloy fractions. References 8, 25, and 26 used a continuum elastic Green's function method for calculating strain. These works assumed spatially uniform elastic and dielectric constants for both the host and dot materials, which was justified for the sizes of their InAs/GaAs and GaN/AlN QD systems but would not be correct for larger dots. Reference 8 presented a method to include different host and dot material elastic constants but did not use it.

In isolated QDs, it is computationally challenging to calculate strain effects since strain generally decays more slowly than bound state wavefunctions. The unit cell must then be chosen large enough to resolve the strain decay, increasing the number of plane waves that must be calculated to resolve the electronic structure, at considerable computational expense. References 20 and 27 presented a more efficient approach that uses two different unit cells: one for the electronic structure and one for strain. This method allows for the modeling of the electronic structure and strain but introduces some complexity in calculating the Hamiltonian, which requires the calculation of multiple composed convolutions on different Fourier space meshes. These convolutions can be computationally costly depending on the sizes of meshes needed for convergence.

In this article, we show the methods for and the importance of including two different sources of spatial variation in Fourier space $\mathbf{k} \cdot \mathbf{p}$ calculations for wurtzite InGaN systems: first, spatially varying elastic and dielectric constants and second, smooth alloy profiles, as are found in experimental devices. Our approach efficiently includes smooth alloy profiles in the strain, piezoelectric potential, and electronic structure calculations. Smooth indium profiles both increase

the accuracy of the simulations for simulating experimental devices and decrease their computational cost by removing the abrupt interfaces that otherwise require large numbers of plane waves to obtain convergence. The spatial variations of elastic and dielectric constants affect the calculation of strain and piezoelectric potentials, and we adapt the calculations presented in Ref. 8 to include the spatially varying dielectric constant. We also present a method to improve the efficiency of calculating the strain in using the two-unit-cell method for isolated QDs. By fixing the strain unit cell to be commensurate with the electronic unit cell, we present an approach that reduces the number of needed convolutions, significantly reducing the computational cost.

We demonstrate our methodology by calculating the electronic structure for a 1D array of InGaN QDs, modeling devices grown as LEDs and used for water splitting.^{1,3} In this example, we show the importance of the inclusion of spatially varying elastic and dielectric constants and smooth indium profiles for accurate electronic structures. We also show that the most important criterion for the convergence of the lowest QD electron and hole energies is the maximum wave vector included in the Fourier space sampling, which can be increased with low computational cost by using a small unit cell.

Section II contains strain and piezoelectric potential calculations using spatially varying elastic and dielectric parameters. Section III presents the $\mathbf{k} \cdot \mathbf{p}$ model used for electronic structure calculations and our novel approach to efficiently include strain through choices of unit cells. Section IV introduces a method to use smooth indium profiles in all aspects of our calculations. Section V demonstrates our entire methodology for the case of a 1D QD array, such as QDs grown inside the nanowires.¹

II. SPATIALLY VARYING ELASTIC AND PIEZOELECTRIC CONSTANTS

We begin by considering QD heterostructures with abrupt changes in alloy fraction. Alloying the host material changes the local lattice constants, leading to a lattice mismatch at the host and dot material boundary. This lattice mismatch is a source of strain throughout the QD system, affecting the electronic states of the system. For example, InN has a larger lattice constant than GaN, so alloying GaN with indium to form a QD produces strain in and near the QD. The propagation of that strain is determined by the elastic constant, which itself also varies with alloy fraction. Additionally, strain can generate strong piezoelectric potentials in materials such as III-nitrides. The piezoelectric potential in III-nitrides is particularly important along the c-axis and can be strong enough to spatially separate electron and hole states through the quantum-confined Stark effect.²⁸

In prior Fourier-space-based work, elastic and dielectric constants are largely assumed to be spatially uniform in Fourier-based calculations of strain and the piezoelectric potential. Reference 8 presented a method to include differing host and dot elastic constants but did not use it in their calculations. They argued that the system's elastic constant largely depends on the nearest neighbor lattice spacing, which is strongly modified inside small dots from bulk values. They therefore used the spatially uniform elastic constant of the host material. In our case, we consider larger dots in which the lattice constant in the center of the dots can be significantly different from that in the barrier material, which implies that the local elastic constants should also be different from those in the barrier material. Having the local elasticity depend on local strain would make a computationally challenging nonlinear problem. Instead, we implement the method presented in Ref. 8 to include differing elastic constants using the bulk elastic constants of alloys at each location. We compare those results to those obtained from using spatially uniform elastic constants, to highlight the differences in strain-induced dot energy levels. For the piezo-electric potential, we use a procedure similar to Ref. 8, but we also include spatially varying dielectric constants. The strain field and the piezoelectric potential are coupled into a $\mathbf{k} \cdot \mathbf{p}$ model, as presented in Sec. III, for electronic structure calculations.

A. Quantum dot system

We consider a superlattice of wurtzite QDs embedded in a bulk host material, as shown in Fig. 1(a). InGaN QDs such as those described in Ref. 1 have a lens-like shape and do not have a sharply defined boundary. We approximate these QDs as being either cylindrical or hexagonal prisms. These choices of dot geometries simplify calculations, as described in Sec. II B, and preserve the $C_{6\nu}$ symmetry of the material, which we take advantage of in Sec. III for electronic structure calculations. Hexagonal periodic boundary conditions are used to also preserve the material's $C_{6\nu}$ symmetry. For single-dot calculations, the superlattice unit cell must be large enough that the choice of cell size does not affect results. For actual QD arrays, we consider only hexagonal superlattices in the plane.

In this periodic system, the real space QD superlattice is defined by the set of lattice vectors \mathbf{L}_i , as shown in Fig. 1(a). We denote the real space unit cell by $\Omega_{\mathbf{e}}^{-1}$. The index "e" indicates that these quantities relate to the electronic cell, as opposed to the strain unit cell, which is introduced in Sec. II B. Imposing periodic conditions in real space implies a discrete reciprocal space with wave vectors

$$\mathbf{q} = i_1 \mathbf{b}_1 + i_2 \mathbf{b}_2 + i_3 \mathbf{b}_3, \quad i_1, i_2, i_3 \in \mathbb{Z},$$
(1)



FIG. 1. (a) Unit cell of the cylindrical QD superlattice and its basis vectors. Dashed lines show the unit cell boundaries of the QD superlattice. White regions are the host material and gray regions are the QDs. (b) Hexagonal prism QD.

where \mathbf{b}_i are the reciprocal basis vectors, similar to that in Ref. 25. Due to the symmetry of the system, we have $L_1 = L_2$, which we define as L_{12} . In our reciprocal-space calculations, we sample on sets of the wave vectors $\mathbf{q} \in \Omega_e^{-1}$. We define m_{12} and m_3 such that $i_1, i_2 = \{-m_{12}, \ldots, 0, \ldots, m_{12}\}$ and $i_3 = \{-m_3, \ldots, 0, \ldots, m_3\}$. This sampling produces a hexagonal mesh of size $N = N_1 N_2 N_3$, where $N_i = 2m_i + 1$. To obtain a C_6 symmetric mesh, we remove points such that $|q_x| > m_{12} \frac{2\pi}{L_1}$, leaving a mesh whose size we denote by N_e .

By choosing the unit cell dimensions L_i large enough, it is possible to remove electronic coupling between neighboring dots. This flexibility allows us to model 3D, 2D, and 1D arrays of coupled dots. The isolated dot case can also be obtained by choosing both L_{12} and L_3 sufficiently large. Section II B presents a method that also uncouples dots in terms of strain, which is based on calculating strain and the electronic structure using different unit cells.

We illustrate the methods presented in this article by modeling a QD system inspired by Ref. 1. That system consists of InGaN dots grown in GaN nanowires. We approximate this system as an infinite 1D QD array by choosing L_3 to match the measured dot-dot spacing and L_{12} large enough to avoid dot-dot coupling, but we do not consider the nanowire itself and instead embed the dots in bulk GaN. We choose the dot indium alloy fraction, radius, and height based on the experimental device. System parameters are listed in Table I, and material parameters are in Appendix A. In some instances, wurtzite QD possesses a clear hexagonal geometry.²⁹ Therefore, we consider both cylindrical and hexagonal prism QDs, as shown in Fig. 1(b). The hexagonal dots have dimensions chosen to give an area of πR^2 , preserving the volume of the cylindrical dots. In all of the cases studied, there is only a small difference between the circular and hexagonal dots, and we show the circular ones in most results. We illustrate the small differences in Sec. V. All results are for the cylindrical case unless stated otherwise.

B. Strain

In this section, we present how we calculate strain with elastic constants that depend on alloy fraction for 3D, 2D, and 1D QD superlattices and isolated dots. Our method follows from

TABLE I. Default quantum dot superlattice parameters used unless specified otherwise.

Parameter	Value	
X_0	0.45	
h	40 Å	
R	200 Å	
L_{12}	500 Å	
L_3	70 Å	
m_{12}	10	
m_3	4	
<i>n</i> ₁₂	6	
n_3	1	
δ	[1.5, 1.5, 2.5] Å	

Refs. 8 and 20. We calculate the strain produced by a single isolated dot and construct the QD superlattice strain by linear superposition.

The calculated strain is to be coupled into the electronic structure calculations. However, strain decays considerably slower than bound electronic wavefunctions. In the case of isolated dots, the unit cell must be large enough to accommodate the strain decay. Choosing a unit cell large enough to capture the strain decay reduces the maximum wave vector attainable when using a fixed number of plane waves. As we demonstrate in Sec. V, accurately describing the electronic states requires using sufficiently large wave vectors, and thus, a large unit cell requires a large number of plane waves. Following Ref. 20, we consider that the electronic model and strain model each have their own real space unit cells. This additional degree of freedom allows accurate and computationally efficient determination of both electronic structure of rapidly decaying confined QD states and longer-range strain effects in isolated dots. In the case of a QD superlattice, different realspace electronic and strain unit cells are not required.

In prior work, lattice-mismatch-driven strain has been calculated for a single dot using a continuum theory with Green's function approach while assuming spatially uniform elastic constants.^{8,26,30} We briefly present the method to include spatially varying elastic constants from Appendix A of Ref. 8, correcting typos in Eqs. (A7) and (A8). We show how the spatially varying elastic constants modify strain and how this modified strain changes the piezoelectric potential in Sec. II C. We show that the elastic constant correction is necessary to obtain accurate strain and piezoelectric potentials.

Reference 8 showed that in the case of differing host and dot elastic constants, λ_{ijmn}^{h} and λ_{ijmn}^{d} , respectively, the strain tensor ϵ_{ij} can be written in Fourier space as

$$\tilde{\boldsymbol{\epsilon}}_{lm}(\mathbf{q}) = \boldsymbol{\epsilon}_{lm}^{\mathsf{T}} \tilde{\boldsymbol{\chi}}_{\mathsf{d}}(\mathbf{q}) + \tilde{\boldsymbol{\epsilon}}_{lm}^{\mathsf{c}}(\mathbf{q}), \qquad (2)$$

$$\boldsymbol{\epsilon}_{lm}^{\mathsf{T}} = \boldsymbol{\varepsilon}_{\mathsf{a}} \delta_{lm} + \boldsymbol{\varepsilon}_{\mathsf{ca}} \delta_{l3} \delta_{m3},$$

where the first term in Eq. (2) is the initial strain due to lattice mismatch and the second term is lattice relaxation due to elasticity. Here, $\varepsilon_{a} = (a^{h} - a^{d})/a^{d}$, $\varepsilon_{c} = (c^{h} - c^{d})/c^{d}$, and $\varepsilon_{ca} = \varepsilon_{c} - \varepsilon_{a}$, and the tildes indicate Fourier-space quantities as described in Appendix B. $\tilde{\chi}_{d}$ is the Fourier transform of the characteristic function of the dot, which is given in Appendix C. a^{h} and c^{h} are the lattice constants of the host material and a^{d} and c^{d} are of the dot material. More specifically, a is the xy-plane lattice constant and c is the lattice constant along the z-axis. $\tilde{\epsilon}^{c}_{lm}(\mathbf{q})$ is then written as a power series

$$\tilde{\boldsymbol{\epsilon}}_{lm}^{\mathsf{c}}(\mathbf{q}) = \tilde{\boldsymbol{\epsilon}}_{lm}^{(0)}(\mathbf{q}) + \tilde{\boldsymbol{\epsilon}}_{lm}^{(1)}(\mathbf{q}) + \tilde{\boldsymbol{\epsilon}}_{lm}^{(2)}(\mathbf{q}) + \cdots, \qquad (3)$$

