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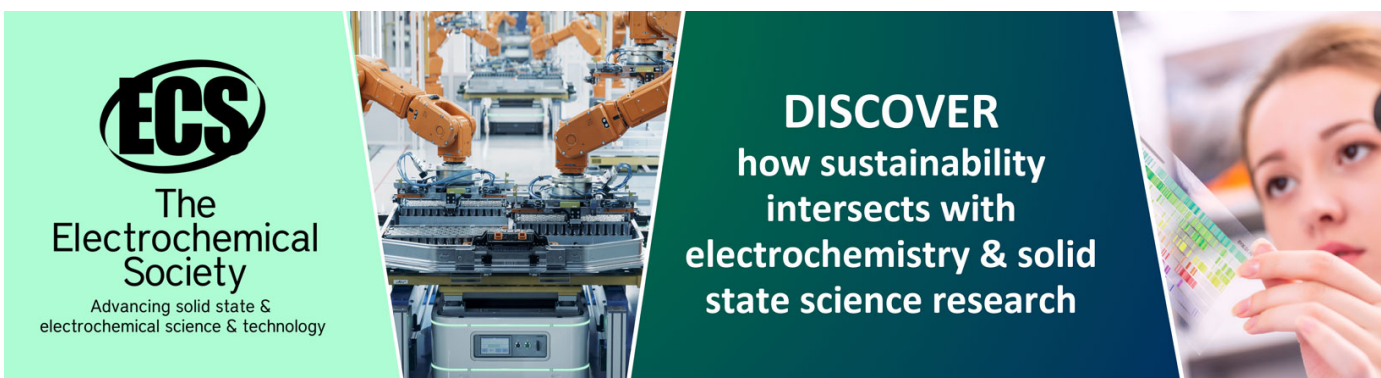
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Biexcitons and quadrons in self-assembled quantum dots

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Abstract

We theoretically study biexcitons and quadrons in quantum dots with parabolic confinement and give a complete comparison between the two excitations. The calculation of quadron and biexciton binding energies as functions of electron-to-hole confinement potentials and mass ratios, using the unrestricted Hartree–Fock method, shows the essential differences between biexcitons and quadrons. The crossover between the negative and positive binding energies is indicated. The effect of an external magnetic field on the quadron and biexciton binding energies has also been investigated. In addition, the crossover between anti-binding and binding of both excited quadron and biexciton states in a certain range of the electron-to-hole oscillator length ratios has been found.

Keywords: exciton, biexciton, quadron, parabolic quantum dot, Hartree–Fock method, binding energy

1. Introduction

Semiconductor optics has been a hot topic for decades. Most of the optics phenomena in semiconductors and their structures are determined by excitons and their associates. The optical processes happening right below the absorption band gap depend strongly on the electronic structures of these quasi-particles, both in bulks and in nanostructures. The application of excitons in nano-optoelectronic devices has been progressing intensively during the last few decades [1–6] due to the fast development of nanotechnology and the realization of many high-quality low-dimensional confinement systems such as quantum wells, quantum wires, microcavities, and quantum dots where the properties of the systems are determined by quantum phenomena. The 2023 Nobel Prize in Chemistry recognized the success in the investigation and production of quantum dots and the importance of confinement systems. One

of the direct consequences of quantum confinement is that elementary excitations in semiconductor nano-structures can reach large binding energies.

A self-assembled semiconductor quantum dot [7] with parabolic confinement potential plays an important role in the study of excitons and their complexities [8–20]. Self-assembled quantum dots are mainly formed through strain-induced mechanisms. The strain confinement in self-assembled quantum dots results in an energy structure of the charge carriers similar to that in a parabolic potential well. This kind of quantum dot has on one hand demonstrates large biexciton binding energies, and on the other hand has easily been incorporated into field-effect structures to study the effect of an external magnetic field without breaking the symmetry of the system.

The spontaneous nature of self-assembled quantum dot formation leads to variations in dot size, which can have both advantages and challenges in applications. The value of the confinement potential in self-assembled quantum dots is determined by the quantum dot size, influencing the electronic

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states within the dots. Randomness in dot size leads to variability in the confinement potentials for electrons and holes, resulting in changes to their energy structure in the quantum dots. Hence, it is essential to investigate and understand the alterations in the energy structure of excitonic systems caused by random changes of the system parameters.

