Bound polaron in a strained wurtzite GaN/Al_xGa_{1-x}N cylindrical quantum dot*

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Abstract: Within the effective-mass approximation, a variational method is adopted to investigate the polaron effect in a strained $GaN/Al_xGa_{1-x}N$ cylindrical quantum dot. The electron couples with both branches of longitudinal optical-like (LO-like) and transverse optical-like (TO-like) phonons and the built-in electric field are taken into account. The numerical results show that the binding energy of the bound polaron is reduced obviously by the polaron effect on the impurity states. Furthermore, the contribution of LO-like phonons to the binding energy is dominant, and the anisotropic angle and Al content influence on the binding energy are small.

Key words:quantum dot;electron-phonon interaction;bound polaron;strainDOI:10.1088/1674-4926/32/6/062003PACC:7320D;7155;6320K

1. Introduction

III-V nitride semiconductors and their alloys with hexagonal wurtzite (WZ) crystal structures and complicated valence band structures are characterized by direct and large band gaps. The band gap energy of aluminum gallium indium nitride (Al-GaInN) varies between 6.2 and 1.9 eV, depending on its composition, at room temperature. Thus, GaN and related materials have been considered as promising systems for optical devices emitting light at wavelengths from ultraviolet to $red^{[1-4]}$. On the other hand, the group-III nitrides are commonly produced in the WZ crystal structure with a strong spontaneous macroscopic polarization. Moreover, due to the large lattice mismatch between GaN and AlGaN, the wurtzite GaN/AlGaN heterostructures can induce a remarkable piezoelectric polarization, which leads to a strong built-in electric field of the order of MV/cm in heterostructures. Such a strong field will give rise to a remarkable reduction in effective band gaps in quantum wells or quantum dots (QDs). Furthermore, the electronic, dielectric and optical properties of the heterostructures are strongly affected by the built-in electric field. Recently, Xia and Wei^[5] have studied hydrogenic impurity states in a wurtzite InGaN QD. It was found that the Al content in the $Al_xGa_{1-x}N$ potential hill layer has a close correlation with the discontinuity and polarization effect on the interface conduction band in a $GaN/Al_xGa_{1-x}N$ heterostructure.

The electron-phonon (e-p) interaction plays an important role in determining the physical properties of QDs consisting of polar materials^[6]. Therefore, there has been much interest in investigating the e-p interaction effects on the energies of electron states. A number of studies have focused on the influence of e-p interactions on the impurity properties in the single QD. Xie and Chen^[7] investigated the phonon contribution to the binding energy of the on-center and off-center impurities in a spherical QD. Szafran *et al.*^[8] studied the effect

of the electron-phonon coupling on the properties of a negatively charged donor center confined in a semiconductor spherical QD embedded in a glass matrix. The binding energy and dipole moment of a hydrogenic impurity confined in a OD with the shape of a spherical cap are calculated by Yuan et al.^[9]. Li and Chen^[10] have derived the longitudinal optical (LO), top surface optical (TSO) and side surface optical (SSO) modes in freestanding cylindrical QDs. Considering the electron and ion couple with the confined LO, TSO and SSO phonon modes, Charrour et al.^[11, 12], and Wang et al.^[13] calculated the binding energy of an on-center donor impurity in a cylindrical QD. Vartanian et al.^[14] presented a systematic study of the binding energy for the ground-state of a hydrogenic impurity in a cylindrical QD in the presence of both an applied electric field and the coupling between an electron and LO phonons. Recently, Zhong and Liu^[15] have derived the general expressions of the interface optical (IO) phonon modes, the dispersion relation, and an electron-IO phonon Fröhlich interaction Hamiltonian in the cylindrical QD heterostructure within the framework of a dielectric continuum approximation. Unfortunately, they neglected the coupling between the impurity and the phonons.