with $\tilde{\boldsymbol{\epsilon}}_{lm}^{(N)}(\mathbf{q}) \propto \left(\frac{\Delta \lambda}{\lambda}\right)^N$, $\Delta \lambda_{ijmn} = \lambda_{ijmn}^{\mathsf{h}} - \lambda_{ijmn}^{\mathsf{d}}$, and the condition $\frac{\Delta \lambda}{\lambda} \ll 1$ ensures the convergence of the series. The leading term $\tilde{\boldsymbol{\epsilon}}_{lm}^{(0)}$ corresponds to uniform elastic constants of the dot with each subsequent term being a correction to include spatial variations due to the alloy profile. Using the Einstein summation convention, each

term has the form

$$\tilde{\boldsymbol{\epsilon}}_{lm}^{(N)}(\mathbf{q}) = \frac{(2\pi)^3}{2} \Big[F_p^{(N)}(\mathbf{q}) q_l \tilde{\boldsymbol{G}}_{mp}^{\mathsf{h}}(\mathbf{q}) + F_p^{(N)}(\mathbf{q}) q_m \tilde{\boldsymbol{G}}_{lp}^{\mathsf{h}}(\mathbf{q}) \Big], \qquad (4)$$

where

$$F_{i}^{(0)}(\mathbf{q}) = -\lambda_{ikpr}^{\mathsf{d}} \boldsymbol{\epsilon}_{pr}^{T} q_{k} \tilde{\boldsymbol{\chi}}_{\mathsf{d}}(\mathbf{q}),$$
(5)

$$F_{i}^{(N)}(\mathbf{q}) = -\Delta\lambda_{iklm}q_{k}\frac{(2\pi)^{3}}{V}\sum_{\mathbf{q}'}\tilde{\chi}_{\mathsf{d}}\left(\mathbf{q}-\mathbf{q}'\right)\tilde{\boldsymbol{\epsilon}}_{lm}^{(N-1)}\left(\mathbf{q}'\right), \quad (6)$$

where Eqs. (4) and (6) are corrected from Ref. 8 and \tilde{G}_{in}^{h} is Green's tensor for the host material.

We compare the strain corrected at various orders according to Eq. (3) to the usually considered case of uniform elastic constants of the host material. Figure 2 shows the convergence of the strain corrections for the 1D QD array system described in Sec. II A. We quantify convergence with the following metric for the norm of the strain:

$$|\tilde{\boldsymbol{\epsilon}}| = \sqrt{\sum_{m \ge l} \frac{V}{(2\pi)^3}} \int d\mathbf{q}^3 \left| \tilde{\boldsymbol{\epsilon}}_{lm}(\mathbf{q}) \right|^2}, \tag{7}$$

where $m \ge l$ indicates the sum of the unique elements of the strain tensor ($\tilde{\epsilon}_{11}$, $\tilde{\epsilon}_{22}$, $\tilde{\epsilon}_{33}$, $\tilde{\epsilon}_{23}$, $\tilde{\epsilon}_{13}$, and $\tilde{\epsilon}_{12}$). The green line in Fig. 2 compares $\tilde{\epsilon}$ when calculated from Eq. (4) to $\tilde{\epsilon}^{\text{GaN}}$, which is calculated assuming spatially uniform elastic constants of GaN. Blue line shows the self-convergence of the power series in Eq. (3). From these results, we conclude that a second order correction is sufficient to have strain converged within 1% in self-convergence and that this converged strain differs from the uniform case by about 6%,



FIG. 2. Convergence of the strain due to spatially varying elastic constants with perturbation order for a 1D QD array as described in Table I. Green line is the relative difference between the corrected strain and the strain with uniform λ of GaN. The zeroth term is the case of uniform λ of InGaN with alloy fraction of the dot. Converged strain has 6% relative difference from the case of λ^{GaN} , indicating that the corrections are necessary for accurate strain fields. Blue line is the magnitude of each term in Eq. (3) as a fraction of the zeroth term. Correction magnitudes are less than 1% starting from second order.



FIG. 3. Hydrostatic strain for increasing correction orders in the elastic constants. Black dashed line is strain assuming uniform elastic constants of the host material λ^{GaN} while solid lines are for spatially varying elastic constants with correction order $\lambda^{(N)}$. As the correction order $\lambda^{(N)}$ increases, the strain moves toward the uniform case λ^{GaN} . Note that the lines for $\lambda^{(2)}$ and $\lambda^{(3)}$ are overlapping. However, the uniform case λ^{GaN} underestimates the strain in the dot.

indicating that the elastic constant corrections produce significant changes in strain fields of InGaN systems. In Sec. II C, we show that the calculated piezoelectric potential remains essentially unchanged from third-order corrections and up, consistent with Fig. 2. Given that including these corrections is not computationally costly, we have included third-order corrections in all of our calculations unless stated otherwise.

Figure 3 shows the hydrostatic strain, $\epsilon_{hydro} = \epsilon_{xx} + \epsilon_{yy} + \epsilon_{zz}$, along a cut through the axis of the dot, showing relaxation of strain inside the dot with each additional correction. For this sample dot, the bulk lattice constants are 5% larger in the dot than in the barrier, and the compressive strain near the center ($\epsilon_{zz} \approx -0.012$, $\epsilon_{xx} \approx -0.028$) produces local lattice constants that are closer to the bulk InGaN values than bulk GaN, which is consistent with the choice to model spatially varying elastic constants.

The strain produced by the QD superlattice can be obtained from the linear superposition of the single-dot strain. However, we want the ability to study dots that are completely uncoupled, both electronically and from strains of the fictitious periodic array. References 20 and 27 proposed a method to allow the simultaneous treatment of a large unit cell for the strain problem and a small unit cell for the electronic problem, which together allow isolated dots to be considered in a computationally tractable manner. In this case of two independent cells, the strain is calculated in its own real space unit cell Ω_s with volume V_s . We denote the strain reciprocal unit cell as Ω_s^{-1} such that it contains the wave vectors \mathbf{Q} , which are defined similarly to Eq. (1) for the electronic cell. Given that strain relaxes more slowly than bound electronic wavefunctions, we only consider $V_s \ge V_e$. In this two-unit-cells approximation, the Fourier transform of the strain produced by the QD array is

$$\tilde{\boldsymbol{\epsilon}}_{ij}^{a}(\mathbf{q}) = \frac{(2\pi)^{3}}{V_{s}} \sum_{\mathbf{Q} \in \Omega_{s}^{-1}} \tilde{\boldsymbol{\epsilon}}_{ij}(\mathbf{Q}) \tilde{\boldsymbol{\chi}}_{e}(\mathbf{q} - \mathbf{Q})$$
$$= \frac{(2\pi)^{3}}{V_{s}} \left(\tilde{\boldsymbol{\epsilon}}_{ij} * \tilde{\boldsymbol{\chi}}_{e} \right)_{s}(\mathbf{q}), \tag{8}$$

where χ_{e} is the characteristic function of the electronic unit cell Ω_{e} in Ω_{s} , which is given for our case in Appendix C. Superscript "a" indicates array. We follow the notation that $\mathbf{q} \in \Omega_{e}^{-1}$ and $\mathbf{Q} \in \Omega_{s}^{-1}$. $(\tilde{\epsilon}_{ij} * \tilde{\chi}_{e})_{s}(\mathbf{q})$ denotes a convolution where the subscript "s" indicates that the convolution is over the wave vectors $\mathbf{Q} \in \Omega_{s}^{-1}$; see Appendix B for Fourier transform and convolution definitions. We show in Sec. III B that choosing the linear dimensions of Ω_{s} to be integer multiples of the linear dimensions of Ω_{e} ensures that all vectors $\mathbf{q} \in \Omega_{e}^{-1}$ are also in Ω_{s}^{-1} . This choice allows Eq. (8) to be evaluated efficiently.

C. Piezoelectric potential

III-nitride materials are strongly piezoelectric, having both spontaneous and strain-driven polarizations.^{31,32} The calculation of the polarization from an electric field requires the knowledge of the static dielectric constant ε of the material. In prior work, Fourier spacebased approaches have assumed a uniform dielectric constant. Given the large difference in ε between GaN and InN (9.8 and 13.8, respectively), spatial variation can be significant in InGaN QD systems. We present a method to obtain the Fourier transform of the scalar potential $\tilde{\varphi}(\mathbf{q})$ assuming $\varepsilon(\mathbf{r})$ changes with the local alloy fraction. We find that correcting for the spatial dependence of the dielectric function leads to important changes in the piezoelectric potential and show in Sec. V that this change significantly shifts the lowest QD energy levels. We do not consider metallic screening, which can be important in highly doped materials.^{33–35}

Generally, we can write the displacement field $D(\mathbf{r})$ as

$$\mathbf{D}(\mathbf{r}) = \boldsymbol{\varepsilon}_0 \mathbf{E}(\mathbf{r}) + \mathbf{P}_{\text{tot}}(\mathbf{r}),$$

where $\mathbf{E}(\mathbf{r})$ is the electric field, ε_0 is the vacuum permittivity, and \mathbf{P}_{tot} is the total polarization. In the strained material, there are three sources of polarization: bound charge, strain, and spontaneous polarization,

$$\mathbf{P}_{\text{tot}}(\mathbf{r}) = \mathbf{P}_{\text{bnd}}(\mathbf{r}) + \mathbf{P}_{\text{st}}(\mathbf{r}) + \mathbf{P}_{\text{sp}}(\mathbf{r}).$$

Here, we assume no free charge screening. Assuming P_{bnd} to be linear with the electric field and incorporated into $\varepsilon(\mathbf{r})$ as usual,

$$\mathbf{D}(\mathbf{r}) = \boldsymbol{\varepsilon}(\mathbf{r})\mathbf{E}(\mathbf{r}) + \mathbf{P}_{\mathsf{st}}(\mathbf{r}) + \mathbf{P}_{\mathsf{sp}}(\mathbf{r}), \tag{9}$$

where $P_{st}(\mathbf{r}) + P_{sp}(\mathbf{r}) = P(\mathbf{r})$ is the residual polarization after electric-field-induced bound charge has been included in $\varepsilon(\mathbf{r})$.

We take $\varepsilon(r)$ to be ε^h in the host material and ε^d in the dot material, so

$$\boldsymbol{\varepsilon}(\mathbf{r}) = \boldsymbol{\varepsilon}^{\mathsf{h}} + (\boldsymbol{\varepsilon}^{\mathsf{d}} - \boldsymbol{\varepsilon}^{\mathsf{h}})\boldsymbol{\chi}_{\mathsf{d}}(\mathbf{r}). \tag{10}$$

We obtain ε^{d} by a linear interpolation of the binary compounds' bulk dielectric constants. Taking the divergence of Eq. (9), using $\nabla \cdot \mathbf{D} = 0$, taking the Fourier transform and solving for the electric field gives

$$E_m(\mathbf{r}) = -\frac{1}{\varepsilon(\mathbf{r})} F^{-1} \bigg\{ \frac{q_n}{q_m} \tilde{P}_n(\mathbf{q}) \bigg\},\tag{11}$$

where F^{-1} represents the inverse Fourier transform. Using $E_m = -\partial_m \varphi$, where $\partial_m \equiv \frac{\partial}{\partial x_m}$ and $\varphi(\mathbf{r})$ is the scalar potential,

$$\tilde{\varphi}(\mathbf{q}) = -\frac{i}{q_m} F\left\{\frac{1}{\varepsilon(\mathbf{r})} F^{-1}\left\{\frac{q'_n}{q'_m} \tilde{P}_n(\mathbf{q}')\right\}(\mathbf{r})\right\}.$$
(12)

For the case of sharp alloy interfaces, $\chi_d(\mathbf{r})$ is either 1 or 0, and Eq. (10) gives

$$\frac{1}{\varepsilon(\mathbf{r})} = \frac{1}{\varepsilon^{\mathsf{h}}} + \left(\frac{1}{\varepsilon^{\mathsf{d}}} - \frac{1}{\varepsilon^{\mathsf{h}}}\right) \chi_{\mathsf{d}}(\mathbf{r}).$$
(13)

We treat the case of smoothly varying alloy fraction in Sec. IV. Putting this result in Eq. (12) gives

$$\tilde{\varphi}(\mathbf{q}) = \tilde{\varphi}^{\mathsf{h}}(\mathbf{q}) + \Delta \tilde{\varphi}(\mathbf{q})$$
 (14)

with

$$\tilde{\varphi}^{\mathsf{h}}(\mathbf{q}) = -\frac{i}{q_m} \frac{1}{\varepsilon^{\mathsf{h}}} \frac{q_n}{q_m} \tilde{P}_n(\mathbf{q}), \qquad (15)$$

$$\Delta \tilde{\varphi} = \frac{-i}{q_m} \left(\frac{1}{\varepsilon^{\mathsf{d}}} - \frac{1}{\varepsilon^{\mathsf{h}}} \right) F \left\{ \chi_{\mathsf{d}} F^{-1} \left\{ \frac{q'_n}{q'_m} \tilde{P}_n(\mathbf{q}') \right\} \right\}.$$
(16)

Here, $\tilde{\varphi}^{h}$ is the contribution to φ with $\varepsilon_{r}(\mathbf{r}) = \varepsilon^{h}$, and $\Delta \tilde{\varphi}$ is the change in $\tilde{\varphi}$ due to the dot material having a different dielectric constant. See Ref. 25 for both spontaneous and strain-induced polarization fields $\tilde{P}_{n}(\mathbf{q})$ for the wurtzite crystal structure.

