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The negatively charged exciton in double-layer quantum dots

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Abstract. The hyperangular equation for charged semiconductor complexes in a double-layer harmonic quantum dot was solved numerically by using the correlated hyperspherical harmonics as basis functions. By using this method, we have calculated the energy spectra of the low-lying states of a charged exciton as a function of the radius of the quantum dot and the binding energy spectra of the ground state as a function of the radius of the quantum dot for a few values of the distance between the vertically coupled dots and the electron-to-hole mass ratio.

PACS. 73.20.Dx Electron states in low-dimensional structures (superlattices, quantum well structures and multilayers) – 71.35.-y Excitons and related phenomena – 78.66.Fd III-V semiconductors

1 Introduction

Recently, there has been considerable experimental and theoretical interest in charged excitons in quantum dots (QD's). The charged excitons result from the binding of an exciton (an electron-hole pair) with an extra electron or hole in semiconductor QD's. The existence of two kinds of charged excitons was first suggested by Lampert [1]. In the past few years, experimental evidence of the existence of X^- (eeh) resulting from the Coulombic interaction between an exciton and an electron in semiconductor quantum well structures [2,3] has stimulated intensive theoretical study of charged excitons in semiconductor heterojunctions [4,5].

Due to relatively the small binding energy of trions, *i.e.* charged excitons, in bulk semiconductors, they can only be observed at very low temperatures ($T \leq 10$ K). This situation limits to some extent their practical importance in semiconductors. Recent progress in semiconductor technology has made possible the fabrication of individual QD's. Such microstructures confine charged particles in all three space dimensions. The reduced dimensionality enhances considerably the binding energy of charged excitons [6], and thus facilitates the experimental observations, X^+ (ehh) involving an exciton and a hole has also been observed experimentally [7]. Theoretically, several models have been proposed for evaluating the binding energy of charged excitons in ideal two dimensions at zero magnetic field; this includes a variational calculation using the Hylleraas-type trial wave function [6,8], analytical results for a linear model [9] and others [10]. From a theoretical point of view, these few-body systems represent a challenging problem. The standard tools of the condensed-matter physicist like the many-body techniques relying on Hartree or Hartree-Fock approximations are often not sufficient since the exchange and correlation energies can be far from negligible [11]. A fully quantum mechanical treatment is needed. In the general case, this requires numerical calculations that can become quite time-consuming as the number of particles grow.

The purpose of this paper is to present a model calculation of negative excitons in a structure consisting of two spatially separated coupled QD's by using the correlated hyperspherical harmonics as basis functions. The center-to-center distance of the two QD's is d. Each layer QD has only one type of charge carrier (two electrons or a hole). The effective separation d may be tuned by an applied electric field. For most QD's, the harmonic oscillator is a very good approximation to describe the lateral confinement [12,13]. Hence, we assume that the confinement of the $x-y$ plane is provided by a harmonic potential, and that the confinement of the z-direction is provided by an infinitely high potential, and the lowest subband in each QD is occupied.

We will investigate the energy spectra of the low-lying states of a negatively charged exciton in double-layer QD's as a function of the dot size. On the other hand, we will show the dependence of the binding energies of the negatively charged excitons on the radius R for a few values of the distance between double-layer QD's and the electron-to-hole mass ratio.

2 Model and method

Consider a charged exciton of strictly 2D e- and h-layers separated by a distance d (see Fig. 1). In both dots the lateral confining potential within each layer is assumed

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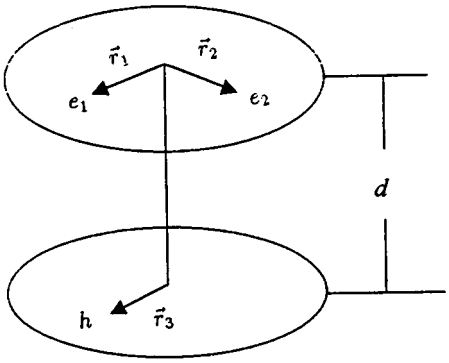

Fig. 1. Schematic configuration of double-layer quantum dots.