While biexciton is a traditional excitation, we recently showed [8–10] that there exists another excitation, also consisting of two electrons and two holes, which is not necessarily the same as the biexciton. Furthermore, we found that under certain conditions in quantum dots this new excitation can be more favorable than the biexciton. We called this new excitation a ‘quadron’ [9], a quasi-particle consisting of these four carriers in quantum dots, to distinguish them from conventional biexcitons. It is interesting to note that a similar idea relating to quadrons has been presented almost at the same time in [12], where the authors show the fundamental differences between the usual biexciton and quantum dot ‘quatuor’ - their term for the system of four carriers—two electrons and two holes in quantum dots, instead of our term ‘quadron’.

A quadron [8–10] or a quatuor [12], in fact, is a system of two electrons and two holes which interact with one another equally. The picture of a conventional biexciton—the coupling of two excitons—is replaced by a totally different picture of pair interactions among the four carriers. In our previous works [8–10] we showed that the nontrivial Coulomb effect in a small 2D InAs quantum dot supports a bound quadron rather than a traditional biexciton. However, the limitation of these studies is that only the ground states were investigated, and only in a small range of electron-to-hole confinement ratios.

In this work, in order to have a complete comparison picture between biexcitons and quadrons, we consider the quadron and biexciton binding energies in the full range of different parameters, namely, i) electron-to-hole confinement ratios for both ground and excited states, ii) electron-to-hole mass ratios, and iii) electron-to-hole oscillator length ratio. We also discuss the effect of the magnetic field on the binding energy of the quadron and biexciton.

It is found that in the ground state of 2D InAs quantum dots, where the spins of the electrons are anti-parallel, the quadron binding energy is positive within a certain range of electron-to-hole oscillator length ratios while the biexciton binding energy is always negative. For the excited state, we find the interval of the electron-to-hole oscillator length ratio where the transition from anti-binding to binding states of biexciton occurs, i.e. the interval where the biexciton is bound inside and unbound outside. Such a crossover of anti-binding to binding states for quadrons has been found as well. Moreover, the range of crossing for the quadron is much larger than that for the biexciton. Our results help further clarify the properties of the basic excitations in self-organized semiconductor quantum dots, in particular clarifying the results in [18] on sensitive changes of the binding energy within quantum dot ensembles.

Our paper is organized as follows. We briefly describe the model in section 2. The numerical results and discussions are presented in section 3. We conclude our work in section 4.

2. The model

We focus on self-assembled quantum dots where charged particles can be modeled as a 2D electron–hole system with parabolic potential. As in [8–10] we use the model of a system of interacting electrons and holes confined in a 2D quantum dot with a parabolic lateral potential in the presence of a perpendicular magnetic field $\vec{B}||z$. Within the effective-mass approximation, for a system of N electrons and M holes ($N = M = 2$ for biexciton and quadron) the total Hamiltonian is written as

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

where the terms are sums of Hamiltonians of single electrons $h(\vec{r}_i)$, single holes $h'(\vec{r}_k)$, the Coulomb interactions between electrons, holes, and electron–hole, respectively. ϵ is the dielectric constant of the material.

The Hamiltonians for a single electron and single hole in a quantum dot with parabolic confinement in a magnetic field, are

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

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

where the spin Zeeman splitting due to interaction of the spin with the magnetic field has been omitted because of its smallness. 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 frequencies for the electron (hole) and the z -components of the orbital angular momentum operators of the electron (hole), respectively.

In the polar coordinate, $\vec{r} = (r, \varphi)$ the eigenfunction and the eigenvalue of the single electron (and the hole, with index e replaced by h) in the quantum states (n, m) reads as:

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

$$E_{n,m}^e = \Omega_e (2n + |m| + 1) + \frac{1}{2} m \omega_{ce}, \quad (5)$$

where $L_n^{|m|}(r)$ is generalized Laguerre polynomial, and $\Omega_e = (\omega_e^2 + \frac{1}{4}\omega_{ce}^2)^{1/2}$, $\alpha_e = \sqrt{m_e^* \Omega_e}$.