We now show the piezoelectric potentials that result from this formulation for our model system in Sec. II A. Figure 4(a) shows $\varphi(z)$ along the central axis of the QD calculated with constant ε of the dot and host and with Eq. (14). As expected, the calculation with spatially varying $\varepsilon(\mathbf{r})$ agrees with φ_{uni}^{d} inside the dot and also agrees with φ_{uni}^{h} outside the dot, with a transition near the boundary that is captured by neither of the uniform cases.

Figure 3 showed how spatially varying elastic constants change strain profiles. Figure 4(b) shows how those strain corrections change φ . The peak correction of 8 mV is significant if looking to converge the energy levels within a few meV.

III. COMMENSURATE UNIT CELLS FOR EFFICIENT COUPLING OF STRAIN IN $k \cdot p$

Here, we briefly present the QD $\mathbf{k} \cdot \mathbf{p}$ model we use for electronic structure calculations. This QD Hamiltonian is written in a symmetry adapted basis, which reduces the computational cost for calculating and diagonalizing the Hamiltonian. In this symmetry adapted basis, we show how the strain produced by the QDs contributes to the Hamiltonian. We also introduce strain effects using a larger unit cell than the electronic cell defined in Fig. 1. In this section, our goal is to show our method of efficiently including strain in the QD $\mathbf{k} \cdot \mathbf{p}$ model, which we do by choosing the strain unit cell's dimensions to be integer multiples of the unit cell used for the electronic structure calculations. The $\mathbf{k} \cdot \mathbf{p}$ model and symmetry adapted basis are standard, and we briefly describe them and refer to the sources for full descriptions.



FIG. 4. (a) Piezoelectric potential $\varphi(z)$ along the central axis, beginning in the center of the dot. Blue and red dashed lines show φ calculated with uniform $\varepsilon(\mathbf{r}) = \varepsilon^{h}$ and $\varepsilon(\mathbf{r}) = \varepsilon^{d}$, respectively. Black line shows the case with spatially varying $\varepsilon(\mathbf{r})$. Note that φ is antisymmetric in z. These results show that the case of uniform ε cannot accurately describe φ throughout the system. Strain calculations include third-order corrections for the nonuniform elastic constants. (b) Potential difference for various elastic constant corrections. Difference is with respect to $\varphi_{h}(\mathbf{r})$, which is calculated with uniform $\lambda(\mathbf{r}) = \lambda^{GaN}$. The zeroth order case (blue line) shows $\lambda(\mathbf{r}) = \lambda^{d}$. Includes a spatially varying dielectric constant. System parameters are given in Table I. Both calculations implement an alloy smoothing of $\delta = [1.5, 1.5, 2.5]$ Å, which is described in Sec. IV. Vertical dashed lines indicate the nominal material interface without smoothing.

We use the 8-band $\mathbf{k} \cdot \mathbf{p}$ model for bulk wurtzite material presented in Ref. 10 in the basis of Γ -point Bloch functions, which includes spin-orbit coupling, crystal field splitting, and strain. Since choosing angular momentum \hat{J}_z eigenfunctions aids in the construction of a symmetry adapted basis, we rotate the 8-band Hamiltonian into the \hat{J}_z eigenbasis, similar to Ref. 36. See Appendix A for details. Recent work has alternatively obtained $\mathbf{k} \cdot \mathbf{p}$ parameters directly in the symmetry adapted basis using *ab initio* calculations.³⁷

A. Quantum dot k · p

We construct the QD Hamiltonian by giving the bulk Hamiltonian a spatial dependence. The solution is expanded using slowly varying envelope functions.^{8,18–20} The envelope functions are periodic with the superlattice and can be expanded in the Fourier domain using the superlattice reciprocal wave vectors **q** defined in Eq. (1). Writing the envelope functions in terms of plane waves leads to a non-sparse Hamiltonian.^{8,19} Due to the

broken translation symmetry, we apply the substitution $k_j \rightarrow -i \frac{\partial}{\partial x_j}$ and symmetrize the operations to preserve Hermiticity.^{18,19,38} For computational efficiency, we use a symmetry adapted basis, which takes advantage of the C_6 symmetry of the wurtzite crystal structure by block diagonalizing the Hamiltonian. Symmetry adapted bases have been fully described for both zincblende and wurtzite systems.^{18,25} The symmetry-adapted basis reduces the Fourier space sampling to a single sextant $0 \le q_y \le \tan(\frac{2\pi}{6})q_x$ of the full space and block diagonalizes the Hamiltonian into six blocks, which are labeled by $m_f = \{-5/2, -3/2, -1/2, 1/2, 3/2, 5/2\}$. The label m_f can be interpreted as a total quasi angular momentum.^{20,25} The basis elements are

$$|m_f, \alpha, \mathbf{q}\rangle = \Lambda(m_f, \alpha, \mathbf{q}, \mathbf{r})|u_{\alpha}\rangle,$$
 (17)

where

$$\Lambda(m_f, \alpha, \mathbf{q}, \mathbf{r}) = \frac{1}{\sqrt{6}} \sum_{l=0}^{5} e^{i \left(\overrightarrow{\mathbf{R}}_l \mathbf{q}\right) \cdot \mathbf{r}} e^{i l \phi \left[m_f - J_z(\alpha)\right]}$$
(18)

when either q_x or q_y is nonzero and $\Lambda(m_f, \alpha, \mathbf{q}, \mathbf{r}) = e^{i\mathbf{q}\cdot\mathbf{r}}$ when $q_x = q_y = 0$ and $J_z(\alpha) = m_f$. \mathbf{R}_l is the $l\phi$ rotation around the z-axis with $\phi = 2\pi/6$. This basis consists of the basis functions of the irreducible representations of the double group \overline{C}_6 . This block diagonalization greatly reduces the computational cost to diagonalize the Hamiltonian. Equation (18) distinguishes wave vectors that are purely along the z-axis from those that have an xy-component, which we denote by \mathbf{q}_z and \mathbf{q} , respectively. These two cases differ because a z-axis rotation leaves \mathbf{q}_z invariant while sending \mathbf{q} to a new wave vector $\mathbf{R}_l \mathbf{q}$. The case of $|m_f, \alpha, \mathbf{q}_z\rangle$ with $J_z(\alpha) \neq m_f$ does

new wave vector $\mathbf{R}_I \mathbf{q}$. The case of $|m_f, \alpha, \mathbf{q}_z\rangle$ with $J_z(\alpha) \neq m_f$ does not exist in the basis set. In this basis, the eigenvalue problem is

$$\sum_{\alpha=1}^{8} \sum_{\mathbf{q}} \mathcal{H}_{m_{f}\alpha'\alpha}(\mathbf{q}',\mathbf{q}) A^{\alpha}_{im_{f}}(\mathbf{q}) = E_{i} A^{\alpha'}_{im_{f}}(\mathbf{q}'), \qquad (19)$$

where

$$\mathcal{H}_{m_{f}\alpha'\alpha}(\mathbf{q}',\mathbf{q}) \equiv \langle m_{f},\alpha',\mathbf{q}'|\mathcal{H}|m_{f},\alpha,\mathbf{q}\rangle$$
$$= \frac{1}{V_{e}} \int_{V_{e}} \mathbf{d}^{3}\mathbf{r} \Lambda^{*}(\mathbf{r}) H_{\alpha'\alpha}\Lambda(\mathbf{r}), \qquad (20)$$

where we have dropped some arguments in Λ for brevity. $H_{\alpha'\alpha}$ are the bulk Hamiltonian matrix elements presented in Appendix A. Expressions for $\mathcal{H}_{m_{f}\alpha'\alpha}(\mathbf{q}', \mathbf{q})$ are fully written out in Appendix D in terms of the bulk Hamiltonian matrix elements and QD characteristic function.

B. Strain and piezoelectric coupling

Deformation potentials and piezoelectric effects, which are both strain-driven, are important for accurate calculations of the electronic structure in III-N materials. However, including deformation potentials can be computationally costly for the case of isolated dots. The two-unit cell approach presented in Sec. II B allows for the study of isolated dots but at the cost of computationally expensive convolutions. Additionally, another layer of convolutions appears in the Hamiltonian matrix elements, leading to composed convolutions. Here, we present the matrix elements due to strain and show our computationally efficient approach of dealing with these composed convolutions by choosing the linear dimensions of the real-space strain cell Ω_s to be integer multiples of those of the electronic cell Ω_e .

The bulk strain Hamiltonian matrix elements in Eqs. (A2) and (A4) can be written as

$$H_{lpha'lpha} = \sum_{ij} f^{ij}_{lpha'lpha} \epsilon_{ij}(\mathbf{r}),$$

where $f_{\alpha\alpha}^{y}$ consist of $\mathbf{k} \cdot \mathbf{p}$ parameters $(a_i, l_i, m_i, \text{ and } n_i)$. Using the prescription of Sec. III A and defining

$$\begin{split} S^{ll'm_f}_{\alpha',\alpha} &= e^{i\phi\left\{l\left[m_f - J_z(\alpha)\right] - l'\left[m_f - J_z(\alpha')\right]\right\}},\\ S^{l'm_f}_{\alpha'} &= e^{-il'\phi\left[m_f - J_z(\alpha')\right]}, \end{split}$$

the strain contributions to the QD Hamiltonian are

$$\begin{split} \mathcal{H}_{m_{f}\alpha'\alpha}^{ij,\text{st}}(\mathbf{q}',\mathbf{q}) &= \frac{1}{6} \sum_{l,l'=0}^{5} S_{\alpha',\alpha}^{ll'm_{f}} h_{\alpha'\alpha}^{ij,\text{st}} \left(\stackrel{\leftrightarrow}{\mathbf{R}}_{l'}\mathbf{q}', \stackrel{\leftrightarrow}{\mathbf{R}}_{l}\mathbf{q} \right), \\ \mathcal{H}_{m_{f}\alpha'\alpha}^{ij,\text{st}}(\mathbf{q}',\mathbf{q}_{z}) &= \frac{1}{\sqrt{6}} \sum_{l'=0}^{5} S_{\alpha'}^{l'm_{f}} h_{\alpha'\alpha}^{ij,\text{st}} \left(\stackrel{\leftrightarrow}{\mathbf{R}}_{l'}\mathbf{q}',\mathbf{q}_{z} \right), \\ \mathcal{H}_{m_{f}\alpha'\alpha}^{ij,\text{st}}(\mathbf{q}_{z}',\mathbf{q}_{z}) &= h_{\alpha'\alpha}^{ij,\text{st}}(\mathbf{q}_{z}',\mathbf{q}_{z}), \end{split}$$

where

$$\begin{aligned} h_{\alpha'\alpha}^{ij,\text{st}}(\mathbf{q}',\mathbf{q}) &= \frac{(2\pi)^3 f_{\alpha'\alpha}^{ij,\text{h}}}{V_{\text{e}}} \tilde{\boldsymbol{\epsilon}}_{ij}^{\text{a}}(\mathbf{q}'-\mathbf{q}) \\ &+ \frac{(2\pi)^6 \left(f_{\alpha'\alpha}^{ij,\text{d}} - f_{\alpha'\alpha}^{ij,\text{h}}\right)}{V_{\text{e}}^2} \left(\tilde{\boldsymbol{\chi}}_{\text{d}} * \tilde{\boldsymbol{\epsilon}}_{ij}^{\text{a}}\right)_{\text{e}} (\mathbf{q}'-\mathbf{q}). \end{aligned}$$

