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Charged excitons and trions in 2D parabolic quantum dots

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This work is dedicated to the bright memory of our mentor, Prof. Nguyen Van Hieu, who passed away recently during the revision of this paper, with our respect and gratitude of being his students 40 years ago

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ABSTRACT

Currently in literature, "charged exciton" and "trion" are often confused with each other and mostly considered the same. In this work we show that these terms might actually describe different quasi-particles. Energies and binding energies of both charged excitons and trions in 2D parabolic quantum dots have been calculated as functions of confinements of charge carriers in a quantum dot, using the unrestricted Hartree–Fock method. It is shown that the binding energies of the charged exciton and the trion behave differently in regard to the ratios of the confinements of the electron and the hole. The effect of the external magnetic field on the binding energies of charged excitons has also been considered.

1. Introduction

Multi-particle bound states play important role in electronics and optical properties of semiconductors and have been studied intensively. Over the past two decades, multi-particles such as excitons, trions and their associates in low-dimensional semiconductor systems have attracted a lot of attention due to their unique properties as well as their critical role in electronics, optoelectric devices and quantum information technology in the future [1–3].

As its name implies, trion is a bound state of three charged particles, which can be two electrons and a hole or two holes and an electron, or, understandably, an exciton with an electron or an exciton with a hole. In the case of excitons coupling with electrons (holes), they are also known as negatively (positively) charged excitons [4–7]. Like excitons and other exciton-based systems, in trion states the charge carriers are correlated with each other through Coulomb interaction, which has attracted great attention recently, especially in atomic-size thin semiconductor layers based on transition metal dichalcogenide monolayers [8–11]. Up to very recent, there is no difference noted between trions and charged excitons. Since the nature of Coulomb interaction between the particles in trions and charged excitons is different, it is worth to study them and their existences more carefully.

Recently several new quasi-particles have been named. "Duo", "trio", "quatuor", for quasi-particle consisting of two, three and four charge carriers, respectively, have been introduced in quantum dots by Combescot [12], and "quadron" for four charged carriers in 2D semiconductor quantum dots by us [13–15] to distinguish from conventional excitons, charged excitons and biexcitons. In [12–15] the nature of the differences between biexciton and quantum dot "quatuor" or "quadron" has been shown. For instance, while both biexciton and quadron consist of four interacting carriers (two electrons and two holes), the traditional biexciton is considered as a bound state of two interacting excitons, while the newly invented quadron is a bound state of two electrons and two holes with a fair multi-particle Coulomb interaction between each pair of particles. It has been shown in our previous works [13–15] that in a small 2D InAs quantum dot it prefers a bound quadron rather than a conventional biexciton.

To develop this problem further, we study the differences between charged excitons and trions in self-assembled semiconductor quantum dots with parabolic confinement potential. Due to their large binding energies and the easiness of incorporation in field effect structures, which allows the study of external magnetic field effect without breaking the symmetry of the system [13–22], self-assembled semiconductor quantum dots with parabolic confinement potential currently are of high interest. Other than charged exciton as a bound state of an exciton with an electron or a hole, in this work trion is considered as a bound

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state of three particles (two electron and a hole or two hole and an electron) with a fair multi-particle Coulomb interaction between them. The binding energies of the charged excitons and trions have been calculated for a full range of electron-to-hole confinement ratio, using the Hartree–Fock method. The binding energies of the charged exciton and the trion will be shown to behave differently in regard to the ratios of the confinements between the electron and hole. The effect of a magnetic field on binding energy of the charged excitons will be presented as well. Based on our results, the trions and charged excitons are shown to be different quasi-particles with different binding energies, different behaviors with respect to electron–hole confinement and magnetic files, and different preferences.

2. Model of interacting confined charged particles

We consider the model of interacting electrons and holes confined in a 2D quantum dot with parabolic lateral potential in the presence of the perpendicular magnetic field $\vec{B} \parallel z$ [13–15]. The Hamiltonian of the system of N electrons and M holes ($N = 2, M = 1$ or $N = 1, M = 2$ for charged excitons and trions) can be written in the framework of the effective-mass approximation as follows

$$\hat{H} = \sum_{i=1}^{N} h(\vec{r}_i) + \sum_{k=1}^{M} h'(\vec{r}_k) + \sum_{i=1;i<j}^{N} \frac{e^2}{\epsilon r_{ij}} + \sum_{k=1;k<l}^{M} \frac{e^2}{\epsilon r_{kl}} - \sum_{i=1}^{N}\sum_{k=1}^{M} \frac{e^2}{\epsilon r_{ik}} , \quad (1)$$