In this paper, considering the uniaxial anisotropy, the built-in electric and the interactions between the impurity and phonon modes (LO-like phonon and TO-like phonon modes), we calculated the e-p interaction effect on an on-center hydrogenic impurity state in the strained GaN/Al_xGa_{1-x}N cylindrical QD. With a variational technique, the binding energy and polaronic shift were performed as functions of the QD sizes, the anisotropy angle and the Al content. An effective Hamiltonian for the ground state of a bound polaron in the strained GaN/Al_xGa_{1-x}N cylindrical QD with an infinite potential is obtained by using a Lee–Low–Pines(LLP)-like method^[16, 17], and the variational calculations for the impurity state binding energy and its polaronic shift are performed. The numerical results and some detailed discussions are presented.

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2. Theory

Let us consider an isolated WZ cylindrical GaN/ Al_xGa_{1-x}N QD with radius *R* and height *L*, ignoring the strain effect of the Al_xGa_{1-x}N potential barrier layer. The hydrogenic donor impurity is located at the center of the QD. For WZ nitride semiconductor materials, we use the London model^[18], taking the *c*-axis along the *z* direction and denoting its perpendicular direction as \perp . $\kappa_{\perp}^{\infty}(\kappa_{z}^{\infty})$ is the high-frequency dielectric constant perpendicular to (along) the *c*-axis. κ_{\perp}^{0} and κ_{z}^{0} are the static dielectric constants. $m_{\perp}(m_{z})$ is the effective mass perpendicular to (along) the *c*-axis.

The Hamiltonian of the electron–impurity system, including the extraordinary LO-like and TO-like phonons and both the e–p and impurity–phonon (i–p) coupling, can be written as

$$\hat{H} = \hat{H}_{\rm e} + \hat{H}_{\rm ph} + \hat{H}_{\rm int}.$$
 (1)

The first term in Eq. (1) is the Hamiltonian for a bare shallow donor and is written as

$$\hat{H}_{\rm e} = \frac{\hat{p}_{\perp}^2}{2m_{\perp}} + \frac{\hat{p}_z^2}{2m_z} - \frac{e^2}{4\pi\varepsilon_0\varepsilon^\infty\sqrt{\rho^2 + z^2}} - eFz + V(\rho, z).$$
(2)

The confinement potential of a QD $V(\rho, z)$ can be described as

$$V(\rho, z) = \begin{cases} 0, & \rho < R, & |z| < L/2, \\ \infty, & \rho \ge R, & |z| \ge L/2, \end{cases}$$
(3)

where *R* and *L* are the radius and length of the QDs, respectively. κ^{∞} is the effective high-frequency dielectric constant and takes the form^[19]

$$\frac{1}{\kappa^{\infty}} = \frac{\cos^2\beta}{\kappa_{\perp}^{\infty}} + \frac{\sin^2\beta}{\kappa_{z}^{\infty}},\tag{4}$$

where β is the angle between the total momentum p and the *c*-axis.

The strength of the built-in electric field *F* caused by spontaneous and piezoelectric polarization in the WZ $GaN/Al_xGa_{1-x}N$ strained quantum dot is expressed as^[20]

$$F = \begin{cases} \left| -\frac{P_{\rm SP}^{\rm GaN} + P_{\rm PE}^{\rm GaN} - P_{\rm SP}^{\rm Al_x Ga_{1-x}N}}{\kappa_{\rm e}^{\rm GaN} \kappa_0} \right|, \quad |z| < L/2, \\ 0, \quad |z| \ge L/2, \end{cases}$$
(5)

$$P_{\rm PE}^{\rm GaN} = e_{31} \left(\varepsilon_{xx} + \varepsilon_{yy} \right) + e_{33} \varepsilon_{zz}, \tag{6}$$

where P_{SP}^{GaN} , P_{PE}^{GaN} and $P_{SP}^{Al_XGa_1-x^N}$ are the spontaneous and piezoelectric polarization of GaN and the spontaneous polarization of $Al_xGa_{1-x}N$, respectively. κ_e^{GaN} is the electronic dielectric constant of material GaN. e_{31} and e_{33} are the piezoelectric coefficient of GaN material, ε_{xx} , ε_{yy} and ε_{zz} are strains in the GaN layer in the *x*, *y* and *z* directions, respectively. In general, the direction of *F* depends on the orientation of the spontaneous and piezoelectric polarizations and can be determined by both the polarity of the crystal and the strain of the quantum well (QW) structure^[20]. Based on Eq. (5), we have calculated the strength of the built-in electric field F in the strained GaN layer as a function of the Al content x for the GaN/Al_xGa_{1-x}N quantum dot. The result shows that the built-in electric field in the strained GaN layer is extremely strong with an order of MV/cm and becomes higher with increasing Al content.