$$(21)$$

Here, $f_{\alpha'\alpha}^{ij,h}$ and $f_{\alpha'\alpha}^{ij,d}$ are the $\mathbf{k} \cdot \mathbf{p}$ parameters for bulk host and dot materials, respectively. $\tilde{\boldsymbol{\epsilon}}_{ij}^{a}$ is the strain produced by the QD array calculated in Sec. II B. The subscript "e" in $\left(\tilde{\chi}_{d} * \tilde{\boldsymbol{\epsilon}}_{ij}^{a}\right)_{e}$ indicates that the convolution is over the wave vectors $\mathbf{q} \in \Omega_{e}^{-1}$. Inserting the superlattice strain from Eq. (8) into the $\mathbf{k} \cdot \mathbf{p}$ strain matrix elements from Eq. (21) leads to composed convolutions,

$$\begin{aligned} \left(\tilde{\chi}_{\mathsf{d}} \star \tilde{\epsilon}_{ij}^{\mathsf{a}} \right)_{\mathsf{e}}(\mathbf{q}) &= \sum_{\mathbf{q}' \in \Omega_{\mathsf{e}}^{-1}} \tilde{\chi}_{\mathsf{d}}(\mathbf{q}') \tilde{\epsilon}_{ij}^{\mathsf{a}}(\mathbf{q} - \mathbf{q}') \\ &= \frac{(2\pi)^3}{V_{\mathsf{s}}} \sum_{\mathbf{q}' \in \Omega_{\mathsf{e}}^{-1}} \tilde{\chi}_{\mathsf{d}}(\mathbf{q}') \sum_{\mathbf{Q} \in \Omega_{\mathsf{s}}^{-1}} \tilde{\epsilon}_{ij}(\mathbf{Q}) \tilde{\chi}_{\mathsf{e}}(\mathbf{q} - \mathbf{q}' - \mathbf{Q}), \end{aligned}$$

$$(22)$$

which can be computationally demanding depending on the number of wave vectors used. The original proposal of using a

large strain cell with a smaller electronic cell imposed no relationship between their sizes.^{20,27} Equation (22) then requires evaluating $\tilde{\chi}_{e}$ at points $\mathbf{q} - \mathbf{q}' - \mathbf{Q}$ that are contained on neither the electronic nor strain meshes, requiring a unique convolution be calculated for every $\mathbf{q}' \in \Omega_{\mathbf{e}}^{-1}$. It is well known that using the convolution theorem to compute a convolution between two vectors of length N has a computational cost that scales as $N \log(N)$. Similarly, the computational cost for a convolution on a 3D $N \times N \times N$ mesh scales as $N^3 \log(N)$. Computing the composed convolutions in Eq. (22) would then scale as $N_e^3 \log(N_e) N_s^3 \log(N_s)$ since a convolution in \mathbf{Q} has to be calculated for each \mathbf{q}' . Note that the convolutions in Eq. (22) are linear convolutions, which implies that the arrays of function values must be padded with zeros before using the convolution theorem, as detailed in Appendix B. This zero padding increases both N_e and N_s . We show that choosing a strain unit cell to be a supercell of the electronic unit cell reduces the required number of convolutions, leading to an improved scaling of $N_{\mathsf{e}}^3 \log(N_{\mathsf{e}}) + N_{\mathsf{s}}^3 \log(N_{\mathsf{s}}).$

Choosing the strain unit cell linear dimensions to be multiples of the electronic cell, we have

$$L_i^{\rm s} = n_i L_i^{\rm e}, \quad i = 12 \text{ and } 3,$$
 (23)

where n_i take positive integer values. This choice of real-space unit cells leads to the electronic Fourier space mesh being contained in the strain mesh $\Omega_e^{-1} \subset \Omega_s^{-1}$. The wave vectors **Q** then have a spacing that is a fraction of the spacing of the electronic wave vectors **q**,

$$\Delta Q_i = \frac{\Delta q_i}{n_i}.$$
 (24)

See Fig. 5 for an example of commensurate meshes and their overlapping Fourier-space meshes. Note that from Eq. (22), $\tilde{\epsilon}_{ij}^a$ is then only sampled at points $\Delta \mathbf{q} = \mathbf{q} - \mathbf{q}'$, which belong to the electronic mesh. Our procedure starts with using the convolution theorem (see Appendix B) to efficiently calculate the inner convolution $(\tilde{\epsilon}_{ij} * \tilde{\chi}_{e})_{s}(\mathbf{Q})$ on the strain mesh to obtain $\tilde{\epsilon}_{ij}^{a}(\mathbf{Q})$. Since the wave vectors \mathbf{Q} also contain the wave vectors \mathbf{q} , we can then extract the points that lie on the electronic mesh to obtain $\tilde{\epsilon}_{ij}^{a}(\mathbf{q})$. Lastly, we perform the second convolution $(\tilde{\chi}_{d} * \tilde{\epsilon}_{ij}^{a})_{e}(\mathbf{q})$, again using the convolution theorem. This workflow is shown in Fig. 6. This method computes only two 3D convolutions and so has a complexity scaling of $N_{e}^{3} \log(N_{e}) + N_{s}^{3} \log(N_{s})$, which is a considerable improvement compared to the non-commensurate case. Note that N_{s} is generally much larger than N_{e} to obtain appropriate convergence, so the computational cost is dominated by the convolutions on Ω_{s}^{-1} .

The piezoelectric potential brings no additional complexity, and the workflow for calculating the piezoelectric potential is shown in Fig. 6. The potential is initially calculated on the strain mesh, and the electronic mesh portion is extracted to calculate the piezoelectric potential contributions to the Hamiltonian, which are written out in Appendix D.

IV. SMOOTH ALLOY PROFILE

When InGaN devices are grown by MBE, indium diffuses between layers.¹ While most studies of MBE-grown materials simulate





abrupt junctions, this diffusion leads to smoothing of the material interfaces, producing a continuously varying alloy fraction, which changes the local band properties and lattice constant, which in turn change strain and polarization fields. This smooth alloy profile must be included for accurate modeling. The smooth indium profiles produced by diffusion differ from the random alloy fluctuations studied in Ref. 7 using atomistic modeling. Reference 12 has used random fluctuations at material interfaces to study quantum well width fluctuations, which is similar to the profiles produced by indium diffusion. In this section, we present a method to include alloy diffusion effects in Fourier-space calculations by effectively smoothing the characteristic function of the dot. Smooth indium profiles also provide a computational benefit, since sharp features of the confining potentials are removed, so fewer wave vectors are required to attain convergence. We use this method to produce an alloy profile that models indium diffusion and to address how spatially varying material parameters can be included within the piezoelectric potential and electronic structure calculations. Our examples focus on indium alloying, but the methods are general for all $\mathbf{k} \cdot \mathbf{p}$ calculations of alloy structures. Reference 39 presents a similar procedure for strain calculations only, with a linearly graded alloy profile.



FIG. 6. Workflows for calculating strain $\tilde{\epsilon}^a$ and piezoelectric potential $\tilde{\phi}^a$ on the electronic mesh of an array of dots, beginning from the strain of a single dot on the strain mesh. Upper path shows the composed convolutions of the form in Eq. (22), which are necessary to compute the matrix elements in Eq. (21). Lower branch shows steps for calculating $\tilde{\phi}^a$, which is used in Eqs. (D1), (D2), and (D3). e-mesh and s-mesh signify the Fourier space electronic and strain meshes, respectively.

In the case of a sharp material interface, the local alloy fraction $X(\mathbf{r})$ can be defined by the characteristic function of the dot

$$X(\mathbf{r}) = X_0 \chi_{\mathsf{d}}(\mathbf{r}),$$

where the characteristic function $\chi_{d}(\mathbf{r})$ defines the geometry of the dot with indium fraction X_0 . By convolving with a Gaussian

 $G(\mathbf{r},\boldsymbol{\delta}) = \frac{1}{(2\pi)^{\frac{1}{2}} \delta_x \delta_y \delta_z} \mathbf{e}^{-\frac{1}{2} \left(\frac{x^2}{\delta_x^2} + \frac{y^2}{\delta_y^2} + \frac{x^2}{\delta_z^2}\right)} \text{ or other kernel, we can obtain a}$ smooth version of the characteristic function

$$\begin{split} X_{\mathsf{sm}}(\mathbf{r}) &= (X_0 \chi_{\mathsf{d}} \star G)(\mathbf{r}) \\ &= X_0 \chi_{\mathsf{sm}}(\mathbf{r}), \end{split}$$

where $\boldsymbol{\delta} = [\delta_x, \delta_y, \delta_z]$ controls the radius of smoothing and needs to be chosen to model the desired alloy diffusion. $G(\mathbf{r}, \boldsymbol{\delta})$ is normalized to preserve the total amount of alloying element, and $\chi_{sm}(\mathbf{r})$ is a smoothed characteristic function. Using the convolution theorem, the smoothed characteristic function satisfies

$$\tilde{\chi}_{\rm sm}(\mathbf{q}) = \tilde{\chi}_{\rm d}(\mathbf{q}) \mathrm{e}^{-\frac{\left(\delta_x^2 q_x^2 + \delta_y^2 q_y^2 + \delta_z^2 q_z^2\right)}{2}}$$

Note that $\chi_{sm}(\mathbf{r})$ is no longer strictly a characteristic function, as it takes values continuously between 0 and 1. We now show that it can be inserted in the place of the characteristic function in the previous sections to give $\mathbf{k} \cdot \mathbf{p}$ parameters, strain, and piezoelectric fields accurately with a smooth alloy profile.

B. Material parameters

We focus on the case of InGaN to illustrate the interpolation of material parameters. In the case of sharp material interfaces, the host and dot regions each consist of a uniform material. The host material is a binary material and has well-defined parameters. The dot region consists of alloyed InGaN, and its parameters are obtained by either linear or bowed interpolation of bulk GaN and InN parameters, which are listed in Appendix A.

In the case of a smooth alloy profile, the dot and host regions are no longer uniform, giving the material parameters a smooth spatial dependence. Parameters that were linearly interpolated in the sharp interface case can still be obtained from a simple linear interpolation based on the local alloy fraction $X(\mathbf{r})$. The bandgap E_q is nonlinear in the alloy fraction due to a bowing factor. This nonlinearity prevents us from using the convolution theorem in calculating the Hamiltonian matrix elements. However, we show that neglecting the bowing parameters in the alloy-smoothing region can still give computationally efficient and accurate smoothed profiles when the alloy fraction is not too large.

The local value for any of the linearly interpolated material parameters depends on the local alloy fraction

$$f(\mathbf{r}) = f^{\mathsf{B}}X(\mathbf{r}) + [1 - X(\mathbf{r})]f^{\mathsf{A}},$$
(25)

scitation.org/journal/jap

where f can be a parameter such as lattice constant, and subscripts A and B stand for the two binary materials, GaN and InN, for example. For this case of linearly interpolated quantities, smoothed parameters can be written as follows:

ARTICLE

$$f(\mathbf{r}) = f^{\mathsf{d}} \chi_{\mathsf{sm}}(\mathbf{r}) + [1 - \chi_{\mathsf{sm}}(\mathbf{r})] f^{\mathsf{A}}, \qquad (26)$$

where f^{d} is the linearly interpolated material parameter at the nominal alloy fraction X_0 of the QD.