where the first two terms are Hamiltonians of single electrons and single holes, the third and fourth terms are electron–electron and hole–hole Coulomb interactions, respectively, and the last term is the electron–hole Coulomb interaction. ϵ is the dielectric constant of the material. In a quantum dot with parabolic confinement by magnetic field the Hamiltonians for a single electron and a single hole can be written as the following:

$$h(\vec{r}_i) = -\frac{\nabla_i^2}{2m_e^*} + \frac{m_e^*}{2}(\omega_e^2 + \frac{1}{4}\omega_{ce}^2) r_i^2 + \frac{1}{2}\omega_{ce}\hat{L}_{zi} , \quad (2)$$

$$h'(\vec{r}_k) = -\frac{\nabla_k^2}{2m_h^*} + \frac{m_h^*}{2}(\omega_h^2 + \frac{1}{4}\omega_{ch}^2) r_k^2 + \frac{1}{2}\omega_{ch}\hat{L}_{zk} , \quad (3)$$

where m_e^* (m_h^*) and ω_e (ω_h) are the effective mass and the confinement potential of the electron (hole), respectively; $\omega_{ce} = eB/m_e^*$ ($\omega_{ch} = eB/m_h^*$) and $\hat{L}_{zi}$ ($\hat{L}_{zk}$) are the cyclotron frequency for the electron (hole) and the z-components of electron (hole) orbital angular momentum operators, respectively. The Zeeman splitting terms for the interaction of the spins with the magnetic field have been omitted here due to their smallness in comparison with other terms.

It should be noted that in the framework of the effective mass approximation, the effective mass of electron or hole in semiconductor quantum dots may be considered depending on the dot's radius (for example, see [23,24] and the references therein). However, in this work for the model of two-dimensional parabolic quantum dot, similarly to [13–22], we take the bulk values for the masses of the electron and the hole in the Eqs. (2) and (3) as a good approximation.

In polar coordinates the eigenfunctions of the single electron in the quantum states (n, m) could be written as the following:

$$\chi^e_{n,m}(r,\varphi) = \frac{1}{\sqrt{2\pi}} e^{im\varphi} \sqrt{\frac{2n!}{(n+|m|)!}} \alpha_e (\alpha_e r)^{|m|} e^{-(\alpha_e r)^2/2} L_n^{|m|}((\alpha_e r)^2) , \quad (4)$$

with $L_n^{|m|}$ being generalized Laguerre polynomial. The eigenvalue of the electron then has the form:

$$E^e_{n,m} = \Omega_e(2n + |m| + 1) + \frac{1}{2} m\omega_{c_e} , \quad (5)$$

where $\Omega_e = (\omega_e^2 + \frac{1}{4}\omega_{c_e}^2)^{1/2} \quad \alpha_e = \sqrt{m_e^* \Omega_e}$.

The wave function of the system is written in the form of direct product of the Slater determinants for N electrons and M holes:

$$\Psi(\xi_1, \ldots, \xi_N, \xi'_1, \ldots, \xi'_M) = |\psi_1(\xi_1)\ldots\psi_N(\xi_N)|.|\psi'_1(\xi'_1)\ldots\psi'_M(\xi'_M)| , \quad (6)$$

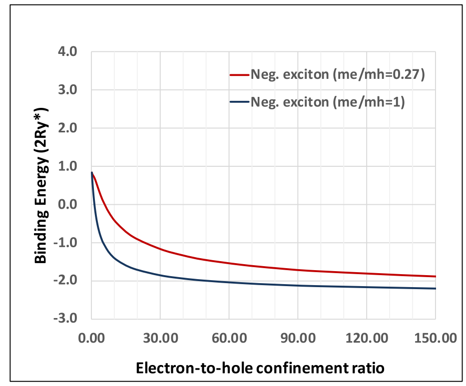

Fig. 1. (Color online) Comparison of negatively charged exciton binding energies with $m_e/m_h = 0.27$ and $m_e/m_h = 1$ as functions of electron-to-hole confinement ratio.

where the electron and hole wave functions $\psi_i(\xi), \psi'_k(\xi)$ in the Slater determinants are spin dependent: $\psi_i(\xi) = \phi_i^\alpha(\vec{r})\sigma(\alpha)$ or $\phi_i^\beta(\vec{r})\sigma(\beta)$ for up- or down-spin electrons, and similar for holes.