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

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

where the electron and hole orbitals $\psi_i(\xi), \psi'_k(\xi)$ in the Slater determinants are spin dependent: $\psi_i(\xi) = \phi_i^\alpha(\vec{r})\sigma(\alpha)$ or $\psi_i(\xi) = \phi_i^\beta(\vec{r})\sigma(\beta)$ for up- or down-spin electrons, and $\psi'_k(\xi) = \phi'_k\alpha(\vec{r})\sigma(\alpha)$ or $\psi'_k(\xi) = \phi'_k\beta(\vec{r})\sigma(\beta)$ for up- or down-spin holes. In the Hartree–Fock–Roothaan approach [8–10, 19], the spatial parts of electron and hole orbitals $\phi_i^{\alpha,\beta}(\vec{r})$ and $\phi'_k\alpha, \beta(\vec{r})$ are expanded in the basis of the single electron and single hole, respectively:

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

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

We then calculate the matrix elements of the Coulomb interactions analytically, as in [20]. Solving this system of equations self-consistently, we obtain the total energy of the system as follows:

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

where the explicit expressions for $P_{\mu\nu}^T, P'_{\mu\nu} T, P_{\mu\nu}^{\alpha}, P_{\mu\nu}^{\beta}, P'_{\mu\nu} \alpha, P'_{\mu\nu} \beta$ and $F_{\mu\nu}^{\alpha,\beta}, F'_{\mu\nu} \alpha, \beta$ are calculated via the expansion coefficients (see in [8]).

3. The numerical results and discussions

In this section we will discuss the differences between the results obtained for quadrans and biexcitons. We use the parameters of the InAs/GaAs self-assembled quantum dots for the numerical computation input, as in [8–10]: $m_e = m_e^*/m_o = 0.067$, $m_h = m_h^*/m_o = 0.25$, $\omega_e = 49$ meV, $\omega_h = 25$ meV, $\epsilon_s = 12.53$. The adopted units of length and energy are $a_B^* = \epsilon_s/m_e^*e^2 = 9.9$ nm, $2Ry^* = m_e^*e^4/\epsilon_s^2 = 11.61$ meV. The oscillator lengths for electrons and holes in the absence of magnetic fields $l_{e,h} = (m_{e,h}^*\omega_{e,h})^{-1/2}$ are 4.8 nm and 3.5 nm, respectively, much smaller than the effective excitonic Bohr radius of about 13 nm, which means that electrons and holes in small InAs/GaAs dots are strongly confined. The binding energies of biexcitons and quadrans will be considered under the following conditions: 1) effect of confinement, 2) effect of exchange interaction, 3) impact of mass values, 4) dependence on oscillator strength and 5) dependence on magnetic field.

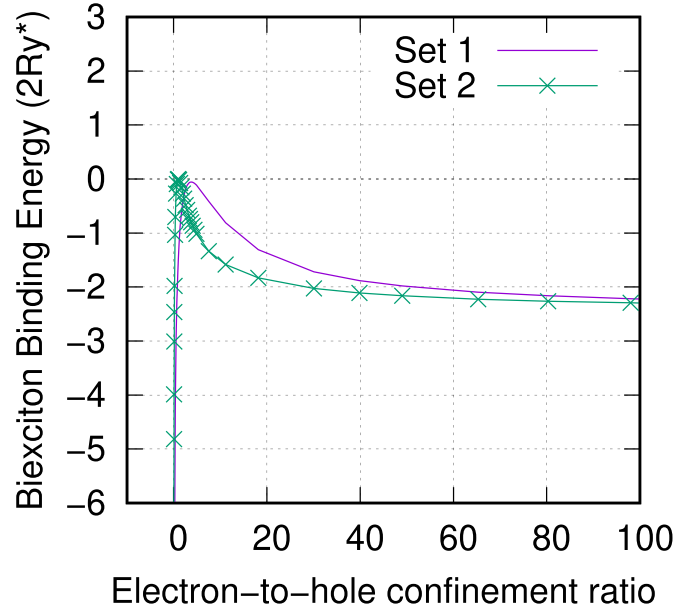


Figure 1. Biexciton binding energies in the ground state as functions of electron-to-hole confinement ratio (ω_e/ω_h) in the absence of magnetic field for two data sets of masses.

3.1. Effect of confinement

To study the effect of confinement on the binding energies, for the purpose of comparison we use two parameter sets, set 1: $\omega_e = 49$ meV, $m_e = 0.067$, and $m_h = 0.25$, set 2: $\omega_e = 49$ meV, $m_e = m_h = 0.067$. The binding energies of the biexciton, exciton and the quadrans in the ground state in the absence of magnetic field as functions of electron-to-hole confinement ratio (ω_e/ω_h) for two parameter sets 1 and 2 are shown in figures 1–3 respectively.