Bandgaps do not vary linearly with alloy fraction and are generally well described with a bowing term, as

$$E_g(\mathbf{r}) = E_g^{\mathsf{B}} X(\mathbf{r}) + [1 - X(\mathbf{r})] E_g^{\mathsf{A}} - X(\mathbf{r}) [1 - X(\mathbf{r})] C,$$

where C is a bowing constant. More complicated forms allow the bowing factor to depend on alloy fractions,⁴⁰ but we do not consider such effects. Following the same procedure as in Eq. (26), a smoothed version can be written as follows:

$$E_{g}(\mathbf{r}) = E_{g}^{A} + \left[E_{g}^{B} - E_{g}^{A} \right] E_{g}^{A} X_{0} \chi_{sm}(\mathbf{r}) - C X_{0} \chi_{sm}(\mathbf{r}) [1 - X_{0} \chi_{sm}(\mathbf{r})], \qquad (27)$$

where the first two terms are the linear interpolation and the last term is the bowing. This bowing term brings additional complexity when performing $\mathbf{k} \cdot \mathbf{p}$ calculations due to the nonlinearity in $\chi_{\rm sm}({\bf r})$. Dealing with this nonlinearity is trivial for the case of sharp material interfaces since $\chi^2 = \chi$, which is not true for χ_{sm} . The bandgap appears in the Hamiltonian through the diagonal element E'_{c} in Eq. (A3). In the plane wave basis, the contributions of the nonlinear term $CX_0^2\chi^2_{sm}$ are

$$\begin{split} h_{E_{\mathbf{g}}}^{\mathrm{nonlinear}}(\mathbf{q}',\mathbf{q}) &= C X_0^2 \frac{1}{V} \int_V \mathbf{d}^3 \mathbf{r} \, \mathbf{e}^{-i\mathbf{q}'\cdot\mathbf{r}} \chi_{\mathrm{sm}}^2(\mathbf{r}) \mathbf{e}^{i\mathbf{q}\cdot\mathbf{r}} \\ &= C X_0^2 \frac{(2\pi)^6}{V^2} (\chi_{\mathrm{sm}} \star \chi_{\mathrm{sm}}) (\mathbf{q}'-\mathbf{q}). \end{split}$$

Additional convolutions must then be calculated to include the bowing of the bandgap, increasing the computational cost. However, we show that we can, to good approximation, use a bandgap that is linearly interpolated between the host and dot bandgaps,

$$E_g(\mathbf{r}) \approx E_g^{\mathsf{d}} \chi_{\mathsf{sm}}(\mathbf{r}) + [1 - \chi_{\mathsf{sm}}(\mathbf{r})] E_g^{\mathsf{h}}.$$
 (28)

Here, E_{α}^{d} is the bulk bandgap at an alloy fraction of X_{0} , and E_{α}^{h} is the bulk bandgap of the host material. This linear interpolation gives a good approximation for the bandgap for most regions, as shown in Fig. 7, and overestimates E_g near the interfaces. Disagreements increase with dot indium fraction. The regions with largest deviation are in the same locations where E_{g} changes over 1.5 eV; so we expect the slight shift of position where each bandgap occurs to have a minimal effect. We can estimate from Fig. 7 that the shift in the effective height of the dot in the z-direction is smaller than $\delta_z/2$, so a particle-in-a-box model allows the fractional change in the most confined energy level to

be estimated to be approximately $\delta_z/h \approx 6\%$. The neglection of the χ^2_{sm} term allows the theory to stay linear and therefore efficiently calculated with the convolution theorem. If the smoothing radius becomes comparable to the dot dimensions, this approximation may need to be reconsidered.

C. Strain and the piezoelectric potential

Here, we show how smoothing is included in the strain and piezoelectric potential calculations. Once calculated, those strains and piezoelectric potentials can be included in the $\mathbf{k} \cdot \mathbf{p}$ model exactly as shown in Sec. III B.

Following the derivations from Refs. 8 and 26, it is not obvious how smoothing is to be implemented in strain calculations since they begin from the stress of the sharp interface dot/barrier interface. However, Ref. 30 presents an alternative derivation for the same strain calculation indicating that $\chi(\mathbf{r})$ in the strain expressions can be exchanged for the smoothed version $\chi_{sm}(\mathbf{r})$ without any further changes, which is the approach taken by Ref. 39.

For the piezoelectric potential, Eq. (13) for the spatially varying inverse dielectric constant assumed sharp interfaces. In the case of a smooth indium profile, we use Eq. (25) to write

$$\boldsymbol{\varepsilon}(\mathbf{r}) = \boldsymbol{\varepsilon}^{\mathsf{B}} X(\mathbf{r}) + [1 - X(\mathbf{r})] \boldsymbol{\varepsilon}^{\mathsf{A}}.$$
(29)



FIG. 7. (a) Bandgap in a QD system obtained by bowed interpolation using Eq. (27) along the z and x axes through the center of the dot. Bandgap obtained from linear interpolation using Eq. (28) is visually indistinguishable. (b) Difference in the bowed and linearly interpolated bandgaps along the z and x axes. QD parameters are listed in Table I. For computational simplicity and smooth curves, these results were obtained using a rectangular real-space unit cell with dimensions $L_x = L_y = 500$ Å, $L_z = 70$ Å and a smoothing of $\delta = [3, 3, 5]$ Å.

In the scenario where $X(\mathbf{r})$ is spatially varying, Eq. (13) can no longer be applied, because the inverse of the dielectric constant is not a linear function of indium. However, similar to the bandgap, we find that

$$\frac{1}{\varepsilon(\mathbf{r})} \approx \frac{1}{\varepsilon^{\mathsf{d}}} \chi_{\mathsf{sm}}(\mathbf{r}) + [1 - \chi_{\mathsf{sm}}(\mathbf{r})] \frac{1}{\varepsilon^{\mathsf{A}}}$$
(30)

still gives an accurate representation of $\varepsilon^{-1}(\mathbf{r})$. Figure 8 shows in our test case a disagreement of less than 1 percent between the inverse dielectric from Eq. (29) and the linear interpolation in Eq. (30). The form of Eq. (30) allows us to use Eqs. (14)–(16) for the piezoelectric potential with a simple substitution of $\chi(\mathbf{r})$ by $\chi_{sm}(\mathbf{r})$.

V. IMPACTS OF CORRECTIONS

In this section, we apply our methodology to study the case of a 1D array of QDs, inspired by those in Ref. 1. Unlike those experimental dot arrays embedded in nanowires, ours is an infinite 1D chain embedded in bulk GaN. We achieve this 1D array by taking $n_3 = 1$ to fully couple the dots in the z-direction and choose L_{12} and n_{12} sufficiently large to avoid both electronic and strain effects from neighboring dots in the xy-plane. In the experimental dot arrays, strain can be significantly modified by the nanowire walls, but the strength of the piezoelectric field along the wire axis, which



FIG. 8. (a) Inverse dielectric constant from Eq. (29) along the z and x axes through the center of the dot. Linearly interpolated inverse from Eq. (30) is visually indistinguishable. (b) Relative difference of the linear and nonlinear interpolations. QD parameters are listed in Table I. Results used the same rectangular mesh and smoothing as in Fig. 7.

plays the most important role in modifying the band structure from a strain-free calculation, is significant in both cases.⁵

We investigate the convergence of the lowest electron and hole state energies E_c and E_v , which define the fundamental gap of the dot $E_0 \equiv E_c - E_v$. More specifically, we show that the largest wave vector sampled plays a dominant role in convergence and that the choice of hexagonal or circular dot cross sections is not significant for dots that maintain the same volume. We also show the energy shifts experienced by these two states when using uniform or spatially varying material parameters and when including alloy smoothing.

We model an infinite 1D QD array with parameters listed in Table I. The 1D dot array has an experimentally well-characterized dot-dot spacing in z, which fixes $L_3^{e} = L_3^{s} = L_3$, leaving L_{12} and n_{12} to be fixed. These QDs have a rather large radius, so the smallest spatial feature that we need to resolve is the decay of the bound wavefunctions into the classically forbidden region. Given that bound wavefunctions decay faster than strain, we need wave vectors that are relatively large to be able to resolve the wavefunctions.



FIG. 9. Convergence of the fundamental gap E_0 for a 1D array of QDs vs the maximum magnitude of q_{12} included in the $\mathbf{k} \cdot \mathbf{p}$ calculations. L_{12}^{e} and n_{12} vary while (a) has R = 200 Å with strain box size held constant at $L_{12}^{e} = 10R$ and (b) has R = 40 Å with $L_{12}^{e} = 16R$. Plane wave sampling m_{12} from 3 to 11 are shown for each choice of L_{12}^{e} ; the number next to each point indicates m_{12} . Different values of m_{12} , L_{12}^{e} that produce the same q_{12}^{max} can be seen to produce approximately the same E_0 , showing that q_{12}^{max} is a useful metric for convergence of these states. Since $q_{12}^{max} = m_{12}\pi/L_{12}^{e}$ and computational cost scales with m_{12} , smaller $L_{12}^{e} = 2R$, so the dots touch each other at the six edges of the hexagonal unit cell. In the larger dot, there is no signature of dot-dot tunneling, while in the smaller dot, tunneling of the wavefunctions into neighboring dots causes E_0 to have a significant change, labeled Δ . For R = 40 Å case, E_c and E_v were found to be converged within 5 meV with $m_{12} = 7$, $m_3 = 4$, and $n_{12} = 4$, smaller than those required for the larger dot, as reported in Table I.

Increasing m_{12} increases the maximum wave vector contained in the mesh, but we can also sample at larger wave vectors by using a smaller L_{12}^{e} . However, if the electronic cell is chosen too small, then there can be electronic wavefunction overlap between states of neighboring dots. We must then choose L_{12}^{e} as small as possible while also avoiding dot-dot interactions. As for strain, in order to study a 1D array, we must choose n_{12} sufficiently large to have $L_{12}^{s} = n_{12}L_{12}^{e}$ large enough that the strain of the QD superlattice does not extend across neighboring strain unit cells in the xy-plane.

With this intuition, we turn to the convergence of E_0 in terms of m_{12} , L_{12}^e , and n_{12} . Figure 9(a) shows the importance of the largest **q** in the electronic mesh, q_{12}^{\max} , for the convergence of E_0 . In this study, L_{12}^e and n_{12} are chosen to keep a constant $L_{12}^s = L_{12}^e n_{12} = 2000$ Å. We observe that E_0 is to good approximation a function of only q_{12}^{\max} and not of m_{12} and L_{12}^e , converging toward the same value for all choices of L_{12}^e . We also observe that the smallest L_{12}^e with highest m_{12} gives the most converged E_0 , since $q_{12}^{\max} = m_{12}\pi/L_{12}^e$. The black line in Fig. 9(a) represents the case of dots touching in the xy plane and, interestingly, does not break the convergence trend. However, we do find a break in the convergence trend for smaller dots in Fig. 9(b). This difference occurs because smaller dots have wavefunctions that extend further outside the dot region, which makes them more able to tunnel to a neighboring dot. Consequently, care has to be taken in choosing the unit cell dimensions for small QDs.

The lowest QD confined electron and hole energies, E_c and E_v , have each been converged within 5 meV by choosing m_{12} , m_3 , and n_{12} sufficiently large, see Table I. Material parameters are listed in Appendix A. Band edges and the lowest energy states are shown in Fig. 10. The thick lines labeled CB and VB represent the bulk



FIG. 10. Electronic structure of a cylindrical (solid, black) and hexagonal prism (dashed, red) QD superlattice system along the central axis of the dot for $\delta = [1.5, 1.5, 2.5]$ Å, corresponding to s = 1 in Fig. 11(a). Other system parameters are in Table I. The circular and hexagonal dots are not significantly different for these band-edge states, with eigenenergies differing by less than 12 meV. Thick lines labeled CB and VB are the bulk band edges under the influence of the piezoelectric field and the diagonal component of the deformation potential. Lowest bound electron and hole state energies, E_c and E_v , are shown by horizontal lines. Thin lines labeled ψ_c and ψ_v are z-axis projections of the probability distributions obtained from the envelope functions for these states. Dashed vertical lines are the nominal material interfaces before smoothing. These calculations include spatially varying elastic and dielectric constants.

TABLE II. Energy shifts due to spatially varying $\lambda(\mathbf{r})$ and $\varepsilon(\mathbf{r})$ relative to the case with uniform constants of the host material λ^{GaN} and ε^{GaN} . Cylindrical dot with parameters in Table I.

Energy shifts (meV)	$\lambda(\mathbf{r})$ and $arepsilon^{GaN}$	λ^{GaN} and $\epsilon(\mathbf{r})$	$\lambda(\mathbf{r})$ and $\varepsilon(\mathbf{r})$
ΔE_{c}	16.7	46.7	64.7
ΔE_{V}	-5.1	-30.6	-37.4
ΔE_0	21.7	77.4	102.1

band edges modified by the piezoelectric potential and the diagonal portion of the deformation potential; the valence band edge includes a third of the trace of the 3×3 valence band block from Eq. (A4). Figure 10 shows the electronic structure of both circular and hexagonal QDs. Convergence parameters were kept the same for both sets of calculations, and the radii were chosen to maintain the same volume. We conclude that the energies of the most confined electron and hole states are insensitive to the dot geometry.