to be parabolic. With the effective-mass approximation, the Hamiltonian in the laboratory frame can be written

$$H = \sum_{i=1}^{3} \left(\frac{p_i^2}{2m_i^*} + \frac{1}{2} m_i^* \omega_0^2 r_i^2 \right) + V_{\rm C} \tag{1}$$

$$V_{\rm C} = \frac{e^2}{4\pi\epsilon} \left(\frac{1}{r_{12}} - \frac{1}{\sqrt{r_{13}^2 + d^2}} - \frac{1}{\sqrt{r_{23}^2 + d^2}} \right) \tag{2}$$

where m_i is the mass of particle i and $m_1 = m_2 = m_{\rm e}^*$ (the effective mass of an electron), $m_3 = m_{\rm h}^*$ (the effective mass of a hole), $\mathbf{r}_i$ ($\mathbf{p}_i$) is the position vector (momentum) of particle i, r_{ij} is the distance between particles i and j, and ω_0 is the strength of the confinement. In this paper, we follow the lines of Que [14] and use the same parabolic frequency for both holes and electrons but quite different masses. The interaction between the three particles is modeled by a Coulomb potential which is screened by a quite phenomenological dielectric constant ϵ. To maintain that the kinetic energy operator is diagonal, we introduce Jacobi coordinates to describe the relative motion. We can choose $\boldsymbol{\rho}_1 = \mathbf{r}_{12}$, the vector from 1 to 2, and $\boldsymbol{\rho}_2 = \mathbf{r}_{12,3}$, the vector from the center of mass of 1 and 2 to particle 3. In this coordinate system, the Schrödinger equation in the center-of-mass frame takes the form

$$\left[-\frac{\hbar^2}{2\mu_1} \nabla_{\rho_1}^2 + \frac{1}{2}\mu_1 \omega_0^2 \rho_1^2 - \frac{\hbar^2}{2\mu_2} \nabla_{\rho_2}^2 + \frac{1}{2}\mu_2 \omega_0^2 \rho_2^2 + V_{\rm C} - E \right] \times \Psi(\boldsymbol{\rho}_1, \boldsymbol{\rho}_2) = 0 \tag{3}$$

where $\mu_1 = m_{\rm e}^*/2$ and $\mu_2 = 2m_{\rm e}^* m_{\rm h}^*/(2m_{\rm e}^* + m_{\rm h}^*)$ are the reduced masses of the pair 1 and 2, and the pair 12 and 3, respectively. This particular set of Jacobi coordinates will be referred to as the α-set. There are two other possible Jacobi coordinates: the β-set where $\boldsymbol{\rho}_1^{(\beta)}$ is the vector from 2 to 3 and $\boldsymbol{\rho}_2^{(\beta)}$ is from the center of mass of 23 to 1; and the γ-set where $\boldsymbol{\rho}_1^{(\gamma)}$ is the vector from 3 to 1 and $\boldsymbol{\rho}_2^{(\gamma)}$ is from the center of mass of 31 to 2. These vectors are depicted in Figure 2. For systems consisting of two identical particles, the β-set and γ-set are equivalent and the wave function should be properly symmetrized.

We also notice that the Schrödinger equation (3) can be written in terms of β-set or γ-set Jacobi coordinates as well, with the corresponding reduced masses μ_1 and μ_2. The superscripts in the coordinates and reduced masses will not be specified unless such a distinction is needed in the discussion.

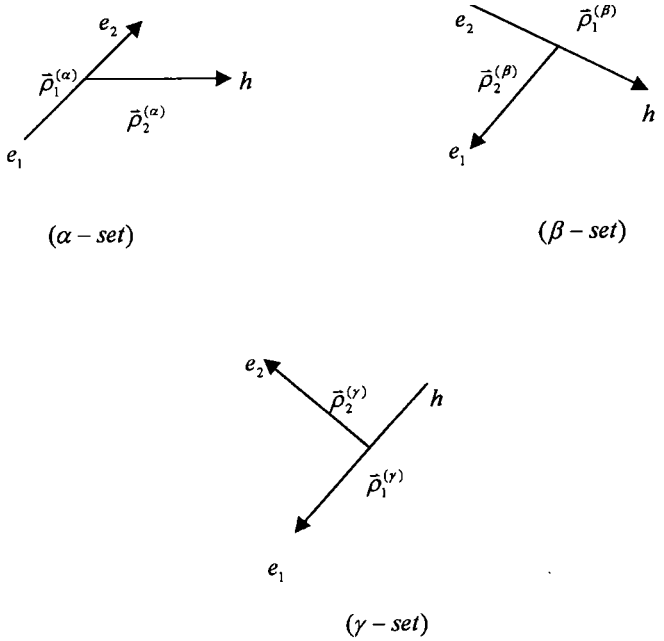

Fig. 2. Three sets of Jacobi coordinates for systems of the negatively charged excitons.