In the Hartree–Fock–Roothaan formulation [13–15,22], the spatial parts of electron and hole wave functions $\phi_i^{\alpha,\beta}(\vec{r})$ and $\phi_k'^{\,\alpha,\beta}(\vec{r})$ are written as expansions with the basis functions being the single electron and single hole wavefunctions in (4):

$$\phi_i^{\alpha,\beta}(\vec{r}) = \sum_\nu C_{i\nu}^{\alpha,\beta} \chi_\nu^e(\vec{r}) , \quad \phi_k'^{\,\alpha,\beta}(\vec{r}) = \sum_\mu C_{k\mu}'^{\,\alpha,\beta} \chi_\mu^h(\vec{r}) ,$$

indexes ν, μ run over all single electron or hole states with quantum numbers (n, m).

Within Hartree–Fock–Roothaan formulation the matrix elements of Coulomb interactions with these basis functions can be calculated analytically [25]. The system of Hartree–Fock–Roothaan equations can be solved self-consistently to give the total energy of the system:

$$E = \frac{1}{2}\sum_{\mu,\nu}\left\{\delta_{\mu\nu}P_{\mu\nu}^T[\Omega_e(2n+|m|+1) + m\omega_{c_e}] + P_{\mu\nu}^\alpha F_{\nu\mu}^\alpha + P_{\mu\nu}^\beta F_{\nu\mu}^\beta\right\}$$
$$+ \frac{1}{2}\sum_{\mu,\nu}\left\{\delta_{\mu\nu}P_{\mu\nu}'^{\,T}[\Omega_h(2n+|m|+1) - m\omega_{c_h}] + P_{\mu\nu}'^{\,\alpha} F_{\nu\mu}'^{\,\alpha} + P_{\mu\nu}'^{\,\beta} F_{\nu\mu}'^{\,\beta}\right\}, \quad (7)$$

where the explicit expressions for $P^T_{\mu\nu}$, $P'^{\,T}_{\mu\nu}$, $P^\alpha_{\mu\nu}$, $P^\beta_{\mu\nu}$, $P'^{\,\alpha}_{\mu\nu}$, $P'^{\,\beta}_{\mu\nu}$ and $F^{\alpha,\beta}_{\mu\nu}$, $F'^{\,\alpha,\beta}_{\mu\nu}$ can be found in [13].

3. Results and discussions

For numerical calculation, the following parameters for self-assembled InAs/GaAs quantum dots [13–15,22] have been used: $m_e^* = 0.067m_o$, $m_h^* = 0.25m_o$, $\omega_e = 49$ meV, $\omega_h = 25$ meV, $\epsilon_s = 12.53$. The adopted length and energy units are $a_B^* = \epsilon_s/m_e^* e^2 = 9.9$ nm, $2Ry^* = m_e^* e^4/2\epsilon_s^2 = 11.61$ meV. The oscillator lengths for electrons (holes) in the absence of magnetic fields $l_{e,h} = (m^*_{e,h}\omega_{e,h})^{-1/2}$ are 4.8 nm (3.5 nm). With the effective excitonic Bohr radius of about 13 nm, these parameters mean strong confinement of the particles in small InAs/GaAs quantum dots.

To study of the effect of confinement on the binding energies, for comparison we use two parameter sets, set 1 with $\omega_e = 49$ meV, $m_e^* = 0.067m_o$, and $m_h^* = 0.25m_o$, and set 2 with $\omega_e = 49$ meV, $m_e^* = 0.067m_o$, and $m_h^* = 0.067m_o$. Denoting $m_e = m_e^*/m_o$, $m_h = m_h^*/m_o$, we have for set 1: $m_e/m_h = 0.27$, and for set 2: $m_e/m_h = 1$.