The second term in Eq. (1) is the phonon Hamiltonian and can be written as

$$\hat{H}_{\rm ph} = \sum_{j\boldsymbol{q}} \hbar \omega_j a_{j\boldsymbol{q}}^+ a_{j\boldsymbol{q}}, \qquad (7)$$

where j (= L, T) is the mode-index of the phonon characteristic frequencies. j = L stands for the LO-like phonon, and j = T for the TO-like phonon. a_{jq}^+ and a_{jq} are respectively the creation operator and annihilation operator. The characteristic phonon frequencies of WZ structures satisfy the following conditions^[18],

$$\omega_{\rm L}^2 = \omega_{z\rm L}^2 \cos^2 \theta + \omega_{\perp\rm L}^2 \sin^2 \theta, \qquad (8)$$

$$\omega_{\rm T}^2 = \omega_{z\rm T}^2 \sin^2 \theta + \omega_{\rm \perp T}^2 \cos^2 \theta, \qquad (9)$$

where θ is the angle between the phonon wave-vector \boldsymbol{q} and the *c*-axis.

The e-p and i-p interactions Hamiltonian in Eq. (1) is given by

$$\hat{H}_{\text{int}} = \sum_{j\boldsymbol{q}} \left[G_{j\boldsymbol{q}} (e^{i\boldsymbol{q}_{\perp} \cdot \boldsymbol{\rho} + q_{z}z} - 1) a_{j\boldsymbol{q}} + h.c. \right], \quad (10)$$

where "-1" term comes from the impurity localized at the origin^[16], and

$$G_{j\boldsymbol{q}} = \left(\frac{e^2\hbar}{\kappa_0 V}\right)^{1/2} \frac{1}{q} \left(\frac{\partial\kappa_{\theta}^j}{\partial\omega_j}\right)^{-1/2}, \qquad (11)$$

$$\frac{\partial \kappa_{\theta}^{J}}{\partial \omega_{j}} = \frac{\partial \kappa_{\perp}(\omega_{j})}{\partial \omega_{j}} \sin^{2} \theta + \frac{\partial \kappa_{z}(\omega_{j})}{\partial \omega_{j}} \cos^{2} \theta.$$
(12)

We first carry out a unitary transformation as follows,

$$U_0 = \exp\left[\sum_{j\boldsymbol{q}} \left(G_{j\boldsymbol{q}}^* a_{j\boldsymbol{q}}^+ - G_{j\boldsymbol{q}} a_{j\boldsymbol{q}}\right)/\hbar\omega_j\right].$$
(13)

Then, the Hamiltonian of the electron-impurity system (see the Eq. (1)) can be transformed into the following form,

$$\hat{H}^{*} = U_{0}^{-1}\hat{H}U_{0}$$

$$= \frac{\hat{p}_{\perp}^{2}}{2m_{\perp}} + \frac{\hat{p}_{z}^{2}}{2m_{z}} - \frac{e^{2}}{\kappa^{*}\sqrt{\rho^{2} + z^{2}}} - eFz + V(\rho, z)$$

$$+ \sum_{jq} \hbar\omega_{j}a_{jq}^{+}a_{jq} + \sum_{jq} \left(G_{jq}e^{iq\cdot\rho + q_{z}z}a_{jq} + h.c.\right),$$
(14)

where

$$\frac{1}{\kappa^*} = \frac{1}{4\pi\kappa_0\kappa^\infty} - \frac{1}{2\pi^2\kappa_0} \times \sum_j \int \left(\omega_j \frac{\partial\kappa_\theta^j}{\partial\omega_j}\right)^{-1} r\sin\theta\cos\left(qr\cos\theta\right) d\theta dq.$$
(15)