One can introduce mass-weighted hyperspherical coordinates by defining

$$\boldsymbol{\xi}_1 = \sqrt{\mu_1/\mu}\,\boldsymbol{\rho}_1, \quad \boldsymbol{\xi}_2 = \sqrt{\mu_2/\mu}\,\boldsymbol{\rho}_2 \tag{4}$$

where μ is arbitrary (taken to be unity in general). In term of $\boldsymbol{\xi}$'s, the Schrödinger equation (3) now takes the form

$$\left[-\frac{\hbar^2}{2\mu} \nabla_{\xi_1}^2 + \frac{1}{2}\mu\omega_0^2 \xi_1^2 - \frac{\hbar^2}{2\mu} \nabla_{\xi_2}^2 + \frac{1}{2}\mu\omega_0^2 \xi_2^2 + V_C - E \right] \times \Psi(\boldsymbol{\xi}_1, \boldsymbol{\xi}_2) = 0 \tag{5}$$

such that the kinetic energy and the confinement potential operators associated with the two "particles" are identical, and all the mass dependence of the three particles is cast in the rescaling of the distances.

From ξ's, it is straightforward to introduce the hyperspherical coordinates

$$\xi = \sqrt{\xi_1^2 + \xi_2^2}, \quad \tan\phi = \xi_2/\xi_1 \tag{6}$$

where ξ is the hyperradius and $\phi = \phi^{(i)}$ ($i = \alpha, \beta, \gamma$) is the hyperangle. We note that the hyperradius ξ is independent of which set of Jacobi coordinates is used. Thus the two vectors $\boldsymbol{\xi}_1$ and $\boldsymbol{\xi}_2$ are replaced by four coordinates (ξ, Ω), where $\Omega = (\phi, \varphi_1, \varphi_2)$ denotes collectively the five angles, with φ_i being the polar angle of the vector $\boldsymbol{\xi}_i$. Physically, ξ measures the size, while Ω describes the shape and orientation of the system.

With hyperspherical coordinates, the Schrödinger equation (3) is then given by

$$\left[-\frac{\hbar^2}{2\mu}\left(\frac{1}{\xi^3}\frac{\mathrm{d}}{\mathrm{d}\xi}\xi^3\frac{\mathrm{d}}{\mathrm{d}\xi}-\frac{\Lambda^2(\Omega)}{\xi^2}\right)+\frac{C}{\xi}+\frac{1}{2}\mu\omega_0^2\xi^2+E\right]\times\Psi(\xi,\Omega)=0 \quad (7)$$

and

$$\Lambda^2(\Omega)=-\frac{\mathrm{d}^2}{\mathrm{d}\phi^2}+\left(\frac{\sin\phi}{\cos\phi}-\frac{\cos\phi}{\sin\phi}\right)\frac{\mathrm{d}}{\mathrm{d}\phi}+\frac{\ell^2(\varphi_1)}{\cos^2\phi}+\frac{\ell^2(\varphi_2)}{\sin^2\phi} \quad (8)$$

where $\ell^2(\varphi_1)=-\mathrm{i}\partial/\partial\varphi_1$. The grand angular momentum operator $\Lambda^2(\Omega)$ is translationally invariant, *i.e.*,

$$\Lambda^2(\Omega^\alpha)=\Lambda^2(\Omega^\beta)=\Lambda^2(\Omega^\gamma), \quad (9)$$

therefore, the hyperspherical harmonics in different sets of hyperangles are simply the different representations. The eigenvalues and eigenfunctions for the $\Lambda^2(\Omega)$ operator are known [15],

$$\Lambda^2(\Omega)Y_{[K]}(\Omega)=\lambda_{[K]}(\lambda_{[K]}+2)Y_{[K]}(\Omega) \quad (10)$$

$$\lambda_{[K]}=2m+|\ell_1|+|\ell_2| \quad (11)$$

where the analytical expression for $Y_{[k]}(\Omega)$ is given in reference [15], $[K]$ denotes the set of quantum numbers, $[K]=(\ell_1,\ell_2,m)$, with m related to the polynomial functions in angle ϕ, $\ell_1+\ell_2=L$ is the total orbital angular momentum. With coordinate set (α) imposing the particle exchange symmetry, $Y_{[K]}(\Omega^{(\alpha)})$ is straightforward, *i.e.*, ℓ_1 is odd for $S_{12}=1$ and ℓ_1 is even for $S_{12}=0$; with coordinate sets (β) and (γ) imposing the exchange symmetry, it is more complicated [15]. The eigenfunctions in one set can be expanded in terms of eigenfunctions of the other set with $\lambda_{[K]}=\lambda_{[K']}$,