In Figs. 1–4 the binding energies of the negatively charged exciton and the negative trion in the ground state without magnetic fields as functions of electron-to-hole confinement ratio have been compared. To see the impact of the carrier masses, we compare the results for both parameter sets with $m_e/m_h = 0.27$, and $m_e/m_h = 1$, respectively. For the whole range of the electron-to-hole confinement ratio ω_e/ω_h, the binding energy of the negative trion is always larger than that of the

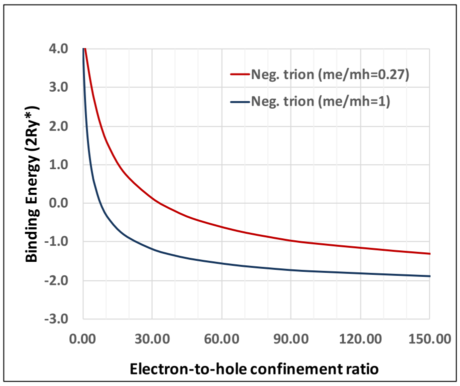

Fig. 2. (Color online) Comparison of negative trion binding energies with the $m_e/m_h = 0.27$ and $m_e/m_h = 1$ as functions of electron-to-hole confinement ratio.

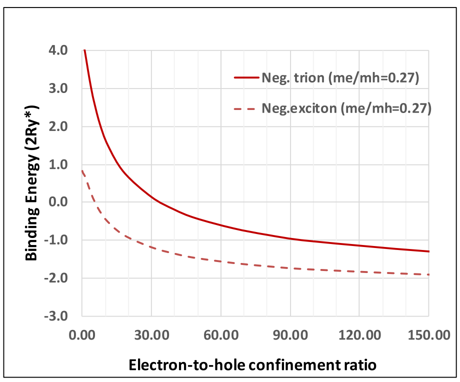

Fig. 3. (Color online) Comparison of binding energies for negative trions and negatively charged excitons with the $m_e/m_h = 0.27$, as functions of electron-to-hole confinement ratio.

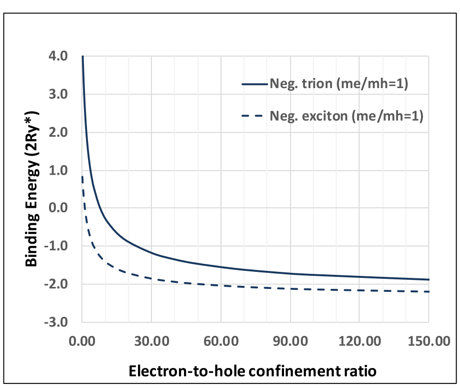

Fig. 4. (Color online) Comparison of binding energies for negative trions and negatively charged excitons with the $m_e/m_h = 1$, as functions of electron-to-hole confinement ratio.

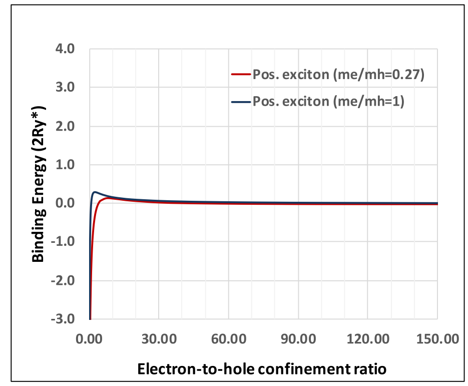

Fig. 5. (Color online) Comparison of positively charged exciton binding energies with the $m_e/m_h = 0.27$ and $m_e/m_h = 1$, as functions of electron-to-hole confinement ratio.

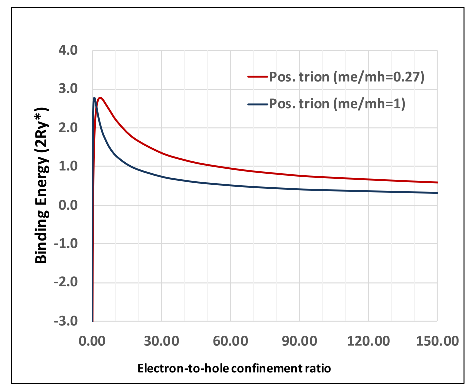

Fig. 6. (Color online) Comparison of positive trion binding energies with the $m_e/m_h = 0.27$ and $m_e/m_h = 1$, as functions of electron-to-hole confinement ratio.

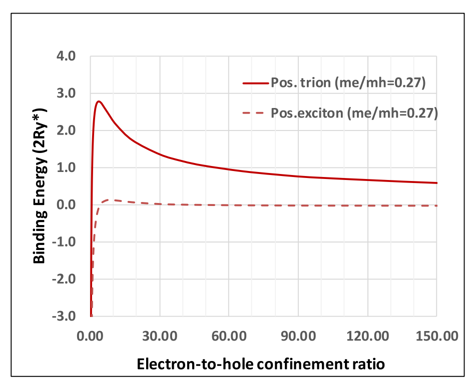

Fig. 7. (Color online) Comparison of binding energies for positive trions and positively charged excitons with the $m_e/m_h = 0.27$, as functions of electron-to-hole confinement ratio.

negatively charged exciton. From the graphs one can see that at small values of the electron-to-hole confinement ratio both negative trions and negatively charged excitons are bound systems but they become unbound when the electron-to-hole confinement ratio increases.