The modifications in both the strain and the piezoelectric potential due to spatially varying elastic and dielectric constants also have effects on the electronic structure. Table II shows how much E_v and E_c shift due to corrections. We find that both corrections push the states apart, leading to an energy gap 100 meV larger than from simpler calculations with uniform ε and λ , a



FIG. 11. Effects of indium diffusion, given by $\delta = s[1.5, 1.5, 2.5]$ Å, for (a) E_c and E_v and (b) bulk conduction band edge. Remaining parameters are as listed in Table I. Increase in indium diffusion leads to less confinement, which pushes the two states apart in energy, widening the electronic gap E_0 . Electronic structure for s = 1 is shown in Fig. 10.

significant change that shows the importance of accurate modeling of dielectric and elastic parameters.

Figure 11(a) shows that indium diffusion pushes the lowest electron and hole states apart, which is due to changes in the confining potentials. From Fig. 11(b), we see that indium diffusion reduces the depth of the confining potential. The valence band shows similar behavior, leading to E_V being pushed down in energy. Consequently, the gap E_0 increases with indium diffusion.

In the case of sharp material interfaces, large wave vectors are needed to resolve the discontinuous parameter profiles. Smoothing removes the sharp interfaces, reducing the required q_{12}^{max} for the same degree of convergence. This reduction gives convergence with smaller m_{12} and m_3 , and therefore, the reduced computational cost is needed with increasing δ . In Figs. 11(a) and 11(b), we converged for the case of s = 1, guaranteeing convergence for the rest of the sweep.

VI. CONCLUSIONS

We have demonstrated techniques for and results of four modifications of standard Fourier space QD $\mathbf{k} \cdot \mathbf{p}$ theory. We have presented a method to include smoothly varying alloy profiles in Fourier-based strain, piezoelectric potential, and $\mathbf{k} \cdot \mathbf{p}$ calculations. We have shown that the effects of spatially varying elastic and dielectric parameters are non-negligible on the strain and piezoelectric potential and also produce important shifts of the lowest electron and hole states, significantly changing the calculated gap of the QD. For the case of $\mathbf{k} \cdot \mathbf{p}$ theory for isolated dots, we have presented a new method of overlapping electronic and strain meshes to facilitate the coupling of strain into the $\mathbf{k} \cdot \mathbf{p}$ Hamiltonian, greatly reducing the computational cost of calculating the Hamiltonian matrix elements. Lastly, we have shown that the maximum wave vector contained in the electronic sampling mesh is the most important criterion for determining the convergence of QD levels.

ACKNOWLEDGMENTS

We acknowledge useful conversation with Stanko Tomić about Fourier space $\mathbf{k} \cdot \mathbf{p}$ methods. We acknowledge funding from the Ontario Early Researcher Award and NSERC CREATE TOP-SET Program (Award No. 497981).

APPENDIX A: BULK k · p PARAMETERS

We use the following basis of \hat{J}_z eigenfunctions:

$$|u_{1}\rangle = |iS, \uparrow\rangle \quad |u_{5}\rangle = |-iS, \downarrow\rangle, |u_{2}\rangle = |-\frac{X+iY}{\sqrt{2}}, \uparrow\rangle \quad |u_{6}\rangle = |\frac{X-iY}{\sqrt{2}}, \downarrow\rangle, |u_{3}\rangle = |\frac{X-iY}{\sqrt{2}}, \uparrow\rangle \quad |u_{7}\rangle = |-\frac{X+iY}{\sqrt{2}}, \downarrow\rangle, |u_{4}\rangle = |Z, \uparrow\rangle \quad |u_{8}\rangle = |Z, \downarrow\rangle,$$
(A1)

where *S*, *X*, *Y*, and *Z* are Γ -point Bloch functions with arrows indicating spin. The eigenvalues of the \hat{f}_z eigenfunctions are as follows:

$$J_z = \left\{ \frac{1}{2}, \ \frac{3}{2}, \ -\frac{1}{2}, \ \frac{1}{2}, \ -\frac{1}{2}, \ -\frac{1}{2}, \ -\frac{3}{2}, \ \frac{1}{2}, \ -\frac{1}{2} \right\}$$

In the basis (A1), the bulk $\mathbf{k} \cdot \mathbf{p}$ Hamiltonian is

where

$$g(\mathbf{k}) = g_1(\mathbf{k}) + g_2(\mathbf{k}) + g_{\mathsf{cr}} + g_{\mathsf{so}} + g_{\mathsf{st}}$$

 $H = \begin{bmatrix} g(\mathbf{k}) & \gamma \\ -\gamma^* & g^*(\mathbf{k}) \end{bmatrix},$

$$g_{2}(\mathbf{k}) = \begin{bmatrix} A_{2}'\left(k_{x}^{2}+k_{y}^{2}\right)+A_{1}'k_{z}^{2} & 0\\ 0 & \left(\frac{L_{1}'+M_{1}}{2}\right)\left(k_{x}^{2}+k_{y}^{2}\right)+M_{2}k_{z}^{2}\\ 0 & -\frac{1}{2}N_{1}'k_{+}^{2}\\ 0 & -\frac{1}{\sqrt{2}}N_{2}'k_{+}k_{z} \end{bmatrix}$$

(A2)

$$g_{st} = \begin{bmatrix} a_2(\boldsymbol{\epsilon}_{xx} + \boldsymbol{\epsilon}_{yy}) + a_1\boldsymbol{\epsilon}_{zz} & 0\\ 0 & \frac{1}{2}(l_1 + m_1)(\boldsymbol{\epsilon}_{xx} + \boldsymbol{\epsilon}_{yy}) + m_2\boldsymbol{\epsilon}_{zz}\\ 0 & -\frac{1}{2}(l_1 - m_1)(\boldsymbol{\epsilon}_{xx} - \boldsymbol{\epsilon}_{yy}) - in_1\boldsymbol{\epsilon}_{xy}\\ 0 & -\frac{n_2(\boldsymbol{\epsilon}_{xz} + i\boldsymbol{\epsilon}_{yz})}{\sqrt{2}} \end{bmatrix}$$

$$g_{1}(\mathbf{k}) = \begin{bmatrix} E'_{\mathbf{c}} & -\frac{P_{2}}{\sqrt{2}}k_{+} & \frac{P_{2}}{\sqrt{2}}k_{-} & P_{1}k_{z} \\ -\frac{P_{2}}{\sqrt{2}}k_{-} & E'_{\mathbf{v}} & 0 & 0 \\ \frac{P_{2}}{\sqrt{2}}k_{+} & 0 & E'_{\mathbf{v}} & 0 \\ P_{1}k_{z} & 0 & 0 & E'_{\mathbf{v}} \end{bmatrix},$$
(A3)

$$\begin{bmatrix} 0 & 0 \\ -\frac{1}{2}N_1'k_-^2 & -\frac{1}{\sqrt{2}}N_2'k_-k_z \\ \left(\frac{L_1'+M_1}{2}\right)\left(k_x^2+k_y^2\right) + M_2k_z^2 & \frac{1}{\sqrt{2}}N_2'k_+k_z \\ \frac{1}{\sqrt{2}}N_2'k_-k_z & M_3\left(k_x^2+k_y^2\right) + L_2'k_z^2 \end{bmatrix},$$

$$\begin{array}{cccc}
0 & 0 \\
-\frac{1}{2}(l_1 - m_1)(\boldsymbol{\epsilon}_{xx} - \boldsymbol{\epsilon}_{yy}) + in_1\boldsymbol{\epsilon}_{xy} & -\frac{n_2(\boldsymbol{\epsilon}_{xz} - i\boldsymbol{\epsilon}_{yz})}{\sqrt{2}} \\
\frac{1}{2}(l_1 + m_1)(\boldsymbol{\epsilon}_{xx} + \boldsymbol{\epsilon}_{yy}) + m_2\boldsymbol{\epsilon}_{zz} & \frac{n_2(\boldsymbol{\epsilon}_{xz} + i\boldsymbol{\epsilon}_{yz})}{\sqrt{2}} \\
& \frac{n_2(\boldsymbol{\epsilon}_{xz} - i\boldsymbol{\epsilon}_{yz})}{\sqrt{2}} & m_3(\boldsymbol{\epsilon}_{xx} + \boldsymbol{\epsilon}_{yy}) + l_2\boldsymbol{\epsilon}_{zz}
\end{array}$$
(A4)

Here, Δ_{cr} and Δ_{so} are crystal field splitting and spin-orbit coupling, respectively. The band edges are $E'_c = E_v + E_g + \Delta_{cr} + \frac{\Delta_{so}}{\beta} + \varphi$ and $E'_v = E_v + \varphi$, where φ is any additional scalar potential such as the piezoelectric potential. g_{st} is the contribution to the Hamiltonian due to strain ϵ_{ij} . The $\mathbf{k} \cdot \mathbf{p}$ parameters A'_i are related to the Kane parameters P_i as

$$\begin{split} A_1' &= \frac{\hbar^2}{2m_{\rm e}^{\parallel}} - \frac{P_1^2}{E_{\rm g}}, \\ A_2' &= \frac{\hbar^2}{2m_{\rm e}^{\perp}} - \frac{P_2^2}{E_{\rm g}}, \end{split}$$

where

$$\begin{split} P_1^2 &= \frac{\hbar^2}{2m_0} \left(\frac{m_0}{m_{\text{e}}^{\parallel}} - 1 \right) \frac{3E_{\text{g}} (\Delta_{\text{so}} + E_{\text{g}}) + \Delta_{\text{cr}} (2\Delta_{\text{so}} + 3E_{\text{g}})}{2\Delta_{\text{so}} + 3E_{\text{g}}}, \\ P_2^2 &= \frac{\hbar^2}{2m_0} \left(\frac{m_0}{m_{\text{e}}^{\perp}} - 1 \right) \frac{E_{\text{g}} [3E_{\text{g}} (\Delta_{\text{so}} + E_{\text{g}})] + \Delta_{\text{cr}} (2\Delta_{\text{so}} + 3E_{\text{g}})}{\Delta_{\text{cr}} \Delta_{\text{so}} + 3\Delta_{\text{cr}} E_{\text{g}} + 2\Delta_{\text{so}} E_{\text{g}} + E_{\text{g}}^2}. \end{split}$$

Here, m_{e}^{\parallel} and m_{e}^{\perp} are the electron effective masses along the z-axis and in the xy-plane, respectively. The Luttinger-like parameters L'_{i} , M_{i} , and N'_{i} are related to the A_{i} parameters by

$$\begin{split} L_1' &= \frac{\hbar^2}{2m_0} \left(A_2 + A_4 + A_5 \right) + \frac{P_2^2}{E_g} \\ L_2' &= \frac{\hbar^2}{2m_0} A_1 + \frac{P_1^2}{E_g}, \\ M_1 &= \frac{\hbar^2}{2m_0} \left(A_2 + A_4 - A_5 \right), \\ M_2 &= \frac{\hbar^2}{2m_0} \left(A_1 + A_3 \right), \\ M_3 &= \frac{\hbar^2}{2m_0} A_2, \\ N_1' &= \frac{\hbar^2}{2m_0} 2A_5 + \frac{P_2^2}{E_g}, \\ N_2' &= \frac{\hbar^2}{2m_0} \sqrt{2}A_6 + \frac{P_1P_2}{E_g}. \end{split}$$

Note that there is an error in the relations for L'_1 , L'_2 , and N'_1 in Ref. 10, which we have corrected in agreement with Appendix B of Ref. 41. The parameters A_i , P_i , and E_g used in our numerical study of InGaN systems are given in Table III.

Similar to the $\mathbf{k} \cdot \mathbf{p}$ parameters L'_i , M_i , and N'_i , the strain parameters are as follows:

$$l_{1} = D_{2} + D_{4} + D_{5},$$

$$l_{2} = D_{1},$$

$$m_{1} = D_{2} + D_{4} - D_{5},$$

$$m_{2} = D_{1} + D_{3},$$

$$m_{2} = D_{1} + D_{3},$$

$$m_{3} = D_{2},$$

$$n_{1} = 2D_{5},$$

$$n_{2} = \sqrt{2}D_{6}.$$

where the deformation potentials D_i are listed in Table III.

