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# Binding energy of biexcitons in quantum dots

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## Abstract

We propose a procedure to solve exactly the Schrödinger equation for a biexciton composed of two electrons and two holes in a harmonic quantum dot by using the hyperspherical coordinates. The binding energy of the ground state has been obtained as a function of the electron–hole mass ratio. Dependence of the ratio of the binding energy of a biexciton to that of an exciton on the mass ratio in a quantum dot has been obtained.

## 1. Introduction

Semiconductor nanocrystals dispersed in a large-band gap matrix can be treated as quantum dots (QD). Three-dimensional (3D) quantum confinement of electrons, holes and excitons makes it possible to observe quantum size effects in QD [1–3]. A biexciton is a system consisting of two excitons which are bound together. Since the first observation of biexcitons in quantum wells reported by Miller *et al* [4], there have been many studies, both experimental and theoretical, on this subject. From a theoretical point of view, they calculated the binding energy of the biexciton ground-state in a spherical QD and the oscillator strength as a function of the QD radius, and the electron–hole mass ratio by using a variational approach [5,6]. Recently, the effect of exciton localization on the binding energy [7–12] and dephasing of biexcitons [13,14] have also been studied, both in III–V and II–VI material systems.

Kleinman [15] developed a variational model that gives the Haynes factor,  $f_H = E_b^{xx}/E_b^x = 0.564$  which is the ratio between the biexciton-binding energy ( $E_b^{xx}$ ) and the exciton-binding energy ( $E_b^x$ ) for  $\sigma = 0$  (hydrogen limit), 0.15 for  $\sigma = 0.68$  (GaAs), and 0.14 for  $\sigma = 1$  (positronium limit) at zero quantum well width. Kleinman identified this ratio with the Haynes factor [16] of 0.1 in 3D. This theoretical result produced as high a value of  $E_b^{xx}/E_b^x$  as predicted by Haynes for 3D biexcitons, therefore Kleinman concluded correctly that the binding energy of 2D biexcitons has to be larger than that of 3D biexcitons. Several papers have reported even higher values of the binding energy of quasi-2D biexcitons [17–22]. It is obvious that the quantum confinement leads to a significant increase in the binding energy of biexcitons. The aim of the present paper is to investigate the quantum confinement effect on the binding energy of the biexciton ground-state in a disk-like QD. In experimentally realized QD, the motion in the  $z$  direction is always frozen out into the lowest subband. Since the corresponding extent of the wave function is much less than the one in the  $x$ – $y$  plane, we can

treat the QD in a disk-like 2D limit. For most QD, the harmonic oscillator is a very good approximation to describe the lateral confinement.

The hyperspherical approach has been applied to solve bound states and scattering problems in many different fields of physics and chemistry. Many of the earlier works dealt with the basic structure of the mathematical functions encountered in hyperspherical coordinates. In recent years, computational techniques have been developed to perform accurate calculations in hyperspherical coordinates and the hyperspherical approach provides a direct and conceptually elegant method for treating problems of three-electron systems [23], charged-exciton complexes [24] and excitons bound to ionized donors [25].

In this paper, we will propose a procedure to diagonalize the Hamiltonian of a biexciton in the QD with a parabolic lateral confining potential by using the correlated hyperspherical harmonics as basis functions. The binding energy of the ground state of a biexciton in 2D QD subjected to a parabolic confinement is obtained as a function of the electron-to-hole mass ratio. The ratio of the binding energy of a biexciton to that of an exciton in QD is larger than those of biexcitons in 2D and 3D semiconductors.