For positively charged excitons and positive trions we get the similar differences. In Figs. 5–8 the binding energies of the positively charged exciton and positive trion in the ground states as functions of electron-to-hole confinement ratio are compared with two parameter sets with $m_e/m_h = 0.27$ and $m_e/m_h = 1$. Again, the binding energy of the positive trion is always larger than that of the positively charged excitons.

However in this case, opposite to the above case of negative trions and negatively charged excitons, we observe the transition from anti-binding to binding state of both the positive trion and the positively charged exciton when increasing the electron-to-hole confinement ratio. This happens due to the contributions of the hole–hole interaction, which is larger than that of electron–electron interaction, to suppress the electron–hole interaction. It is also interesting to note that our new results can help to understand and clarify the sensitive changes in the binding energy of trions in natural ensembles of InAs/GaAs quantum dots with randomly fluctuating parameters [21].

On Figs. 9–10 the effect of magnetic fields on charged exciton states has been shown. It is shown that magnetic fields increase the binding

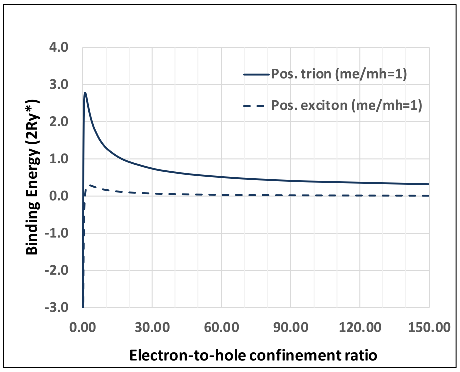

Fig. 8. (Color online) Comparison of binding energies for positive trions and positively charged excitons with the $m_e/m_h = 1$, as functions of electron-to-hole confinement ratio.

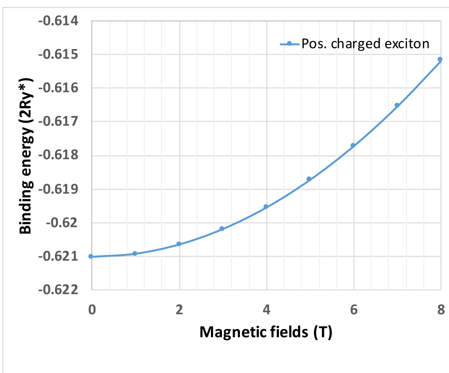

Fig. 9. (Color online) Change of the negatively charged exciton binding energy as a function of magnetic field.

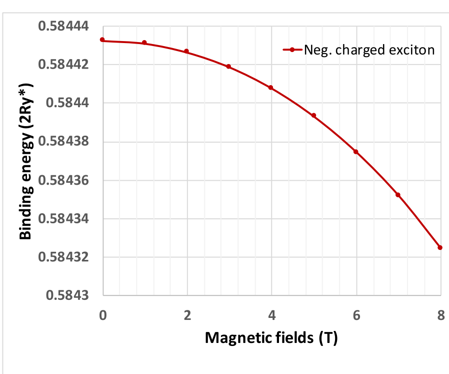

Fig. 10. (Color online) Change of the positively charged exciton binding energy as a function of magnetic field.

energy of the positively charged exciton but decrease the one of the negatively charged exciton, although the effect is rather small.

In [14,15] we have analyzed the agreements between our results using Hartree–Fock method and the experimental data for the biexciton binding energy [19]. In this work in InAs/GaAs self-assembled quantum dot we obtained a good agreement between our results and the experimental data for the negatively and positively charged exciton binding energies as well. Indeed, the value of electron-to-hole confinement ratio in InAs/GaAs self-assembled quantum dot is $\omega_e/\omega_h = 1.96$, corresponding to the values of the negatively and positively charged exciton binding energies $0.58\ (2Ry^*) \approx 6.73$ meV and $-0.62\ (2Ry^*) \approx -7.2$ meV, respectively. Our results agree very well with the experimental value of 6.2 ± 0.4 meV for negatively exciton and rather well with the experimental value interval of [-1 meV:-6 meV] for positively charged exciton, respectively [19].