In the above equation, r is the electron distance from the center. For convenience, we have dropped the infinite selfenergy of a static point charge $-\sum_{j\mathbf{q}} |G_{j\mathbf{q}}|^2 / \hbar \omega_j$. Now we extend the LLP method, making two unitary

transformations,

$$U_1 = \exp[-i\sum_{j\mathbf{q}} a_{j\mathbf{q}}^+ a_{j\mathbf{q}} \left(\mathbf{q}_\perp \cdot \boldsymbol{\rho} + q_z z\right)], \qquad (16)$$

$$U_{2} = \exp\{\sum_{j q} [a_{j q}^{+} f_{j}(q) - a_{j q} f_{j}^{*}(q)]\}, \qquad (17)$$

where $f_j(q)$ and $f_j^*(q)$ are the variational functions, which can be determined by minimizing the expectation value of the bound polaron energy. As a first approximation, we confined our discussion in the low-temperature limit, i.e., the phonon vacuum state $|0\rangle(\langle 0|0\rangle = 1)$. After the two transformations, the effective Hamiltonian for the bound polaron in the phonon vacuum state can be derived as

$$H_{\text{eff}} = \langle 0 | U_2^{-1} U_1^{-1} \hat{H}^* U_1 U_2 | 0 \rangle$$

$$= \frac{p_\perp^2}{2m_\perp} + \frac{p_z^2}{2m_z} - eFz + V(\rho, z) - \frac{e^2}{\kappa^* \sqrt{\rho^2 + z^2}}$$

$$+ \sum_{jq} \left[G_{jq} f_j(q) + G_{jq}^* f_j^*(q) \right]$$

$$+ \frac{\hbar^2}{2m_\perp} \left[\sum_{jq} |f_j(q)|^2 q_\perp \right]^2$$

$$+ \frac{\hbar^2}{2m_z} \left[\sum_{jq} |f_j(q)|^2 q_z \right]^2$$

$$+ \sum_{jq} |f_j(q)|^2 \left(\hbar \omega_j - \frac{\hbar}{m_\perp} q_\perp \cdot p_\perp - \frac{\hbar}{m_z} q_z p_z + \frac{\hbar^2 q_\perp^2}{2m_\perp} + \frac{\hbar^2 q_z^2}{2m_z} \right).$$
(18)

After some tedious algebra, we finally obtain the effective Hamiltonian of the bound phonon in cylindrical QD,

$$H_{\rm eff} = \frac{p^2}{2m^*} - eFz + V(\rho, z) - \frac{e^2}{\kappa^* \sqrt{\rho^2 + z^2}} - \sum_{jq} \frac{|G_{jq}|^2}{\hbar\omega_j + \frac{\hbar^2 q^2}{2m}},$$
(19)

where m^* is the effective mass of a polaron and can be given by

$$m^* = m'(1 + \Delta m).$$
 (20)

Here.

$$\frac{1}{m'} = \frac{\sin^2 \beta}{m_\perp} + \frac{\cos^2 \beta}{m_z},\tag{21}$$

$$\Delta m = \frac{A^2 + B}{1 + 2A - B},$$
 (22)

$$A = \sum_{j\mathbf{q}} \frac{2 \left| G_{j\mathbf{q}} \right|^2 \hbar^2 \mathbf{q}^2 D \cos(\beta - \theta)}{\left(\hbar \omega_j + \frac{\hbar^2 \mathbf{q}^2}{2m} \right)^3}, \qquad (23)$$

$$B = \sum_{j\boldsymbol{q}} \frac{2 \left| G_{j\boldsymbol{q}} \right|^2 \hbar^2 \boldsymbol{q}^2 D^2 m'}{\left(\hbar \omega_j + \frac{\hbar^2 \boldsymbol{q}^2}{2m} \right)^3},$$
(24)

$$\frac{1}{m} = \frac{\sin^2 \theta}{m_\perp} + \frac{\cos^2 \theta}{m_z},\tag{25}$$

$$D = \frac{\sin\theta\sin\beta}{m_{\perp}} + \frac{\cos\theta\cos\beta}{m_{z}}.$$
 (26)

In the above equation, $p = (p_{\perp}, p_z)$ stands for the total momentum and $q = (q_{\perp}, q_z)$ is the phonon wave-vector. For convenience, but without loss of generality, we assume that p, q and c-axis are in the same plane.