## 2. Formalism

Let us consider a system of two electrons and two holes moving in the  $x$ - $y$  plane subjected to a parabolic confinement. In our model, we will follow [26] and use the same parabolic frequency for both holes and electrons but with widely different masses. Furthermore, we suppose that the effective mass of electrons (holes) in the QD is the same as that in the barrier materials. With the effective mass approximation, the Hamiltonian for two electrons and two holes in a disk-like QD reads

$$H = \sum_{i=1}^4 \left[ \frac{P_i^2}{2m_i} + \frac{1}{2} m_i \omega^2 r_i^2 \right] + V_c \quad (1)$$

where  $m_1 = m_2 = m_e^*$  and  $m_3 = m_4 = m_h^*$  are the effective masses of particles,  $\vec{r}_i$  and  $\vec{p}_i$  denote the position vector and the momentum vector of particle  $i$ , respectively, and  $\omega$  is the strength of the confinement. The interaction between the four particles is modelled by a Coulombic potential which is screened by a quite phenomenological dielectric constant  $\epsilon$

$$V_c = \frac{1}{4\pi\epsilon} \left( \frac{1}{r_{12}} + \frac{1}{r_{34}} - \frac{1}{r_{13}} - \frac{1}{r_{14}} - \frac{1}{r_{23}} - \frac{1}{r_{24}} \right). \quad (2)$$

To maintain that the kinetic energy operator is diagonal, we introduce a set of the centre of mass ( $c.m.$ ) and Jacobi coordinates to describe the motion of the particles,

$$\vec{R} = \frac{m_e^*(\vec{r}_1 + \vec{r}_2) + m_h^*(\vec{r}_3 + \vec{r}_4)}{M} \quad (3)$$

$$\vec{\xi}_1 = \vec{r}_2 - \vec{r}_1, \quad \vec{\xi}_2 = \vec{r}_4 - \vec{r}_3 \quad (4)$$

$$\vec{\xi}_3 = [m_h^*(\vec{r}_3 + \vec{r}_4) - m_e^*(\vec{r}_1 + \vec{r}_2)] / M. \quad (5)$$

Equation (1) can then be divided into two independent parts

$$H = H_{c.m.} + H_{rel} \quad (6)$$

with

$$H_{c.m.} = \frac{P_{c.m.}^2}{2M} + \frac{1}{2} M \omega^2 R_{c.m.}^2 \quad (7)$$

$$H_{rel} = \sum_{v=1}^3 \left( \frac{p_v^2}{2\mu_v} + \frac{1}{2} \mu_v \omega^2 \xi_v^2 \right) + V_C \quad (8)$$

where  $H_{c.m.}$  describes the *c.m.* motion,  $H_{rel}$  describes the relative motion,  $M = 2m_e^* + 2m_h^*$  is the total mass,

$$\mu_1 = \frac{m_e^*}{2} \quad \mu_2 = \frac{m_h^*}{2} \quad \mu_3 = \frac{2m_e^*m_h^*}{m_e^* + m_h^*}$$

are the reduced masses, respectively. The eigenvalues of  $H_{c.m.}$  are obviously the ordinary 2D harmonic oscillator functions.

Let  $\Psi_{LS}$  be the wave function of the state with total orbital-angular momentum  $L$  and total spin-angular momentum  $S$ . In this coordinates system, the Schrödinger equation in the *c.m.* frame takes the form

$$\left[ \sum_{v=1}^3 \left( \frac{p_v^2}{2\mu_v} + \frac{1}{2} \mu_v \omega^2 \xi_v^2 \right) + V_C - E \right] \Psi_{LS}(\vec{\xi}_1, \vec{\xi}_2, \vec{\xi}_3) = 0. \quad (9)$$

This particular set of Jacobi coordinates will be referred to as the  $\alpha$ -set. There are two other possible Jacobi coordinates: the  $\beta$ -set where  $\vec{\xi}_1^{(\beta)}$  is the vector from 1 to 3,  $\vec{\xi}_2^{(\beta)}$  is from 2 to 4 and  $\vec{\xi}_3^{(\beta)}$  is from the *c.m.* of 1 and 3 to the *c.m.* of 2 and 4; and the  $\gamma$ -set where  $\vec{\xi}_1^{(\gamma)}$  is the vector from 1 to 4,  $\vec{\xi}_2^{(\gamma)}$  is from 2 to 3 and  $\vec{\xi}_3^{(\gamma)}$  is from the *c.m.* of 1 and 4 to the *c.m.* of 2 and 3. These vectors are depicted in figure 1. The  $\beta$ - and  $\gamma$ -set are similar to those used to describe the biexciton systems. We also notice that the Schrödinger equation (3) can be written in terms of the  $\beta$ -set or  $\gamma$ -set Jacobi coordinates as well, with the corresponding reduced masses  $\mu_1$ ,  $\mu_2$  and  $\mu_3$ . The superscripts in the coordinates and reduced masses will not be specified unless such a distinction is necessary in the discussion.

