Electronic and optical properties of GaN/AlN quantum dots with adjacent threading dislocations*

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We present a theory to simulate a coherent GaN QD with an adjacent pure edge threading dislocation by using a finite element method. The piezoelectric effects and the strain modified band edges are investigated in the framework of multi-band $\boldsymbol{k}\cdot\boldsymbol{p}$ theory to calculate the electron and the heavy hole energy levels. The linear optical absorption coefficients corresponding to the interband ground state transition are obtained via the density matrix approach and perturbation expansion method. The results indicate that the strain distribution of the threading dislocation affects the electronic structure. Moreover, the ground state transition behaviour is also influenced by the position of the adjacent threading dislocation.

Keywords: quantum dot, threading dislocation, electronic structure, absorption efficiency

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1. Introduction

During the last decade, semiconductor quantum-dot (QD) heterostructures have been a subject of extensive research due to their applications in fabricating a wide variety of optoelectronic and microelectronic devices. In particular, nitride QDs have potential applications both in the visible and ultraviolet energy ranges. Stranski–Krastanow GaN QDs grown on AlN have been realized by molecular beam epitaxy (MBE) and metal organic chemical vapour deposition (MOCVD) techniques.[1,2] GaN and relevant nitride alloys generally have a hexagonal structure, which leads to a strong built-in piezoelectric field in the heterostructure. Consequently, self-organized GaN/AlN QDs can exhibit a large red shift at an energy of the photoluminescence, about 0.5 eV below the GaN bulk bandgap.[3] Large built-in electric fields of pyroelectric and piezoelectric materials make the GaN/AlN QDs interesting candidates in quantum information processing.[4] Recently, the preferential nucleation of GaN QDs at the edge of AlN threading dislocations has been observed via high resolution transmission electron microscopy (HRTEM).[5] The large strain near the edge threading dislocation may serve as the QD nucleation template. However, most of the GaN QDs are coherent, which can be supported by the intense and temperature-independent photoluminescence (PL) spectrum. Obviously, it is important to consider both piezoelectric and dislocation-induced strain effects in the theoretic analysis of electronic and optical properties.

In a series of previous articles and letters,[6−9] the electronic structure of hexagonal GaN/AlN QD structures has been studied, including the strain effects and the piezoelectric effects. The main aim of the present work is to study more clearly how the adjacent threading dislocation influences the electronic structures and optical properties. We present a method to simultaneously simulate the adjacent pure edge threading dislocation within QD heterostructures by using a finite element method (FEM). The interaction between strain fields introduced by threading dislocation and by misfit is taken into consideration. Then, the piezoelectric effects and the strain modified band edges are investigated. Based on the single-band effective mass approximation, we calculate the wavefunction and the energy levels of the electron and heavy hole, from which the linear optical absorption coefficients

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can also be obtained. The results are compared with the experimental data available and the previous theoretical results.

2. Numerical calculation

As presented in Fig. 1, a truncated hexagonal GaN QD embedded in AlN matrix, together with an adjacent edge threading dislocation, is considered in this work. In our simulation, Cartesian coordinates are adopted. The x-direction corresponds to the $[11\bar{2}0]$ direction and the epitaxial growth is along the [0001] direction with the interface at $z = 0$. The QD size is determined by the diameters of the circles that circumscribe the polygons at the bottom and the top, $R_{\rm b}$ and $R_{\rm t}$. $H_{\rm dot}$ is the sum of QD height and thickness of the wetting layer. The geometry varies with QD height as follows: the wetting layer width is kept at a constant of 0.5 nm and the QD height is varied from 1.8 nm to 3.6 nm. The top diameter equals the total QD height $2R_{\rm t} = H_{\rm dot}$ while the bottom diameter $2R_{\rm b}$ varies linearly with QD height from $R_{\rm b} = 8.5$ nm to $R_{\rm b} = 4.0$ nm. The dimensions of the substrate and the capping layer in this model are both set to be 30×30×15 nm. The threading dislocation is aligned along one of the edge of the coherent QD. Calculations are carried out in the framework of anisotropic elasticity, with the adoption of different elastic constants for the quantum dot, substrate and capping layer. All the material parameters for the calculation are cited from Ref. [9].