One can see in figure 1 that with any value of the electron-to-hole confinement ratio ω_e/ω_h , the biexciton binding energy is always negative, indicating that the biexciton is anti-binding. The positive binding energies of the excitons are clearly seen in figure 2. However, the quadrans binding energy, as shown in figure 3, can have both positive and negative values, depending on the confinement ratio. Namely, it is positive within the interval of $\omega_e/\omega_h \in [0.2 : 55]$ for the parameter set 1 ($m_e = 0.067; m_h = 0.25$) and within the interval of $\omega_e/\omega_h \in [0.2 : 19]$ for the parameter set 2 ($m_e = m_h = 0.067$), indicating the quadrans is bound in this interval. It becomes unbound when crossing outward in this interval. We thus show the essential differences between biexcitons and quadrans. This will be further confirmed in the next subsections.

3.2. Effect of exchange interactions

As the system we consider consists of two electrons and two holes, the exchange interaction between two electrons and between two holes must be taken into account. In the ground state, the spins of electrons are anti-parallel so there is no exchange interaction either between the electrons or between the holes. In the first excited states where the two electron spins

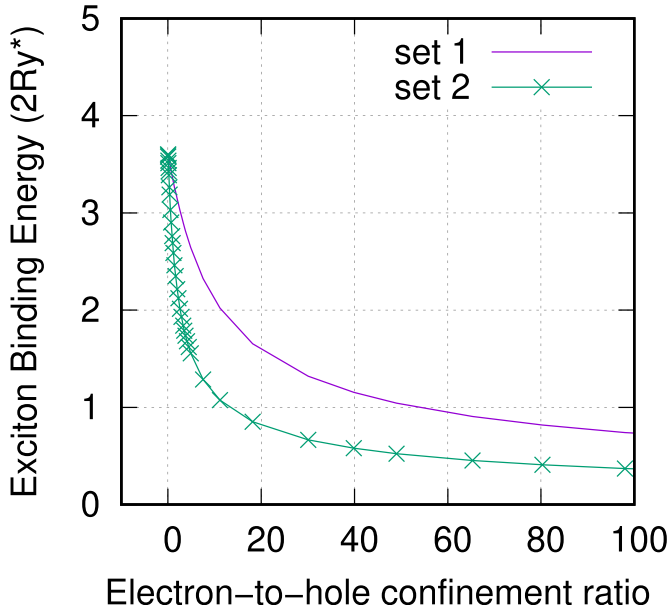


Figure 2. Exciton binding energies in the ground state as functions of electron-to-hole confinement ratio (ω_e/ω_h) in the absence of magnetic field for two data sets of masses.

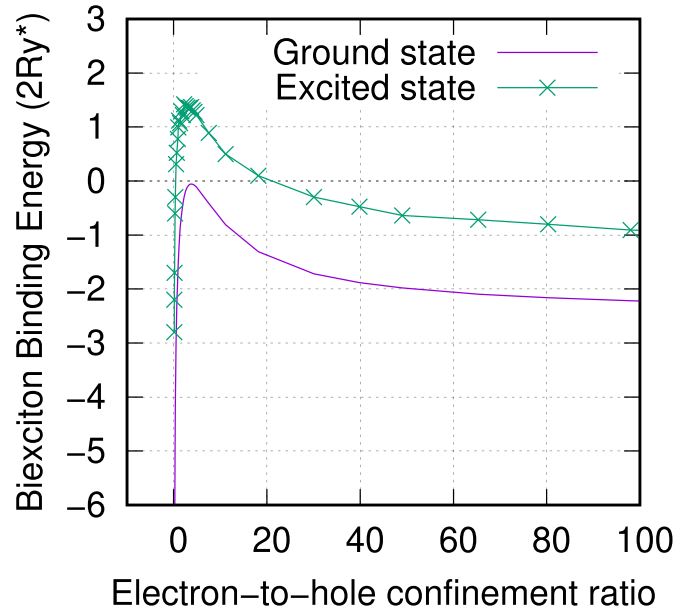


Figure 4. Biexciton binding energies in the ground and excited states as functions of electron-to-hole confinement ratio ω_e/ω_h in the absence of magnetic field for the parameter set 1 with $m_e = 0.067; m_h = 0.25$.