One can introduce mass-weighted hyperspherical coordinates by defining

$$\vec{\eta}_1 = \sqrt{\mu_1/\mu} \vec{\xi}_1 \quad \vec{\eta}_2 = \sqrt{\mu_2/\mu} \vec{\xi}_2 \quad \vec{\eta}_3 = \sqrt{\mu_3/\mu} \vec{\xi}_3 \quad (10)$$

where  $\mu$  is arbitrary (taken to be unity in general). In terms of  $\vec{\eta}$ , the Schrödinger equation (3) now takes the form

$$\left[ \sum_{i=1}^3 \left( -\frac{\hbar^2}{2\mu} \nabla_{\eta_i}^2 + \frac{1}{2} \mu \omega^2 \eta_i^2 \right) + V_C - E \right] \Psi_{LS}(\vec{\eta}_1, \vec{\eta}_2, \vec{\eta}_3) = 0. \quad (11)$$

Such that the kinetic energy and the confinement potential operators associated with the three 'particles' are identical, and the mass dependence of the four particles is cast in the rescaling of the distances.

From  $\vec{\eta}$ , it is straightforward to introduce the hyper-spherical coordinates

$$\eta = \sqrt{\eta_1^2 + \eta_2^2 + \eta_3^2} \quad \eta_1 = \eta \cos \phi_1 \quad \eta_2 = \eta \sin \phi_1 \cos \phi_2 \quad \eta_3 = \eta \sin \phi_1 \sin \phi_2 \quad (12)$$

where  $\eta$  is the hyper-radius and  $\phi = \phi^{(i)}$  ( $i = \alpha, \beta, \gamma$ ) is the hyper-angle. We note that the hyper-radius  $\eta$  is independent of which set of Jacobi coordinates is used. Thus the three vectors  $\vec{\eta}_1$ ,  $\vec{\eta}_2$  and  $\vec{\eta}_3$  are replaced by nine coordinates  $(\eta, \Omega)$ , where  $\Omega = (\phi_1, \phi_2, \theta_1, \varphi_1, \theta_2, \varphi_2, \theta_3, \varphi_3)$  denotes collectively the eight angles,  $\theta_i$  and  $\varphi_i$  being the spherical angles of the vector  $\vec{\eta}_i$ .

With hyper-spherical coordinates splits, the Schrödinger equation of the relative motion for the biexciton in a harmonic well is given by

$$\left[ -\frac{\hbar^2}{2\mu} \left( \frac{1}{\eta^5} \frac{d}{d\eta} \eta^5 \frac{d}{d\eta} - \frac{\Lambda^2(\Omega)}{\eta^2} \right) + \frac{C}{\eta} + \frac{1}{2} \mu \omega^2 \eta^2 + E \right] \Psi_{LS}(\eta, \Omega) = 0 \quad (13)$$