Parameters	GaN	InN
a (Å)	3.189	3.545
c (Å)	5.185	5.703
C_{11} (GPa)	390	223
C_{12} (GPa)	145	115
C ₁₃ (GPa)	106	92
C ₃₃ (GPa)	398	224
C_{44} (GPa)	105	48
$e_{15} ({\rm C}/{\rm m}^2)$	0.326	0.264
e_{31} (C/m ²)	-0.527	-0.484
e_{33} (C/m ²)	0.895	1.06
$P_{\rm sp}~({\rm C}/{\rm m}^2)$	-0.034	-0.042
ε _r	9.8	13.8
E_{g} (eV)	3.51	0.78
E_{v} (eV)	0	0.5
Δ_{cr} (eV)	0.010	0.040
Δ_{so} (eV)	0.017	0.005
m_{\parallel}/m_0	0.20	0.07
m_{\perp}/m_0	0.20	0.07
A_1	-7.21	-8.21
A_2	-0.44	-0.68
A_3	6.68	7.57
A_4	-3.46	-5.23
A_5	-3.40	-5.11
A_6	-4.90	-5.96
$a_1 (eV)$	-4.9	-3.5
$a_2 (eV)$	-11.3	-3.5
D_1 (eV)	-3.7	-3.7
D_2 (eV)	4.5	4.5
D_3 (eV)	8.2	8.2
D_4 (eV)	-4.1	-4.1
D_5 (eV)	-4.0	-4.0
<i>D</i> ₆ (eV)	-5.5	-5.5

APPENDIX B: FOURIER AND CONVOLUTION CONVENTIONS

We define the Fourier forward and inverse transforms of a function $g(\mathbf{r})$ as

$$F\{g(\mathbf{r})\}(\mathbf{q}) \equiv \tilde{g}(\mathbf{q}) = \frac{1}{(2\pi)^3} \int_{-\infty}^{\infty} d^3 \mathbf{r} g(\mathbf{r}) e^{-i\mathbf{q}\cdot\mathbf{r}}$$
$$F^{-1}\{\tilde{g}(\mathbf{q})\}(\mathbf{r}) = g(\mathbf{r}) = \int_{-\infty}^{\infty} d^3 \mathbf{q} \,\tilde{g}(\mathbf{q}) e^{i\mathbf{q}\cdot\mathbf{r}}.$$

A convolution is denoted by

$$(f * g)(\mathbf{r}) = \int_{-\infty}^{\infty} f(\mathbf{r}')g(\mathbf{r} - \mathbf{r}')d\mathbf{r}'.$$

The convolution theorem states that

$$\left(\tilde{f} \star \tilde{g}\right)(\mathbf{q}) = F\{f(\mathbf{r})g(\mathbf{r})\}(\mathbf{q}).$$

For a system that is periodic in real space with a unit cell Ω of volume *V*, we define

$$\tilde{g}(\mathbf{q}) = \frac{1}{(2\pi)^3} \int_{\Omega} \mathsf{d}^3 \mathbf{r} \, g(\mathbf{r}) \mathsf{e}^{-i\mathbf{q}\cdot\mathbf{r}},$$

$$g(\mathbf{r}) = \frac{(2\pi)^3}{V} \sum_{\mathbf{q} \in \Omega^{-1}} \tilde{g}(\mathbf{q}) \mathbf{e}^{i\mathbf{q}\cdot\mathbf{r}},$$

where Ω^{-1} is the reciprocal space to the unit cell Ω . Defining the Fourier space convolution as

$$\left(\tilde{f} \star \tilde{g}\right)(\mathbf{q}) = \sum_{\mathbf{q}' \in \Omega^{-1}} \tilde{f}(\mathbf{q}') \tilde{g}(\mathbf{q} - \mathbf{q}'), \tag{B1}$$

the convolution theorem is

$$\left(\tilde{f} \star \tilde{g}\right)(\mathbf{q}) = \frac{V}{(2\pi)^3} F\{f(\mathbf{r})g(\mathbf{r})\}(\mathbf{q}).$$

Since we consider different superlattice unit cells for the electronic and strain properties, $(\tilde{f} * \tilde{g})_{e}$ indicates a convolution on the electronic space Ω_{e}^{-1} and $(\tilde{f} * \tilde{g})_{s}$ on the strain space Ω_{s}^{-1} . The reciprocal space Ω_{e}^{-1} contains a discrete infinity of wave

The reciprocal space Ω^{-1} contains a discrete infinity of wave vectors on which to evaluate \tilde{f} and \tilde{g} . The convolution in Eq. (B1) then sums over the infinite set of wave vectors, with \tilde{f} and \tilde{g} being functions that decay at large wave vectors. For the calculations in this article, we choose a finite number of wave vectors. By choosing this mesh to contain wave vectors sufficiently large to capture the decay of \tilde{f} and \tilde{g} , we can calculate the linear convolution in

Eq. (B1) to good approximation by padding the f and \tilde{g} arrays with zeros and performing a circular convolution, as defined below.

We denote the finite Fourier space mesh by $\mathbf{q}_{i_1i_2i_3}$ with $-m_j \leq i_j < m_j$ such that $N_j = 2m_j + 1$ is the dimension of the mesh in each direction. For convenience, we use the mapping $p_j = i_j + m_j + 1$, which runs from 1 to N_j . Evaluating a function \tilde{f} on the mesh $\mathbf{q}_{p_1p_2p_3}$ gives the array $\tilde{f}_{p_1p_2p_3}$. The Fourier space array $\tilde{f}_{p_1p_2p_3}$ and its real-space counterpart $f_{\nu_1\nu_2\nu_3}$ are then related through the discrete Fourier transform and its inverse,

$$\tilde{f}_{p_1 p_2 p_3} = F\{f\}_{p_1 p_2 p_3}$$

$$= \frac{1}{(2\pi)^3} \sum_{\nu_1, \nu_2, \nu_3 = 1}^{N_1, N_2, N_3} f_{\nu_1 \nu_2 \nu_3} e^{-i2\pi (p_1 \nu_1 / N_1 + p_2 \nu_2 / N_2 + p_3 \nu_3 / N_3)}, \quad (B2)$$

$$f_{\nu_1\nu_2\nu_3} = F^{-1}\left\{\tilde{f}\right\}_{\nu_1\nu_2\nu_3} = \frac{(2\pi)^3}{N} \sum_{p_1,p_2,p_3=1}^{N_1,N_2,N_3} \tilde{f}_{p_1p_2p_3} e^{i2\pi(p_1\nu_1/N_1+p_2\nu_2/N_2+p_3\nu_3/N_3)}, \quad (B3)$$

where $N = N_1 N_2 N_3$. The circular convolution is defined as

$$\left(\tilde{f}\cdot\tilde{g}\right)_{p_1p_2p_3} = \sum_{p_1',p_2',p_3'=1}^{N_1,N_2,N_3} \tilde{f}_{p_1'p_2'p_3'}\tilde{g}_{\left(p_1-p_1'\right),\left(p_2-p_2'\right),\left(p_3-p_3'\right)}$$

where $\tilde{g}_{(p_1+N_1),(p_2+N_2),(p_3+N_3)} = \tilde{g}_{p_1p_2p_3}$. The convolution theorem is then

$$\left(\tilde{f}\cdot\tilde{g}\right)_{p_1p_2p_3} = \frac{N}{\left(2\pi\right)^3}F\{fg\}_{p_1p_2p_3}$$

To perform a linear convolution, we pad the arrays with zeros, which increases the dimensions of the mesh to $N_j^p = 2N_j - 1$ and yields the padded array $\tilde{f}_{p_1p_2p_3}^p$ containing $N^p = N_1^p N_2^p N_3^p$ elements. The linear convolution of the original functions is contained in the circular convolution of the padded function

$$\left(\tilde{f} * \tilde{g}\right)_{p_1 p_2 p_3} = \left(\tilde{f}^{\mathsf{p}} \cdot \tilde{g}^{\mathsf{p}}\right)_{p_1 p_2 p_3}.$$
 (B4)

This result is independent of the basis used to generate the mesh and is valid in the case of hexagonal meshes.

APPENDIX C: CHARACTERISTIC FUNCTIONS

The characteristic function of a single dot is unity inside the dot and zero outside,

$$\chi_{\mathsf{d}}(\mathbf{r}) = \begin{cases} 1, & \mathbf{r} \in \Omega_{\mathsf{d}}, \\ 0, & \text{otherwise,} \end{cases}$$

where Ω_d is the space inside the dot. For a cylindrical dot centered on the origin with radius *R* and height *h* along the z-axis, the

Fourier transform of χ_d is

$$\tilde{\chi}_{\mathsf{d}}(\mathbf{q}) = \frac{1}{(2\pi)^3} \frac{4\pi R}{q_3 \sqrt{q_x^2 + q_y^2}} \sin\left(\frac{h}{2}q_3\right) J_1\left(R\sqrt{q_x^2 + q_y^2}\right).$$

The characteristic function of the electronic cell, the hexagonal prism shown in Fig. 1, is defined as

$$\chi_{\mathsf{e}}(\mathbf{r}) = \begin{cases} 1, & \mathbf{r} \in \Omega_{\mathsf{e}}, \\ 0, & \text{otherwise,} \end{cases}$$

and its Fourier transform is

$$\tilde{\chi}_{\mathsf{e}}(\mathbf{q}) = \frac{1}{(2\pi)^3} L_3 \operatorname{sinc}\left(q_3 \frac{L_3}{2}\right) \\ \times \frac{q_1 \cos\left(\frac{L_{12}q_1}{2}\right) + q_2 \cos\left(\frac{L_{12}q_1}{2}\right) - (q_1 + q_2) \cos\left(L_{12} \frac{q_1 + q_2}{2}\right)}{\sqrt{3}q_1q_2(q_1 + q_2)}$$

APPENDIX D: QD k · p HAMILTONIAN

For the QD superlattice system, the $\mathbf{k} \cdot \mathbf{p}$ Hamiltonian matrix elements of Eq. (20) in the symmetry adapted basis are given in terms of parameters $f_{\alpha'\alpha}^d$ and $f_{\alpha'\alpha}^h$ that make up the bulk $\mathbf{k} \cdot \mathbf{p}$ Hamiltonian matrix elements $H_{\alpha'\alpha}$, as presented in Sec. III. We take the convention where superscript "(0)" indicates a bulk Hamiltonian matrix element containing no wave vector, "(*i*)" indicates a single wave vector k_i , and "(*i*, *j*)" indicate two wave vectors k_i and k_j . By defining $\phi = \frac{2\pi}{6}$ and

$$\begin{split} S^{ll'm_f}_{\alpha',\alpha} &= e^{i\phi\left\{l\left[m_f - J_z(\alpha)\right] - l'\left[m_f - J_z(\alpha')\right]\right\}},\\ S^{l'm_f}_{\alpha'} &= e^{-il'\phi\left[m_f - J_z(\alpha')\right]}, \end{split}$$

the QD Hamiltonian matrix elements are

$$\begin{aligned} \mathcal{H}_{m_{f}\alpha'\alpha}^{(0)}(\mathbf{q}',\mathbf{q}) &= \frac{1}{6} \sum_{l=0}^{5} \sum_{l=0}^{5} S_{\alpha',\alpha}^{ll'm_{f}} h_{\alpha'\alpha} \Big(\overset{\leftrightarrow}{\mathbf{R}}_{l'}\mathbf{q}', \overset{\leftrightarrow}{\mathbf{R}}_{l} \mathbf{q} \Big), \\ \mathcal{H}_{m_{f}\alpha'\alpha}^{(i)}(\mathbf{q}',\mathbf{q}) &= \frac{1}{6} \sum_{l=0}^{5} \sum_{l=0}^{5} S_{\alpha',\alpha}^{ll'm_{f}} \frac{\Big(\overset{\leftrightarrow}{\mathbf{R}}_{l'}\mathbf{q}' \Big)_{i} + \Big(\overset{\leftrightarrow}{\mathbf{R}}_{l} \mathbf{q} \Big)_{i}}{2} h_{\alpha'\alpha} \Big(\overset{\leftrightarrow}{\mathbf{R}}_{l'}\mathbf{q}', \overset{\leftrightarrow}{\mathbf{R}}_{l} \mathbf{q} \Big), \end{aligned}$$

$$\begin{aligned} \mathcal{H}_{m_{f}\alpha'\alpha}^{(i,j)}(\mathbf{q}',\mathbf{q}) = &\frac{1}{6} \sum_{l'=0}^{5} \sum_{l=0}^{5} S_{\alpha',\alpha}^{ll'm_{f}} \frac{\left(\overset{\leftrightarrow}{\mathbf{R}}_{l}\mathbf{q}\right)_{j} \left(\overset{\leftrightarrow}{\mathbf{R}}_{l'}\mathbf{q}'\right)_{i} + \left(\overset{\leftrightarrow}{\mathbf{R}}_{l}\mathbf{q}\right)_{i} \left(\overset{\leftrightarrow}{\mathbf{R}}_{l'}\mathbf{q}'\right)_{j}}{2} \\ &\times h_{\alpha'\alpha} \left(\overset{\leftrightarrow}{\mathbf{R}}_{l'}\mathbf{q}', \overset{\leftrightarrow}{\mathbf{R}}_{l}\mathbf{q}\right), \end{aligned}$$