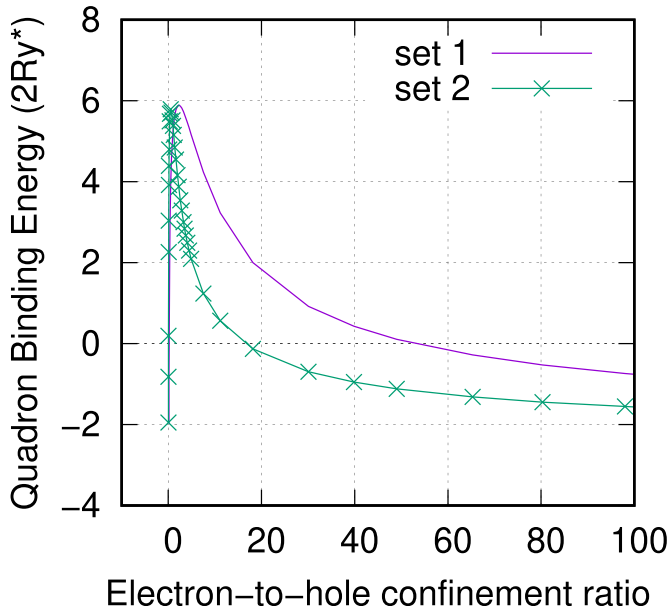


Figure 3. Quadron binding energies in the ground state as functions of electron-to-hole confinement ratio (ω_e/ω_h) in the absence of magnetic field for two data sets of masses.

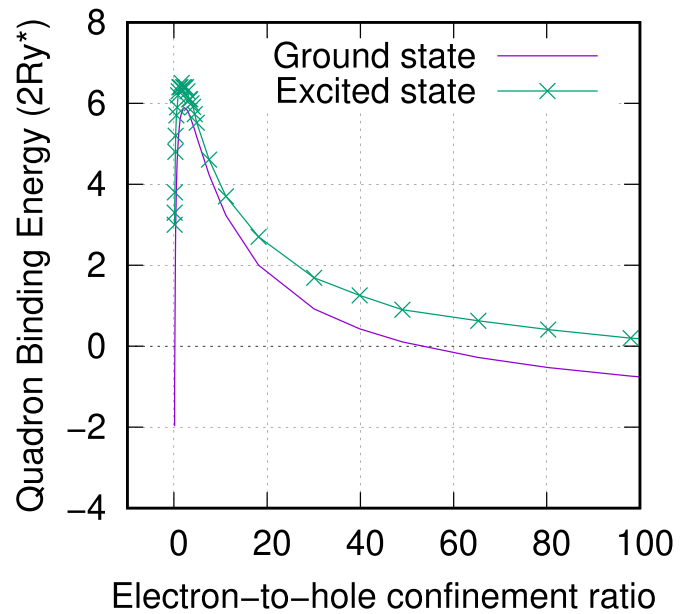


Figure 5. Quadron binding energies in the ground and excited states as functions of electron-to-hole confinement ratio (ω_e/ω_h) in the absence of magnetic field for the case $m_e = 0.067; m_h = 0.25$.

and the hole spins are parallel, there arise the exchange interactions between the electrons and between the holes.

In figures 4 and 5 we show the biexciton and quadron binding energies in the ground and excited states as functions of electron-to-hole confinement ratio ω_e/ω_h in the absence of magnetic field for the parameter set 1: $m_e = 0.067; m_h = 0.25$, where the exchange interaction is included for the excited state. We now discuss some features of these figures. First, one should note that the binding energy in the excited state is always larger than that in the ground state due to the

contributions of the exchange interactions of charged particles. Second, in figure 4 we notice that while the biexciton in the ground state is unbound for all values of ω_e/ω_h , its excited state becomes bound in the certain interval of ω_e/ω_h where the binding energy changes from negative to positive and vice versa. For the quadron, in figure 5 one can see that the bound states exist in both ground and excited states. However, the bound state in the excited state appears in the larger range of ω_e/ω_h in comparison with the one in the

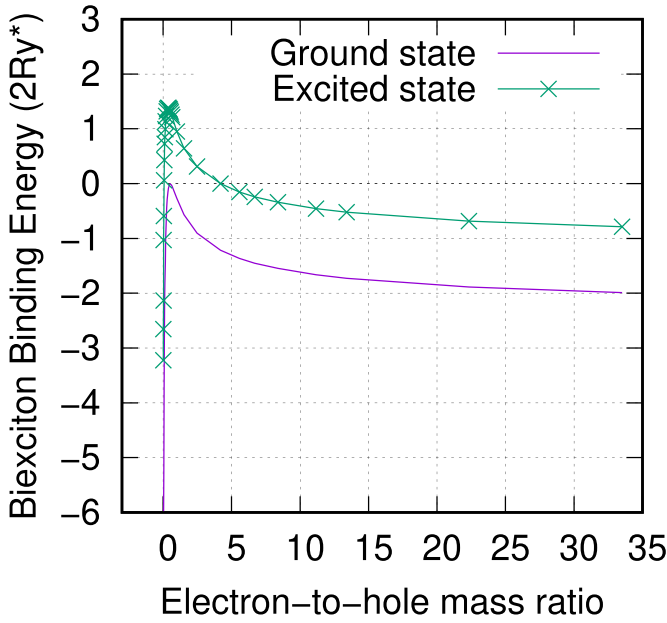


Figure 6. Biexciton binding energies in the ground and excited states as functions of electron-to-hole mass ratio (m_e/m_h) in the absence of magnetic field for the case $\omega_e = 49$ meV; $\omega_h = 25$ meV.