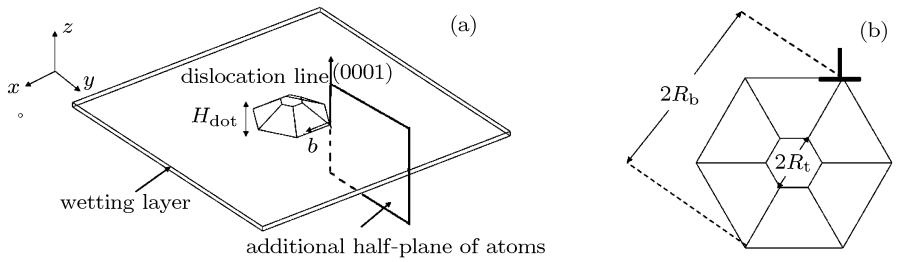


Fig. 1. A three-dimensional (3D) view of GaN/AlN QD with threading dislocation (a) and view of the QD structure in the x–y plane (b).

2.1. Strain distributions

We use the stiffness matrix to describe the dependence of the strain component on the stress component. For the wurtzite crystal and coordinates adopted, this dependence is expressed as

$$\begin{pmatrix} \sigma_{xx} \\ \sigma_{yy} \\ \sigma_{zz} \\ \sigma_{xy} \\ \sigma_{yz} \\ \sigma_{xz} \end{pmatrix} = \begin{bmatrix} C_{11} & C_{12} & C_{13} & 0 & 0 & 0 \\ C_{12} & C_{11} & C_{13} & 0 & 0 & 0 \\ C_{13} & C_{13} & C_{33} & 0 & 0 & 0 \\ 0 & 0 & 0 & \frac{(C_{11}-C_{12})}{2} & 0 & 0 \\ 0 & 0 & 0 & 0 & C_{44} & 0 \\ 0 & 0 & 0 & 0 & 0 & C_{44} \end{bmatrix} \begin{pmatrix} \varepsilon_{xx} - \varepsilon^0_{xx} \\ \varepsilon_{yy} - \varepsilon^0_{yy} \\ \varepsilon_{zz} - \varepsilon^0_{zz} \\ \varepsilon_{xy} \\ \varepsilon_{yz} \\ \varepsilon_{xz} \end{pmatrix} + \sigma^0, \tag{1}$$

where the components of ε^0 are the initial strain used in the FEM calculation, while the components of σ^0 are the initial stress, which are set to zero.

To simulate the strained GaN QD, coherency is imposed in the quantum dot by initial strains. The initial strain components in the x-, y-, and z-directions equal the lattice mismatch

$$\varepsilon^0_{ij} = \begin{cases} \varepsilon^0_{xx} = \varepsilon^0_{yy} = \dfrac{a_{\rm sub} - a_{\rm film}}{a_{\rm film}}, \\ \varepsilon^0_{zz} = \dfrac{c_{\rm sub} - c_{\rm film}}{c_{\rm film}}, \\ \varepsilon^0_{ij} = 0 \quad i \neq j. \end{cases} \tag{2}$$

Then, we add a pure-edge threading dislocation with a Burgers vector of $b_{\rm td} = \frac{1}{3}a[11\bar{2}0]$ into the model as shown in Fig. 1. This type of dislocation can be simulated by feeding an extra initial strain, which corresponds to the displacement of an extra half-plane of atoms, in a region extending from the bottom of the substrate up to the top of the capping layer. This specific region is along the dislocation line [0001] and its width equals the lattice constant in $[11\bar{2}0]$. The extra initial strain is defined as[11]

$$\varepsilon_{\rm td}^0 = [(a[11\bar{2}0] + b_{\rm td}) - a[11\bar{2}0]]/(a[11\bar{2}0] + b_{\rm td}). \quad (3)$$

In the present work, only the strain effect of the threading dislocation is taken into consideration.

2.2. Piezoelectric effects

The piezoelectric effects in the GaN/AlN system are considerably stronger than those observed in cubic GaAs-based structures.[12] This built-in piezoelectric field significantly modifies the electronic structure. The piezoelectric potential can be obtained by solving the Poisson equation

$$-\varepsilon_0 \nabla \cdot (\varepsilon_{\rm r}(r)\nabla V_{\rm P}) = \rho, \quad (4)$$

where $\varepsilon_{\rm r}$ is the relative dielectric constant, and ρ is the piezoelectric charge density and is expressed as

$$\rho\,(r) = -\boldsymbol{\nabla} \cdot (\boldsymbol{P}_{\rm strain} + \boldsymbol{P}_{\rm spont})\,, \quad (5)$$

where $\boldsymbol{P}_{\rm strain}$ and $\boldsymbol{P}_{\rm spont}$ are the strain-induced polarization and the spontaneous polarization respectively. For GaN and AlN materials the strain-induced polarization can be expressed as $P_i^{\rm strain} = \sum_{lm} e_{ilm}(r)\varepsilon_{lm}(r)$, in the common matrix representation it is written as