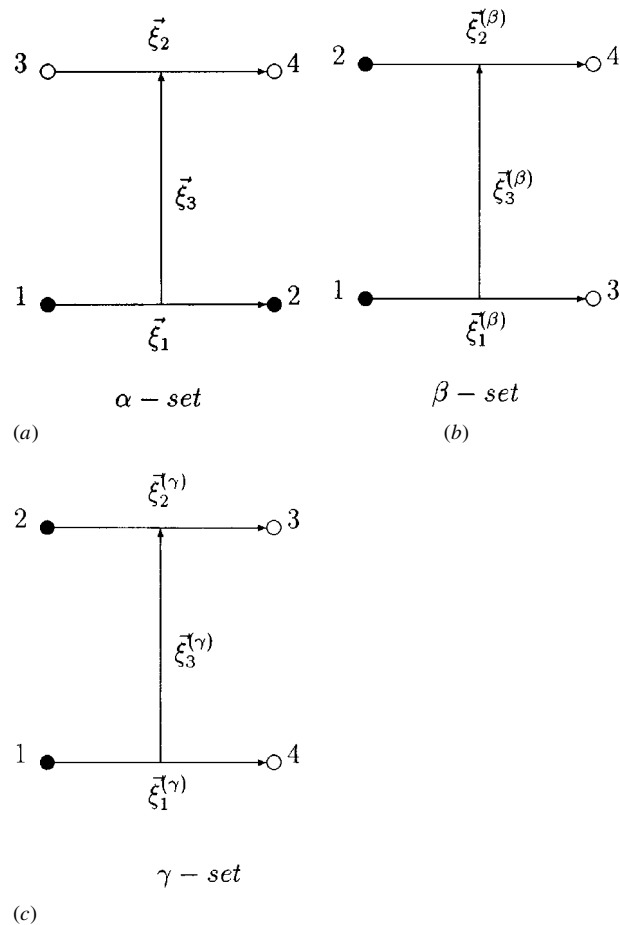


Figure 1. Three sets of Jacobi relative coordinates for a biexciton system

where  $\Lambda^2(\Omega)$  is the grand angular momentum operator, defined by

$$\Lambda^2(\Omega) = \frac{\partial^2}{\partial^2 \phi_1} + \left[ 3 \frac{\cos \phi_1}{\sin \phi_1} - \frac{\sin \phi_1}{\cos \phi_1} \right] \frac{\partial}{\partial \phi_1} + \frac{1}{\sin^2 \phi_1} \frac{\partial^2}{\partial^2 \phi_2} + \left[ \frac{\cos \phi_2}{\sin \phi_2} - \frac{\sin \phi_2}{\cos \phi_2} \right] \frac{\partial}{\partial \phi_2} - \frac{\ell^2(\varphi_1)}{\cos^2 \phi_1} - \frac{\ell^2(\varphi_2)}{\sin^2 \phi_1 \cos^2 \phi_2} - \frac{\ell^2(\varphi_3)}{\sin^2 \phi_1 \sin^2 \phi_2} \quad (14)$$

where  $\ell^2(\varphi_i) = -i\partial/\partial\varphi_i$ ,  $\varphi_i$  is the polar angle of  $\vec{\eta}_i$ . The eigenvalues and eigenfunctions for the  $\Lambda^2(\Omega)$  operator are known,

$$\Lambda^2(\Omega)Y_{[K]}(\Omega) = \lambda(\lambda + 4)Y_{[K]}(\Omega) \quad (15)$$

where

$$\lambda = 2m_1 + 2m_2 + |\ell_1| + |\ell_2| + |\ell_3| \quad (16)$$

and the eigenfunctions are

$$Y_{[K]}(\Omega) = N_{m_1 m_2}^{\ell_1 \ell_2 \ell_3} P_{m_1}^{\ell_1 \ell_2}(\phi_1) P_{m_1 m_2}^{\ell_1 \ell_2 \ell_3}(\phi_2) e^{i\ell_1 \varphi_1} e^{i\ell_2 \varphi_2} e^{i\ell_3 \varphi_3} \quad (17)$$

where  $[K]$  denotes the set of quantum numbers,  $[K] = (\ell_1, \ell_2, \ell_3, m_1, m_2)$ , with  $m_i$  related to the polynomial functions in angle  $\phi_i$ ,  $\ell_1 + \ell_2 + \ell_3 = L$  is the total orbital angular momentum,

where  $N_{m_1 m_2}^{\ell_1 \ell_2 \ell_3}$  is the normalization constant and  $P_{m_1}^{\ell_1 \ell_2}(\phi_1)$  is a Jacobi polynomial. In the coordinate  $\alpha$ -set, the particle exchange symmetry is automatic, i.e.  $\ell_1(\ell_2) = \text{odd}$  for spin-triplet states of two identical particles and  $\ell_1(\ell_2) = \text{even}$  for spin-single states of two identical particles.