ground state. The above results show that the range of the ratio ω_e/ω_h for the binding energy to be positive is much larger for the quadron than for the biexciton, indicating that the quadron excitation is more supported by the quantum dot confinement.

3.3. Impact of mass values

The problem that we would like to investigate in this subsection is how the electron-to-hole mass ratio affects the formation of biexcitons and quadrons. Does there exist any range of the ratio that prefers one excitation more than the other? In order to see this, in figures 6 and 7 we show the biexciton and quadron binding energies in the ground and excited states as a function of electron-to-hole mass ratio m_e/m_h for the case $\omega_e = 49$ meV; $\omega_h = 25$ meV. As in the previous subsection, here we also see the similar behavior of the mass effect on the biexciton and quadron binding energies. First, in figure 6, as in figure 4, we notice that while the biexciton in the ground state is unbound for the entire range of electron-to-hole mass ratio m_e/m_h , its excited state becomes bound in certain intervals of m_e/m_h where the binding energy changes its sign from minus to plus and vice versa. Second, the binding energy in the excited state is always larger than at the ground state due to the additional contributions of the exchange interaction of charged particles. For the quadron, in figure 7, similarly to in figure 5, one can see that the bound states can exist in both ground and excited states and the value range of m_e/m_h for the bound state has been much enlarged in the excited states compared to the ground one. Again, we see that in the quantum dot the formation of the quadron is more supported than the biexciton.

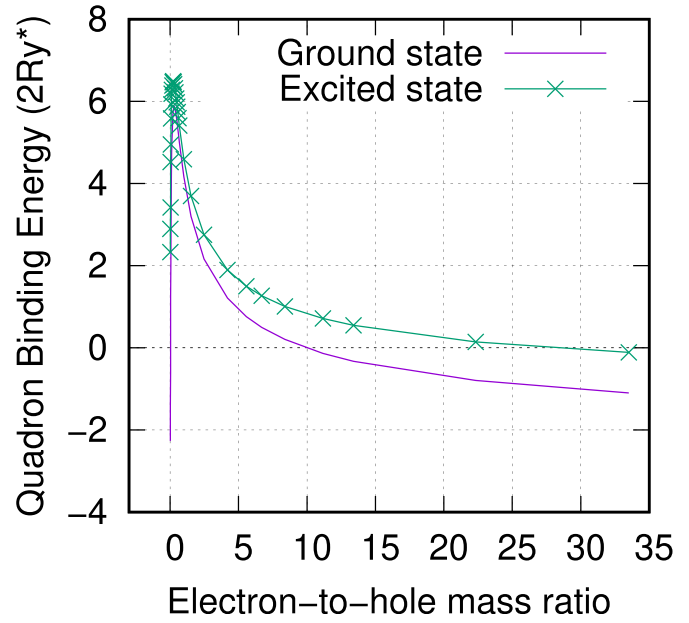


Figure 7. Quadron binding energies in the ground and excited states as functions of electron-to-hole mass ratio (m_e/m_h) in the absence of magnetic field for the case $\omega_e = 49$ meV; $\omega_h = 25$ meV.

3.4. Dependence on oscillator length

In the two previous subsections, we showed the dependence of the binding energy of the biexciton and the quadron on the ratio of the confinement potential, as well as the mass between the electron and the hole. However, we know that the carrier oscillator length is an important parameter that has a more universal meaning and could affect the binding energy and the formation of an excitation when considering systems of interacting charge carriers. Therefore, in this subsection, we recalculate the binding energies as a function of this important parameter—the oscillator length ratio between electrons and holes.

Thus, the binding energies of the exciton, biexciton and quadron in the ground state in the absence of magnetic field as functions of electron-to-hole oscillator length ratio are shown in figure 8 for two parameter sets, set 1: $m_e = 0.067$; $m_h = 0.25$ and set 2: $m_e = m_h = 0.067$. It is noted that these binding energies are almost unchanged when the masses are varied.