The total trial wave function for the system is a product of the wave function $\phi(\rho, z)$ for the electron and the phonon vacuum state $|0\rangle$,

 $|\psi\rangle = \phi(\rho, z) |0\rangle,$

(27)

with

$$\phi(\rho, z) = J_0(\sqrt{\mu}\rho) \cos\left(\frac{\pi}{L}z\right) e^{-\tau \frac{z}{L}} e^{-\alpha \rho_{\rm ei}^2} e^{-\gamma z_{\rm ei}^2}, \quad (28)$$

where α , τ and γ are the variational parameters. Then, the ground state energy of the bound polaron can be obtained by a variational procedure,

$$E_{\rm g} = \min_{\alpha, \gamma, \tau} \frac{\langle \phi | H_{\rm eff} | \phi \rangle}{\langle \phi | \phi \rangle}.$$
 (29)

The binding energy of the impurity state (bound polaron) $E_{\rm b}$ is defined as the difference between the ground state energy of a free polaron and that of a bound polaron,

$$E_{\rm b} = E_{\rm free} - E_{\rm g}.$$
 (30)

The polaronic shift for the binding energy can be defined as the difference between $E_{\rm b}$ with phonons and that without phonons,

$$\Delta E_{\rm ph} = E_{\rm b}$$
(with phonons) – $E_{\rm b}$ (without phonons). (31)

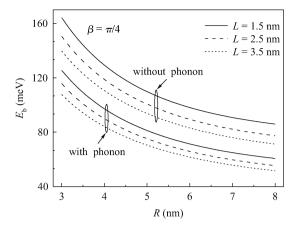


Fig. 1. Binding energies of impurity states with and without impurity state-phonon interaction in GaN/Al_{0.15}Ga_{0.85}N QDs as functions of the QD radius for the different QD heights.

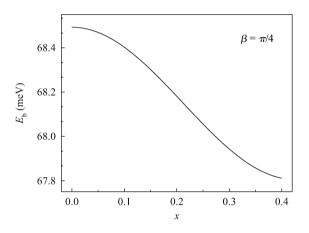


Fig. 2. Binding energy as a function of the Al content for the QD radius R = 5 nm and height L = 4 nm, taking impurity-phonon interation into account

3. Numerical results and discussions

We have calculated numerically the polaronic binding energy and its polaronic shift in a GaN cylindrical QD. In the calculations, we use the following parameter values^[21], $\omega_{zT} = 66.06 \text{ meV}$, $\omega_{zL} = 91.10 \text{ meV}$, $\omega_{\perp T} = 69.53 \text{ meV}$, $\omega_{\perp L} = 91.97 \text{ meV}$, $\kappa_z^0 = 10.18$, $\kappa_{\perp}^0 = 9.36$, $\kappa_z^\infty = \kappa_{\perp}^\infty = 5.35$, $m_{\perp} = m_z = 0.2 m_e$, and the results are illustrated and discussed as follows.

In Fig. 1, the binding energies of impurity states with and without phonons as a function of QD radius for different values of the QD height (L = 1.5, 2.5 and 3.5 nm) with the anisotropic angle $\beta = \pi/4$ are presented. As expected, the binding energies with and without phonons diminish with an increase in the QD radius. This is because the Coulomb interaction between the electron and impurity is reduced due to increasing their relative distance in the ρ -direction if the QD radius *R* increases. In addition, the binding energy decreases with increasing QD height for the same QD radius. One also notices from Fig. 1 that the binding energy with e-p and i-p interactions is obviously lower than that without the interactions. This result indicates that the Coulomb interaction is reduced due to the e-p and i-p interactions.

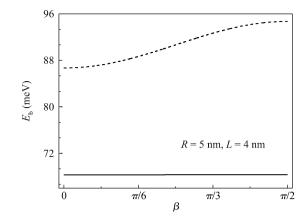


Fig. 3. Binding energy with and without impurity state-phonon interaction as a function of the anisotropy angle for the GaN/Al_{0.15}Ga_{0.85}N QD radius R = 5 nm and height L = 4 nm.