The grand angular momentum operator does not depend on the Jacobi coordinates

$$\Lambda^2(\Omega^\alpha) = \Lambda^2(\Omega^\beta) = \Lambda^2(\Omega^\gamma) \quad (18)$$

therefore, the hyper-spherical harmonics in different sets of hyper-angles are simply the different representations. The eigenfunctions in one set can be expanded in terms of eigenfunctions of the other set with  $\lambda_{[K]} = \lambda_{[K']}$ ,

$$Y_{[K]}(\Omega^j) = \sum_{[K']} B_{[K],[K']}(\eta) Y_{[K']}(\Omega^j) \quad (19)$$

where  $\eta$  depends on the mass ratio of the system, and the expansion coefficients can be evaluated from

$$B_{[K],[K']}(\eta) = \int d\Omega^j Y_{[K]}^*(\Omega^j) Y_{[K']}(\Omega^j). \quad (20)$$

These coefficients are called the transformation bracket and a program for their evaluation has been published [27]. These transformation brackets are useful when evaluating integrals involving functions of different sets of Jacobi coordinates.  $C/\eta$  is the total Coulomb interaction potential among the four charged particles, with  $C$  given by

$$C = \frac{e^2}{\epsilon} \left[ \sqrt{\frac{\mu_1^\alpha}{\mu}} \frac{1}{\cos \phi_1^\alpha} + \sqrt{\frac{\mu_2^\alpha}{\mu}} \frac{1}{\sin \phi_1^\alpha \cos \phi_2^\alpha} - \sqrt{\frac{\mu_1^\beta}{\mu}} \frac{1}{\cos \phi_1^\beta} - \sqrt{\frac{\mu_2^\beta}{\mu}} \frac{1}{\sin \phi_1^\beta \cos \phi_2^\beta} \right. \\ \left. - \sqrt{\frac{\mu_1^\gamma}{\mu}} \frac{1}{\cos \phi_1^\gamma} - \sqrt{\frac{\mu_2^\gamma}{\mu}} \frac{1}{\sin \phi_1^\gamma \cos \phi_2^\gamma} \right]. \quad (21)$$

Since the set of hyper-spherical harmonics forms a complete set on each hyper-spherical surface, the solution of the Schrödinger equation can be expanded as

$$\Psi_{LS} = \sum_{[K]} R_{[K]}(\eta) Y_{[K]}(\Omega). \quad (22)$$

By projecting out the hyper-spherical harmonics, a set of coupled second-order hyperradial differential equations is obtained

$$\left[ -\frac{\hbar^2}{2\mu} \left( \frac{1}{\eta^5} \frac{d}{d\eta} \eta^5 \frac{d}{d\eta} - \frac{\lambda_{[K]}(\lambda_{[K]} + 4)}{\eta^2} \right) + \frac{1}{2} \mu \omega^2 \eta^2 - E \right] R_{[K]}(\eta) \\ - \frac{1}{\eta} \sum_{[K']} U_{[K],[K']} R_{[K']}(\eta) = 0 \quad (23)$$

where

$$U_{[K],[K']} = \langle Y_{[K]}(\Omega) | C | Y_{[K']}(\Omega) \rangle \quad (24)$$

is the matrix element of the effective charge  $C$  evaluated between two hyper-spherical harmonics. Equation (23) can be solved to obtain the eigenvalues if convergence can be achieved by using a reasonable truncated set of hyper-spherical harmonics.