In figure 9 the binding energies of the biexciton and quadron in the ground state as a function of electron-to-hole oscillator length ratio are compared in two cases. In the first case we fix the masses of the electron and hole, namely with $m_e = 0.067$, $m_h = 0.25$ and change the electron-to-hole confinement ratio. In the second case we fix the confinement, namely with $\omega_e = 49$ meV, $\omega_h = 25$ meV and change the mass ratio between the electron and hole. One can clearly notice in both figures 8 and 9 that when scaled to the electron-to-hole oscillator length ratio the binding energies of all quasi-particles calculated in the two cases are almost the same. This means that neither the electron-to-hole confinement ratio nor the electron-to-hole mass ratio separately, but their combined

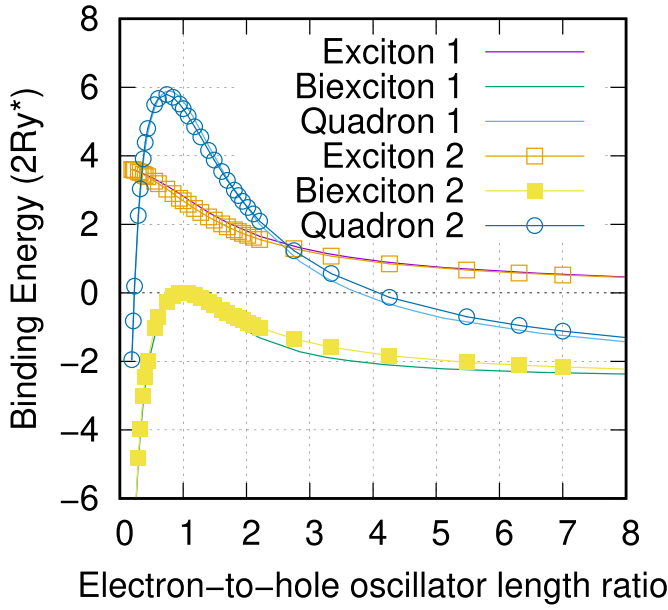


Figure 8. Binding energies of the exciton, biexciton and quadron in the ground state as functions of electron-to-hole oscillator length ratio (l_e/l_h) in the absence of a magnetic field for parameter set 1 ($m_e = 0.067; m_h = 0.25$) and set 2 ($m_e = m_h = 0.067$).

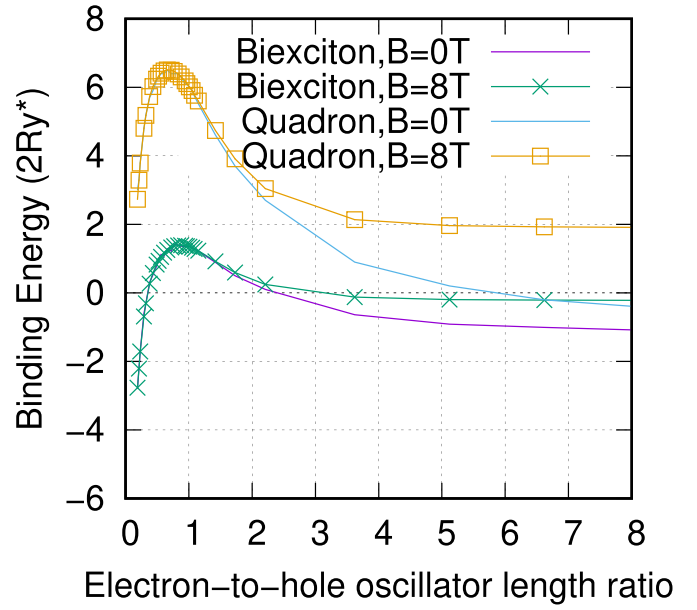


Figure 10. The binding energies of the biexciton and quadron in the excited state as functions of electron-to-hole oscillator length ratio (l_e/l_h) for parameter set 1 with $m_e = 0.067; m_h = 0.25$ in two cases $B = 0T$ and $B = 8T$.