$$\begin{pmatrix} P_x^{\rm strain} \\ P_y^{\rm strain} \\ P_z^{\rm strain} \end{pmatrix} = \begin{pmatrix} e_{11} & e_{12} & e_{13} & e_{14} & e_{15} & e_{16} \\ e_{21} & e_{22} & e_{23} & e_{24} & e_{25} & e_{26} \\ e_{31} & e_{32} & e_{33} & e_{34} & e_{35} & e_{36} \end{pmatrix} \begin{pmatrix} \varepsilon_{xx} \\ \varepsilon_{yy} \\ \varepsilon_{zz} \\ 2\varepsilon_{yz} \\ 2\varepsilon_{zx} \\ 2\varepsilon_{xy} \end{pmatrix}. \quad (6)$$

Only three components of the piezoelectric matrix are nonzero, $e_{31} = e_{32}$, e_{33}, and $e_{24} = e_{15}$.

2.3. Electronic structure

The single band effective-mass Hamiltonians for electron and heavy hole are assumed, respectively, to be expressed as[6,13]

$$H_{\rm e} = -\frac{\hbar^2}{2m_{\rm e}^*}\nabla^2 + E_c + \delta E_c, \quad (7)$$

$$H_{\rm hole} = -\nabla_z \cdot (A_1 + A_3)\,\nabla_z - \nabla_{x,y} \cdot (A_2 + A_4 - A_5)\,\nabla_{x,y} + E_v + \delta E_v, \quad (8)$$

where δE_c and δE_v are both the modified band edges, due to the strain and the piezoelectric effects. The modified band edges can be written as

$$\delta E_c = a_c^{\perp}\left(\varepsilon_{\rm hyd}^{\perp}\right) + a_c^{\parallel}\varepsilon_{\rm hyd}^{\parallel} - eV_p, \quad (9)$$

$$\delta E_v = (D_1 + D_3)\varepsilon_{\rm hyd}^{\parallel} + (D_2 + D_4 - D_5)\varepsilon_{\rm hyd}^{\perp} - eV_p. \quad (10)$$

The boundary conditions of the wave function are as follows: the Dirichlet boundary condition is applied to the outer boundaries while the Neumann boundary condition is used for the inner boundaries. Similar boundary conditions are used in solving the Poisson equation.

2.4. Optical absorption coefficients

To calculate the optical absorption coefficients of the QD with adjacent threading dislocation, we employ the density matrix approach and perturbation expansion method. For simplicity, only the optical transition between the ground states of the electron and heavy hole is considered. Considering the incident light propagating along the x-direction and polarizing in the z-direction, the time-dependent Schrödinger equation of the matrix operator ρ is given by

$$\frac{\partial \rho}{\partial t} = \frac{1}{\mathrm{i}\,\hbar}[H_0 - \boldsymbol{M} \cdot \boldsymbol{E}(t), \rho] - \frac{1}{2}[\Gamma(\rho - \rho^{(0)}) + (\rho - \rho^{(0)})\,\Gamma], \quad (11)$$

where ρ is the density matrix operator, H_0 is the unperturbed Hamiltonian, $\rho^{(0)}$ is the unperturbed density matrix operator, M is the dipole operator, and Γ is the phenomenological operator responsible for the damping due to the electron–phonon interaction and so on. We can expand ρ in series,[14,15] then the linear optical susceptibility can be written as

$$\varepsilon_0\chi^{(1)}(\omega) = \frac{|M_{21}|^2\,\sigma_V}{E_{21} - \hbar\omega - \mathrm{i}\,\hbar\gamma_{21}}. \quad (12)$$

3. Result and discussion

First we take into account only the mismatch strain and the piezoelectric effects to calculate the electronic structure for a coherent GaN QD. The bound states are determined by solving Eqs. (7) and

8) numerically. The calculated probability density distributions of electrons and holes for the ground state are shown in Fig. 2. As can be seen, the electron wavefunction is confined up to the top of the GaN QD, while the hole wavefunction is pushed to the wetting layer. This figure also shows that the wavefunctions are quite strongly confined in the centre of the dot. All these confinements derive from the large built-in electrostatic potential in the GaN/AlN system. The dependence of the ground state transition energy E1–H1 on the total QD height is shown in Fig. 3. Till now, few absorption spectra of GaN/AlN QDs, which correspond to the E1–H1 transition energy, have been reported. As a common treatment method, we take photoluminescence as a comparison, although the Stokes shift exists. For small dots of 2.3 nm in total height, the calculated transition energy nearly coincides with the peak of the PL spectrum,[3] while the calculated transition energy is 200 meV lower than that for large dots. Since the width of the quantum dot height distribution in the PL experiment is about

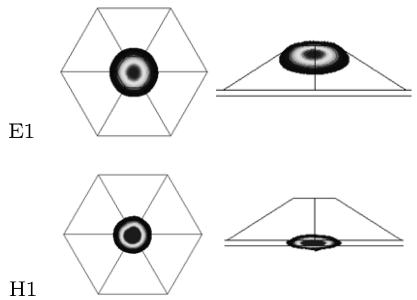


Fig. 2. Contours of probability densities for electron and hole ground states in a QD without dislocation, side view from the x–y plane and z–y plane respectively.