When  $C = 0$ , the eigensolutions of equation (23) satisfying the conventional boundary condition with a finite  $R(0)$  given by

$$R_{n\lambda}(\eta) = N_{n\lambda} \left( \frac{\eta}{\eta_0} \right)^\lambda L_n^{\lambda+N-2} \left( \frac{\eta^2}{\eta_0^2} \right) e^{-\eta^2/2\eta_0^2} \quad (25)$$

where  $N_{n\lambda}$  is the normalization constant,  $\eta_0^2 = \hbar/(\mu\omega)$ , and  $L_n^{\lambda+N-2}$  is a Laguerre polynomial. The associated eigenvalue is  $\hbar\omega(2n + \lambda + 3)$ ,  $n = 0, 1, 2, \dots$ . The accuracy of the solutions depends on how large the model space is. Since we are interested only in the low-lying states and in the qualitative aspect, the model space adopted is neither very large to facilitate numerical calculation, nor very small to assure the qualitative accuracy. This is achieved by extending the dimension of the model space step by step, in each step the new results are compared with the previous results from a smaller space, until satisfactory convergence is achieved. Our main interest is the ground state, and consequently, both the total angular momentum  $L$  and the total spin  $S$  are zero.

### 3. Numerical results and discussions

In what follows the energies are in meV,  $m_e^* = 0.067m_e$  ( $m_e$  is the mass of a free electron),  $\epsilon = 12.4$ , which are relevant to GaAs, are adopted in the calculation. The biexciton-binding energy  $E_b^{xx}$  is defined as the difference between twice the binding energy  $E_b^x$  of the ground-state energy of the exciton and the biexciton ground-state energy  $E_{xx}$  that is

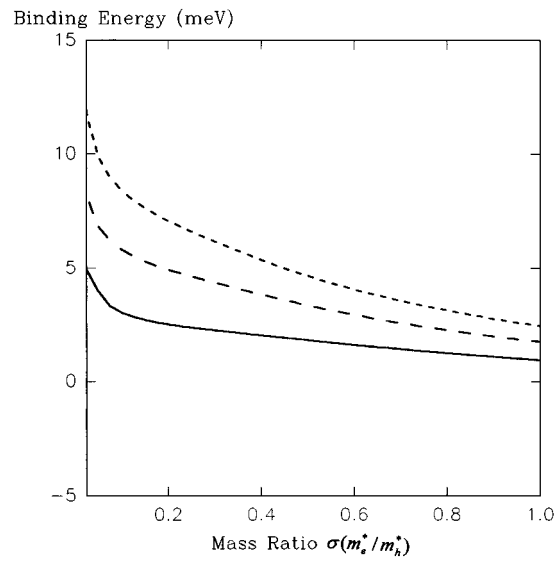
$$E_b^{xx} = 2E_b^x - E_{xx}. \quad (26)$$

As indicated in table 1, the convergence of the biexciton energy of the ground state is fast with the number of basic functions. The dependence of  $E_b^{xx}$  on the electron-to-hole mass ratio  $\sigma$  for the two values of quantum confinement  $\hbar\omega = 0$  (solid curve), 0.3 meV (dashed curve) and 0.5 meV (dotted curve) is plotted in figure 2. We observe that the biexciton confined in a QD has, in general, a larger binding energy than those in the 3D and 2D semiconductors [28]. This physical origin is that in semiconductor microstructures of lower dimensionality, the spatial overlap between an electron and a hole is increased, leading to the increase in the Coulomb binding energy. As an ultimate limit of the reduced dimensionality, the zero-dimensional materials such as QD show a much more enhanced binding energy of the biexciton ground-state. The binding energy increases with the decreasing electron-to-hole mass ratio  $\sigma$  as expected, i.e. the heavy hole gives rise to larger binding energies than the light hole, and takes a maximum value at the hydrogen limit ( $\sigma = 0$ ) as in the case of bulk materials [29]. Compared with the binding energies for different confinements, we find that the binding energy depends strongly on the strength of confinement of QD as a manifestation of the quantum confinement effect, i.e. the stronger the confinement, the higher the binding energy at the same  $\sigma$  value.