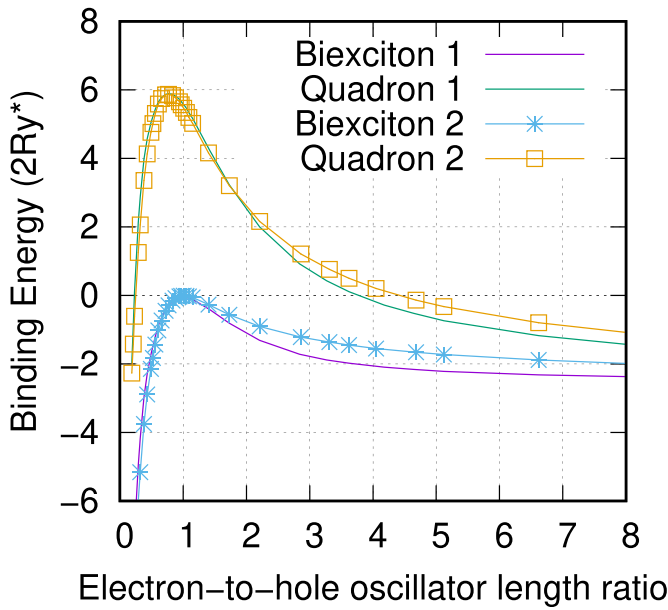


Figure 9. Binding energies of the biexciton and quadron in the ground state as functions of electron-to-hole oscillator length ratio (l_e/l_h) in the absence of a magnetic field are compared in two cases: case1 with $m_e = 0.067; m_h = 0.25$ and case 2 with $\omega_e = 49$ meV; $\omega_h = 25$ meV.

electron-to-hole oscillator length ratio does play a role of universal parameter that controls the binding of the excitonic system.

Again, here we observe the transition from a binding to an anti-binding state of the quadron for regions of too large or too small values of the electron-to-hole oscillator length ratio.

3.5. Dependence on magnetic field

To complete the picture, the effect of magnetic field on the formation of both the quadron and the biexciton has been considered. Figure 10 shows the dependence of the binding energies of the biexciton and quadron in the excited state on the magnetic field in two cases $B = 0T$ and $B = 8T$ for parameter set 1 ($m_e = 0.067; m_h = 0.25$). The magnetic field enhances the binding energies due to additional confinement caused by the field on the electron and hole (see (2) and (3)). However, this enhancement is weak at small values of electron-to-hole oscillator length ratio, about $0.03 (2Ry^*) \approx 0.35$ meV at $B = 8T$. At large ratios of the oscillator strengths between the electron and the hole the magnetic field effect is more evident.

To confirm the method of our calculation, a comparison with experimental results was made. A good agreement between our results and the experimental data for the binding energy of the biexciton [8, 9] and charged excitons [11] has been obtained. Here we note that for an InAs/GaAs self-assembled quantum dot, the electron-to-hole oscillator length ratio $l_e/l_h = 1.3$ corresponds to the value of our calculated biexciton binding energy $-0.45 (2Ry^*) \approx -5.1$ meV, which is well consistent with the experimental data interval $[-1 \text{ meV} : -6 \text{ meV}]$ in [16]. Interestingly, our new results can help to understand and shed light on sensitive changes in the binding energy of a biexciton in natural ensembles of InAs/GaAs quantum dots with randomly fluctuating parameters [18].

4. Conclusion

In conclusion, we have completed a comparison between the conventional biexciton and the quadron, a new type of

four-carrier excitation, in 2D parabolic quantum dots by unrestricted Hartree–Fock method.

The biexciton and quadron binding energies have been calculated for a full range of electron-to-hole confinement and mass ratios, and exchange interaction. It is found that in the ground state of 2D InAs quantum dots, where the spins of the electrons are anti-parallel, the biexciton binding energy is negative in the whole range while the quadron binding energy is positive within a certain range of electron-to-hole oscillator length ratios. For the excited state with parallel carrier spins, we find that both the biexciton and quadron binding energies are larger than those in the ground state, and the value range in which the quadron binding energy is positive is larger than that of the biexciton. The crossover between anti-binding and binding states and vice versa for the biexciton and quadron have been predicted.

The magnetic field dependence of the binding energy has also been investigated and it is shown that a magnetic field increases the binding energies of biexcitons and quadrons but the effect is rather small. However, the impact of the magnetic field is enhanced at large ratios of the oscillator strengths between the electron and the hole.

In a full account, our results show that not only are the biexciton and the quadron different quasiparticles but also in the self-assembled quantum dot configuration the formation of the quadron is more favorable than that of the biexciton. Our study of binding energies in semiconductor quantum dots across a wide range of parameters offers valuable insights for tailoring their properties and facilitating advancements in the design of materials with customized electronic properties. This, in turn, allows for the customization and optimization of materials with specific optical and electronic characteristics tailored for distinct electronic applications. They are particularly useful for experimentalists and serve as suggestions for experimental findings related to quadrons in quantum dots.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

Acknowledgments

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