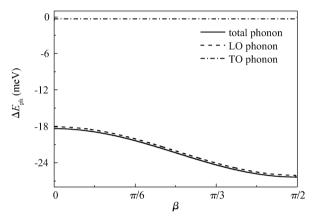


Fig. 4. Polaronic shifts of binding energy as a function of the anisotropy angle for the GaN/ Al_{0.15}Ga_{0.85}N QD radius R = 5 nm and height L = 4 nm.

To clearly understand the effects from Al content x on the binding energy, the polaronic binding energy as a function of Al content x for the QD height L = 4 nm, radius R = 5 nm and the anisotropic angle $\beta = \pi/4$ are plotted in Fig. 2. It can be seen that the binding energy decreases slightly with the Al content x increasing from 0.0 to 0.4. The explanation for such a behavior is as follows. The increase in the Al content induces an increase in the built-in electric field, but the depolarization field induced by the i-p interaction counteracts the effect of the built-in electric field. Thus the effect of the Al component on the binding energy is negligible.

In Fig. 3, we display the polaronic binding energies with (solid curve) and without phonons (dashed curve) as a function of the anisotropic angle β for the QD height L = 4 nm, radius R = 5 nm. This indicates that the binding energies with e-p and i-p interactions are obviously lower than those without the interactions for the same anisotropic angle β . It also shows that the binding energy increases with increasing anisotropic angle β when phonons are out of consideration. It is explained that the motion of the electron tends to the x-y plane and the electron is closer to the impurity with the anisotropic angle β increasing, therefore the Coulomb interaction becomes stronger

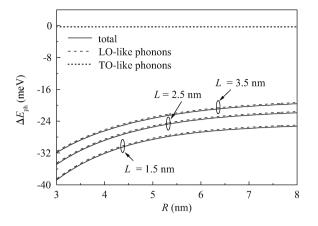


Fig. 5. Polaronic shifts of binding energies as functions of the QD radius R for the GaN/Al_{0.15}Ga_{0.85}N QD heights L.

and the binding energy increases. However, the change in the binding energy is inconspicuous with the variety of anisotropy angle when phonons are considered. This is mainly because the Coulomb potential between the electron and the impurity is screened by the i-p interaction.

To understand the effects from LO-like and TO-like phonons on the binding energy, Figure 4 shows the polaronic shifts for the binding energies as a function of the anisotropic angle β for the QD height L = 4 nm and radius R = 5 nm. It is clear that the total polaronic shift and the shift from the LOlike phonon are always monotonically increased with increasing anisotropic angle β . It is also found that the LO-like phonon influence on the polaronic shift is dominant, and the contribution from TO-like phonons is guite small and is a relatively unimportant role. We have also calculated polaronic shifts for binding energies as a function of the QD radius for different values of the QD height (L = 1.5, 2.5 and 3.5 nm) with the anisotropic angle $\beta = \pi/4$ in Fig. 5. It can be seen that the polaronic shift for the binding energy decreases with increasing QD radius and height. This indicates that the influences of the phonons become stronger as the QD confinement increases. It can also be observed that the LO-like phonon plays a dominant role in the polaronic shift, and the polaronic shift is hardly changed with respect to the QD radius because the contribution from the TO-like phonon is small. This is in good agreement with the result of Mora-Ramos et al.^[22].

4. Conclusions

In conclusion, an electron bound to a hydrogenic donor impurity is discussed by considering the influences of both branches of LO-like and TO-like phonons with a variational method in a GaN/Al_{0.15}Ga_{0.85}N cylindrical QD. In our calculations, the interaction between an impurity and phonons has also been considered to obtain the binding energy of a bound polaron. Numerical results show that the binding energy is generally reduced by the e–p and i–p interactions, and the contribution of the LO-like phonon to the binding energy is dominant. The binding energy of the bound polaron decreases with increasing QD radius. At the same time, the binding energy decreases slightly with increasing Al content, and it is not sensitive to the variety of anisotropy angle.

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