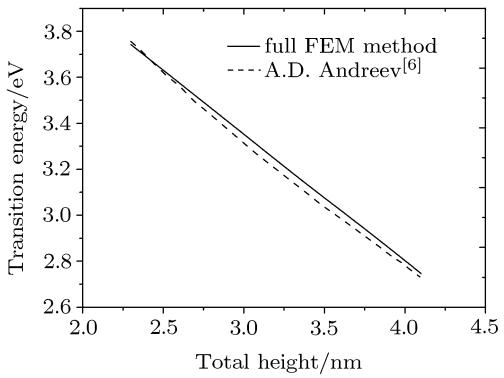


Fig. 3. Dependence of the ground state transition energy E1–H1 on the total QD height.

0.4 nm, our results are in reasonable agreement with available experimental results. Moreover, the calculated transition energy is satisfactorily consistent with that obtained from previous theory work.[6]

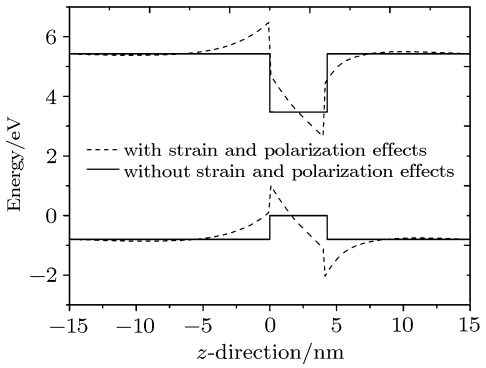


Fig. 4. Strain and polarization modified conduction band edge and heavy hole valence band edge along the [0001] direction.

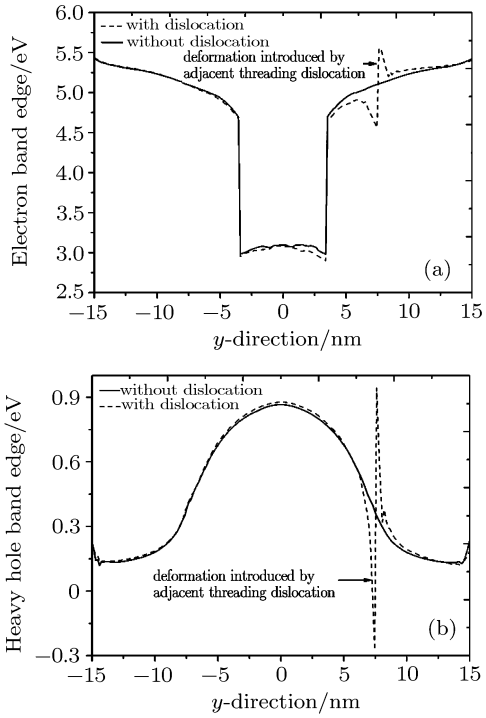


Fig. 5. (a) Electron band edge along the y-direction though the core of threading dislocation (a), and heavy hole band edge along the y-direction though the core of threading dislocation (b).

We now consider the effects of an adjacent threading dislocation on electronic structure. As an example, we introduce the adjacent threading dislocation in the centre of an edge of the large QD via the extra initial strain aforementioned. The dot height is 4.1 nm. The strain and polarization modified conduction band edge and heavy hole valence band edge along the central [0001] axis are shown in Fig. 4. The band edges are the same as the case without dislocation because the strain field of the dislocation becomes

trivial on the central axis of the QD. The electron and the hole states are still spatially separated. But in the lateral direction, the band edges are significantly modified near the threading dislocation due to the strong strain field near the core as illustrated in Fig. 5. The potential barrier between GaN QD and the AlN capping layer leads the electron states to be largely unaffected because of the presence of the dislocation. The threading dislocation affects the heavy hole states in a more direct way since the hole wavefunction is nearly confined in the wetting layer.