**Table 1.** The convergence of the biexciton ground-state energies with the number  $N_H$  of the basic functions for two values of the confinement in the case of  $\sigma = 0.68$ ,  $\hbar\omega_0 = 0.5Ry^*$  and  $d = 10.0$  nm.

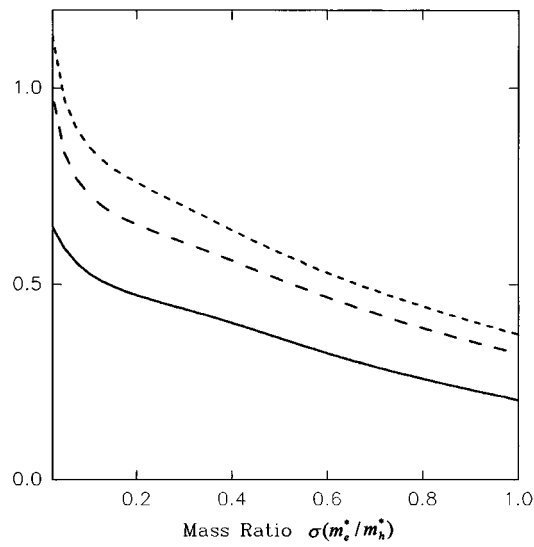
$N_H/\hbar\omega$	0.5 meV	1.0 meV
320	-10.172	-12.735
500	-10.790	-13.460
750	-11.049	-13.803

The Haynes factor  $f_H$  is the ratio between the biexciton-binding energy and the exciton-binding energy. Using a very simple model structure, Singh *et al* obtained  $f_H = E_b^{xx}/E_b^x = 0.228$  which is independent of  $\sigma$  [21]. However, the results obtained by Liu *et al* were that  $f_H$  ranged from 0.580 to 0.220, which varied with  $\sigma$  in 2D semiconductors [22]. In the present case, the ratio  $f_H = E_b^{xx}/E_b^x$  is calculated as a function of the electron-hole mass ratio  $\sigma$  and the result is depicted for  $\hbar\omega = 0$  (solid curve), 0.3 meV (dashed curve) and 0.5 meV (dotted curve) in figure 3. Our calculation shows that  $f_H = 0.645$  at  $\sigma = 0$  and  $f_H = 0.202$



**Figure 2.** Variations of the biexciton binding energies in a QD as a function of the mass ratio  $\sigma = m_e^*/m_h^*$  for  $\hbar\omega = 0$  (solid curve),  $\hbar\omega = 0.3$  meV (dashed curve) and  $\hbar\omega = 0.5$  meV (dotted curve).

$$f_H = E_b^{xx} / E_b^x$$



**Figure 3.** Ratio of the biexciton-binding energy to the exciton-binding energy  $f_H = E_b^{xx} / E_b^x$  in a QD as a function of the mass ratio  $\sigma = m_e^*/m_h^*$  for  $\hbar\omega = 0$  (solid curve),  $\hbar\omega = 0.3$  meV (dashed curve) and  $\hbar\omega = 0.5$  meV (dotted curve).

at  $\sigma = 1.0$  for  $\hbar\omega = 0$ . It is obvious that our result basically agrees with that of [22], i.e. the  $f_H$  varies with  $\sigma$ . On the other hand, we also see that qualitatively the zero-dimensional, two-dimensional and three-dimensional cases behave similarly. Experimentally, we know that the ratio  $f_H = 0.1$  from the Haynes rule for 3D case [16] and about 0.2 for quantum wells [20].



It is obvious that our calculated result of  $f_H$  is higher than these experimental values. Hence this is consistent with the trend that  $f_H$  increases with decreasing dimension.

Although, to our knowledge, no experimental results involving the biexcitons in QD have been reported, we hope that our results might be useful in the interpretation of future experimental data.

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