4. Conclusion

In conclusion, in this work charged exciton and trion states in 2D parabolic quantum dots have been compared the first time.

The binding energies of trions and charged excitons have been studied using unrestricted Hartree–Fock method for a full range of electron-to-hole confinement ratios. It is shown that in the ground state of 2D InAs quantum dots, the binding energies of charged excitons and trions can be either negative or positive, depending on the correlation ratio of the electron–hole confinement potential. Namely, at small values of the electron-to-hole confinement ratio both negative trions and negatively charged excitons are bound systems but they become unbound when the electron-to-hole confinement ratio increases. However, the opposite happens for the positive trion and the positively charged exciton, there occurs a transition from anti-binding to binding state when the electron-to-hole confinement ratio increases. Magnetic fields increase the binding energy of positively charged excitons but decrease it for the case of negatively charged exciton, although the effect is rather small.

The results of this work show that charged excitons and trions are different quasi-particles and should not be considered the same. In all cases, the binding energies of negative or positive trion systems are generally larger than that of the corresponding negatively or positively charged excitons and are more preferable.

Our theoretical calculation results are in good agreements with the experimental data on the binding energies of negative and positive charged excitons in small self-organized InAs/GaAs quantum dots. Our results help to further clarify the properties of the elementary excitations and the differences between charged excitons and trions in semiconductor quantum dots.

CRediT authorship contribution statement

Nguyen Hong Quang: Conceptualization, Methodology, Software, Data curation, Visualization, Writing – original draft, Reviewing, Acquisition of the financial support. **Nguyen Que Huong:** Conceptualization, Methodology, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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References

[1] D. Bimberg, NanoScience and Technology: Semiconductor Nanostructures, Springer, Berlin, 2008.
[2] G. Wang G, et al., Rev. Modern Phys. 90 (2018) 021001.
[3] M. Peter (Ed.), Quantum Dots for Quantum Information Technologies, Springer, Berlin, 2018.
[4] M.A. Lampert, Phys. Rev. Lett. 1 (1958) 450.
[5] K. Kheng, et al., Phys. Rev. Lett. 71 (1993) 1752.
[6] J.I. Climente, A. Bertoni, G. Goldoni, Phys. Rev. B 78 (2008) 155316.
[7] B. Ganchev, et al., Phys. Rev. Lett. 114 (2015) 107401.
[8] K.F. Mak, et al., J. Nat. Mater. 112 (2013) 207.
[9] K.F. Ross, et al., J. Nat. Commun. 4 (2013) 1474.

[10] M.M. Glazov, J. Chem. Phys. 153 (2020) 034703.
[11] W. Lan, Appl. Phys. Lett. 117 (2020) 083107.
[12] M. Combescot, V. Voliotis, S.V. Shiau 2020, Semicond. Sci. Technol. 35 (2020) 045013.
[13] N.H. Quang, T.A. Dung, J. Phys. Conf. Ser. 1506 (2020) 012010.
[14] N.H. Quang, N.Q. Huong, T.A. Dung, H.A. Tuan, N.T. Thang, Physica B 602 (2021) 412591.
[15] N.H. Quang, T.A. Dung, J. Phys. Conf. Ser. 1932 (2021) 012011.
[16] S. Rodt, et al., Phys. Rev. B 68 (2003) 035331.
[17] G. Bester, A. Zunger, Phys. Rev. B 68 (2003) 073309.
[18] A. Schliwa, M. Winkelnkemper, D. Bimberg, Phys. Rev. B 79 (2009) 075443.
[19] M.F. Tsai, et al., Phys. Rev. Lett. 101 (2008) 267402.
[20] M. Abbarchi, et al., Phys. Rev. B 82 (2010) 201301.
[21] M. Zielinski, et al., Phys. Rev. B 91 (2015) 085303.
[22] N.H. Quang, S. Ohnuma, A. Natori, Phys. Rev. B 62 (2000) 12955.
[23] S.I. Pokutnyi, Semiconductors 41 (11) (2007) 1323.
[24] N.H. Quang, N.T. Truc, Y.-M. Niquet, Comput. Mater. Sci. 44 (2008) 21.
[25] V. Halonen, T. Chakraborty, P. Pietilainen, Phys. Rev. B 45 (1992) 5980.