Figure 6 shows the calculated contours of constant probability densities for hole states in the large and the small QDs with an adjacent dislocation in the centre of edge. The deformation of the wavefunction of the heavy hole in a large dot is negligible while the deformation is obvious in a small dot. For a large dot, the transition energy reduces 29.7 meV when the dislocation appears at the corner of the QD edge, while the transition energy reduces 46.1 meV when the dislocation at the edge centre. For a small QD, the transition energies reduce 31.4 meV and 57.6 meV with the two different positions of dislocation. Our results are consistent with the decreased effect of the dislocation on the transition energy as dot size increases.[16] This phenomenon derives from two aspects: 1) the confinement induced by the piezoelectric effect is stronger for large dots; 2) the strong dislocation strain field which modifies the band edge decays within only several nanometres. Furthermore, it is obvious that the transition energy is also influenced by the position of adjacent edge dislocation. The changes of the transition energy, with and without the appearance of dislocation, are dependent on the location of the extra-half plane of atoms as demonstrated in Fig. 7. Moreover, the linear absorption coefficients for small and large QDs in the presence and the absence of threading dislocations are shown in Fig. 8. The magnitude of the absorption coefficient depends on the overlap of the electron and heavy hole wavefunctions. The spatial separation caused by the piezoelectric effects leads the absorption coefficient to be greatly reduced for a large QD. The positions of peaks for the different dislocation conditions correspond to the transition energies calculated. Interestingly, it can be seen that the magnitudes of the peaks are almost unchanged for dislocations appearing at different positions. Our method is still expected to be validated by relevant experiments.

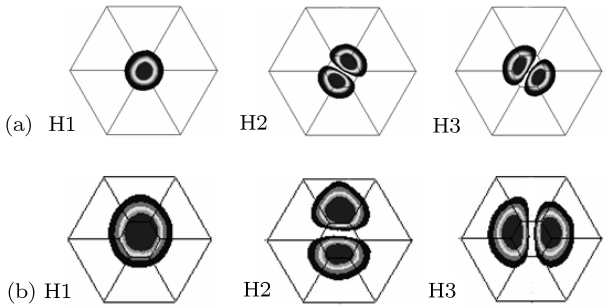


Fig. 6. (a) Contours of constant probability densities for hole states in a large dot with dislocation in the centre of one edge (a), and (b) contours of constant probability density for hole states in a small dot with dislocation in the centre of one edge.

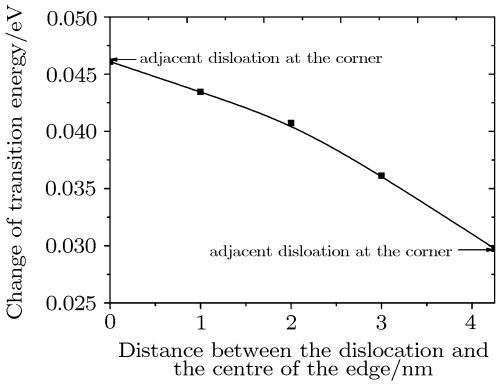


Fig. 7. Dependence of transition energy change on position of dislocation.

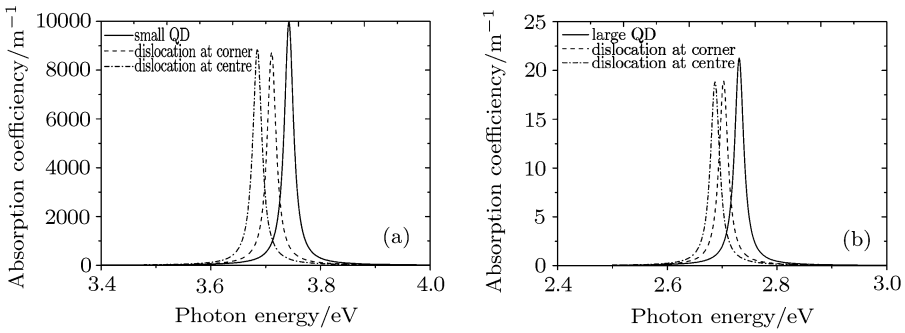


Fig. 8. Variations of linear absorption coefficients with photon energy for small (a) and large (b) QDs.

4. Conclusion

The strain components of a truncated hexagonal pyramid with an adjacent pure edge dislocation are calculated via continuum elastic theory. The threading dislocation is simulated by feeding an extra initial strain corresponding to an extra-half plane. Taking the influences of strain and piezoelectric effects into account, we solve the Schrödinger equation based on the single-band effective mass approximation. The effects of the dislocation are not so huge because the built-in electric potential laterally confines the electrons and holes to the centre of the dot. When the dot is relatively small, the heavy hole wavefunctions are deformed due to the occurrence of the dislocation. The interband ground state transition energy is affected not only by the occurring of the threading dislocation but also by the position of the dislocation.

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