$$\begin{split} \mathcal{H}_{m_{f}\alpha'\alpha}^{(0)}(\mathbf{q}',\mathbf{q}_{z}) &= \frac{1}{\sqrt{6}} \sum_{l'=0}^{5} S_{\alpha'}^{l'm_{f}} h_{\alpha'\alpha} \Big(\overrightarrow{\mathbf{R}}_{l'}\mathbf{q}',\mathbf{q}_{z} \Big), \\ \mathcal{H}_{m_{f}\alpha'\alpha}^{(i)}(\mathbf{q}',\mathbf{q}_{z}) &= \frac{1}{\sqrt{6}} \sum_{l'=0}^{5} S_{\alpha'}^{l'm_{f}} \frac{\left(\overrightarrow{\mathbf{R}}_{l'}\mathbf{q}' \right)_{i} + (\mathbf{q}_{z})_{i}}{2} h_{\alpha'\alpha} \Big(\overrightarrow{\mathbf{R}}_{l'}\mathbf{q}',\mathbf{q}_{z} \Big), \end{split}$$

$$\begin{split} \mathcal{H}_{m_{f}\alpha'\alpha}^{(i,j)}(\mathbf{q}',\mathbf{q}_z) = & \frac{1}{\sqrt{6}} \sum_{l'=0}^{5} S_{\alpha'}^{l'm_f} \frac{(\mathbf{q}_z)_j (\mathbf{R}_l' \mathbf{q}')_i + (\mathbf{q}_z)_i (\mathbf{R}_l' \mathbf{q}')_j}{2} \\ & \times h_{\alpha'\alpha} (\overrightarrow{\mathbf{R}}_{l'} \mathbf{q}', \mathbf{q}_z), \\ \mathcal{H}_{m_{f}\alpha'\alpha}^{(0)} (\mathbf{q}_z', \mathbf{q}_z) = h_{\alpha'\alpha} (\mathbf{q}_z', \mathbf{q}_z), \\ \mathcal{H}_{m_{f}\alpha'\alpha}^{(0)} (\mathbf{q}_z', \mathbf{q}_z) = \frac{(\mathbf{q}_z')_i + (\mathbf{q}_z)_i}{2} h_{\alpha'\alpha} (\mathbf{q}_z', \mathbf{q}_z), \\ \mathcal{H}_{m_{f}\alpha'\alpha}^{(i,j)} (\mathbf{q}_z', \mathbf{q}_z) = \frac{(\mathbf{q}_z)_j (\mathbf{q}_z')_i + (\mathbf{q}_z)_i (\mathbf{q}_z')_j}{2} h_{\alpha'\alpha} (\mathbf{q}_z', \mathbf{q}_z), \end{split}$$

 (\leftrightarrow)

 (\leftrightarrow)

where

$$h_{\alpha'\alpha}\left(\overset{\leftrightarrow}{\mathbf{R}}_{l'}\mathbf{q}',\overset{\leftrightarrow}{\mathbf{R}}_{l}\mathbf{q}\right) = f^{\mathsf{h}}_{\alpha'\alpha}\delta_{l,l'}\delta_{q,q'} + \frac{\left(2\pi\right)^{3}\left(f^{\mathsf{d}}_{\alpha'\alpha} - f^{\mathsf{h}}_{\alpha'\alpha}\right)}{V_{e}}\tilde{\chi}_{\mathsf{d}}\left(\overset{\leftrightarrow}{\mathbf{R}}_{l'}\mathbf{q}' - \overset{\leftrightarrow}{\mathbf{R}}_{l}\mathbf{q}\right).$$

The contributions of the piezoelectric potential to the Hamiltonian are

$$\mathcal{H}_{m_{f}\alpha'\alpha}^{\mathsf{pz}}(\mathbf{q}',\mathbf{q}) = -\delta_{\alpha',\alpha} \frac{(2\pi)^{3} e_{\mathsf{c}}}{6V_{e}} \sum_{l'=0}^{5} \sum_{l=0}^{5} e^{i\phi(l-l') \left[m_{f}-J_{z}(\alpha)\right]} \\ \times \tilde{\varphi}^{\mathsf{a}} \left(\stackrel{\leftrightarrow}{\mathbf{R}}_{l'} \mathbf{q}' - \stackrel{\leftrightarrow}{\mathbf{R}}_{l} \mathbf{q}\right), \tag{D1}$$

$$\mathcal{H}_{m_{f}\alpha'\alpha}^{\mathsf{pz}}(\mathbf{q}',\mathbf{q}_{z}) = -\delta_{\alpha',\alpha} \frac{(2\pi)^{3} e_{\mathbf{c}}}{V_{e}\sqrt{6}} \sum_{l'=0}^{5} e^{-il'\phi[m_{f}-J_{z}(\alpha')]} \tilde{\varphi}^{\mathbf{a}} \Big(\overset{\leftrightarrow}{\mathbf{R}}_{l'}\mathbf{q}'-\mathbf{q}_{z}\Big),$$
(D2)

$$\mathcal{H}_{m_{f}\alpha'\alpha}^{\mathsf{pz}}(\mathbf{q}_{z}',\mathbf{q}_{z}) = -\delta_{\alpha',\alpha} \frac{(2\pi)^{3} e_{\mathsf{c}}}{V_{e}} \tilde{\varphi}^{\mathsf{a}}(\mathbf{q}_{z}'-\mathbf{q}_{z}), \tag{D3}$$

where $\tilde{\varphi}^{a}$ is described in Secs. II C and III B and e_{c} is the electric charge.

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

REFERENCES

¹H. P. T. Nguyen, S. Zhang, K. Cui, X. Han, S. Fathololoumi, M. Couillard, G. A. Botton, and Z. Mi, Nano Lett. **11**, 1919 (2011).

²T. J. Puchtler, T. Wang, C. X. Ren, F. Tang, R. A. Oliver, R. A. Taylor, and T. Zhu, Nano Lett. **16**, 7779 (2016).

³M. G. Kibria, H. P. Nguyen, K. Cui, S. Zhao, D. Liu, H. Guo, M. L. Trudeau, S. Paradis, A. R. Hakima, and Z. Mi, <u>ACS Nano 7</u>, 7886 (2013).

⁴L. Sang, M. Liao, Q. Liang, M. Takeguchi, B. Dierre, B. Shen, T. Sekiguchi, Y. Koide, and M. Sumiya, Adv. Mater. 26, 1414 (2014).

⁵R. Cheriton, S. M. Sadaf, L. Robichaud, J. J. Krich, Z. Mi, and K. Hinzer, Commun. Mater. 1, 63 (2020).

- ⁶K. M. Gambaryan, T. Boeck, A. Trampert, and O. Marquardt, ACS Appl. Electron. Mater. **2**, 646 (2020).
- ⁷M. A. der Maur, A. Pecchia, G. Penazzi, F. Sacconi, and A. D. Carlo, J. Comput. Electron. 12, 553 (2013).

⁸A. D. Andreev and E. P. O'Reilly, Phys. Rev. B **62**, 15851 (2000).

- ⁹V. A. Fonoberov and A. A. Balandin, J. Appl. Phys. **94**, 7178 (2003).
- ¹⁰M. Winkelnkemper, A. Schliwa, and D. Bimberg, Phys. Rev. B 74, 155322 (2006).

¹¹T. Saito and Y. Arakawa, Physica E **15**, 169 (2002).

¹²S. Schulz, M. A. Caro, C. Coughlan, and E. P. O'Reilly, Phys. Rev. B 91, 035439 (2015).

¹³R. M. Martin, *Electronic Structure: Basic Theory and Practical Methods*, 2nd ed. (Cambridge University Press, 2020).

¹⁴D. R. Bowler and T. Miyazaki, Rep. Prog. Phys. 75, 036503 (2012).

 ¹⁵O. Marquardt, D. Mourad, S. Schulz, T. Hickel, G. Czycholl, and J. Neugebauer, *Phys. Rev. B* 78, 235302 (2008).
 ¹⁶A. V. Rodina, A. Y. Alekseev, A. L. Efros, M. Rosen, and B. K. Meyer, *Phys.*

¹⁰A. V. Rodina, A. Y. Alekseev, A. L. Efros, M. Rosen, and B. K. Meyer, Phys. Rev. B **65**, 125302 (2002).

¹⁷B. A. Foreman, Phys. Rev. B 75, 235331 (2007).

¹⁸N. Vukmirović, D. Indjin, V. D. Jovanović, Z. Ikonić, and P. Harrison, Phys. Rev. B 72, 075356 (2005).

¹⁹S. Tomić, A. G. Sunderland, and I. J. Bush, J. Mater. Chem. 16, 1963 (2006).

²⁰N. Vukmirovć and S. Tomić, J. Appl. Phys. 103, 103718 (2008).

²¹S. Tomić, T. Sogabe, and Y. Okada, Prog. Photovoltaics Res. Appl. 23, 546 (2015).

²²N. S. Beattie, P. See, G. Zoppi, P. M. Ushasree, M. Duchamp, I. Farrer, D. A. Ritchie, and S. Tomić, ACS Photonics 4, 2745 (2017).

²³O. Marquardt, S. Boeck, C. Freysoldt, T. Hickel, and J. Neugebauer, Comput. Phys. Commun. **181**, 765 (2010).

²⁴O. Marquardt, S. Boeck, C. Freysoldt, T. Hickel, S. Schulz, J. Neugebauer, and E. P. O'Reilly, Comput. Mater. Sci. **95**, 280 (2014).

²⁵N. Vukmirović, Z. Ikonić, D. Indjin, and P. Harrison, J. Phys.: Condens. Matter 18, 6249 (2006).

²⁶A. D. Andreev, J. R. Downes, D. A. Faux, and E. P. O'Reilly, J. Appl. Phys. 86, 297 (1999).

27S. Tomić and N. Vukmirović, Phys. Rev. B 79, 245330 (2009).

²⁸J. Renard, R. Songmuang, G. Tourbot, C. Bougerol, B. Daudin, and B. Gayral, Phys. Rev. B **80**, 121305 (2009).

²⁹M. J. Holmes, K. Choi, S. Kako, M. Arita, and Y. Arakawa, Nano Lett. 14, 982 (2014).

³⁰A. V. Nenashev, A. A. Koshkarev, and A. V. Dvurechenskii, J. Appl. Phys. **123**, 105104 (2018).

³¹F. Bernardini, V. Fiorentini, and D. Vanderbilt, Phys. Rev. B 56, R10024 (1997).

32A. Zoroddu, F. Bernardini, P. Ruggerone, and V. Fiorentini, Phys. Rev. B 64, 045208 (2001).

³³S. F. Chichibu, A. C. Abare, M. S. Minsky, S. Keller, S. B. Fleischer, J. E. Bowers, E. Hu, U. K. Mishra, L. A. Coldren, S. P. DenBaars, and T. Sota, Appl. Phys. Lett. **73**, 2006 (1998).

³⁴J. P. Ibbetson, P. T. Fini, K. D. Ness, S. P. DenBaars, J. S. Speck, and U. K. Mishra, Appl. Phys. Lett. 77, 250 (2000).

³⁵H.-M. Kim, Y.-H. Cho, H. Lee, S. I. Kim, S. R. Ryu, D. Y. Kim, T. W. Kang, and K. S. Chung, Nano Lett. 4, 1059 (2004).

³⁶S. L. Chuang and C. S. Chang, Phys. Rev. B 54, 2491 (1996).

³⁷M. J. Jocić and N. Vukmirović, Phys. Rev. B 102, 085121 (2020).

³⁸R. A. Morrow and K. R. Brownstein, Phys. Rev. B 30, 678 (1984).

³⁹S. Tomić, P. Howe, N. M. Harrison, and T. S. Jones, J. Appl. Phys. **99**, 093522 (2006).

⁴⁰M. A. Caro, S. Schulz, and E. P. O'Reilly, Phys. Rev. B **88**, 214103 (2013).

⁽⁴⁾E. Berkowicz, D. Gershoni, G. Bahir, E. Lakin, D. Shilo, E. Zolotoyabko, A. C. Abare, S. P. Denbaars, and L. A. Coldren, Phys. Rev. B 61, 010994 (2000).