$$Y_{[K]}(\Omega^i)=\sum_{[K']}B_{[K],[K']}Y_{[K]}(\Omega^j) \quad (12)$$

where the expansion coefficients $B_{[K],[K']}$ can be evaluated from

$$B_{[K],[K']}=\int \mathrm{d}\Omega^j Y^*_{[K]}(\Omega^i)Y_{[K]}(\Omega^j). \quad (13)$$

These coefficients are called the transformation bracket and a program for their evaluation has been published [16]. These transformation brackets are useful when evaluating integrals involving functions of different sets of Jacobi coordinates. C/ξ is the total Coulomb interaction potential among the three charged particles, with C given by

$$C=\frac{e^2}{4\pi\epsilon}\left[\sqrt{\mu\mu_1^\alpha}\frac{1}{\cos\phi^\alpha}-\sqrt{\mu\mu_1^\beta}\frac{1}{\sqrt{\cos^2\phi^\beta+d^2}}-\sqrt{\mu\mu_1^\gamma}\frac{1}{\sqrt{\cos^2\phi^\gamma+d^2}}\right]. \quad (14)$$

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

$$\Psi=\sum_{[K]}R_{[K]}(\xi)Y_{[K]}(\Omega). \quad (15)$$

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

$$\left[-\frac{\hbar^2}{2\mu}\left(\frac{1}{\xi^3}\frac{\mathrm{d}}{\mathrm{d}\xi}\xi^3\frac{\mathrm{d}}{\mathrm{d}\xi}-\frac{\lambda_{[K]}(\lambda_{[K]}+2)}{\xi^2}\right)+\frac{1}{2}\mu\omega_0^2\xi^2-E\right]\times R_{[K]}(\xi)-\frac{1}{\xi}\sum_{[K']}U_{[K],[K']}R_{[K']}(\xi)=0 \quad (16)$$

where

$$U_{[K],[K']}=\left\langle Y_{[K]}(\Omega)\left|C\right|Y_{[K']}(\Omega)\right\rangle \quad (17)$$

is the matrix element of the effective charge C evaluated between two hyperspherical harmonics. Equation (16) can be solved to obtain the eigenvalues if convergence can be achieved by using a reasonable truncated set of hyperspherical harmonics. The method has been applied by a number of authors to H^- and He [17,18].

As the two bound electrons of the trion are identical, so a proper picture must include the antisymmetry of the state when the electrons are exchanged. The role of exchange for two electrons is the same as in the textbook example of the He atom. The wave function factorizes into a spatial part and a spin part, which can have either total $S_{12}=0$ (singlet) or $S_{12}=1$ (triplet). The singlet state is antisymmetric under the exchange of spins, so to retain the correct overall antisymmetry of the wave function, the spatial part must be symmetric. The triplet, by contrast, is symmetric, so the corresponding spatial part must be antisymmetric. Hence, the eigenstates are classified by the total spin S_{12} of the two electrons and the total orbital angular momentum L of the trion.

3 Numerical results and discussion

In this paper, effective atomic units are used so that all energies are measured in units of the effective Rydberg Ry^* and all distances are measured in units of the effective Bohr radius a_{B}^*. Using $\sigma=m_{\mathrm{e}}^*/m_{\mathrm{h}}^*=0.196$, *i.e.*, $m_{\mathrm{e}}^*=0.067m_{\mathrm{e}}$ (m_{e} is the free-electron mass), $m_{\mathrm{h}}^*=0.342m_{\mathrm{e}}$, $\epsilon=12.4$ for GaAs, we find $Ry^*=4.955$ meV and $a_{\mathrm{B}}^*=117$ Å. The dimension of the basis is increased until the required accuracy is achieved. The states will be denoted by $^{2S_{12}+1}L$.

The correlation energy of a negatively charged exciton is defined as

$$E^{\mathrm{c}}=E-2E_{\mathrm{e}}-E_{\mathrm{h}}$$

with E the energy level of the negatively charged exciton, and E_{e} and E_{h} the energy levels of the free electron and

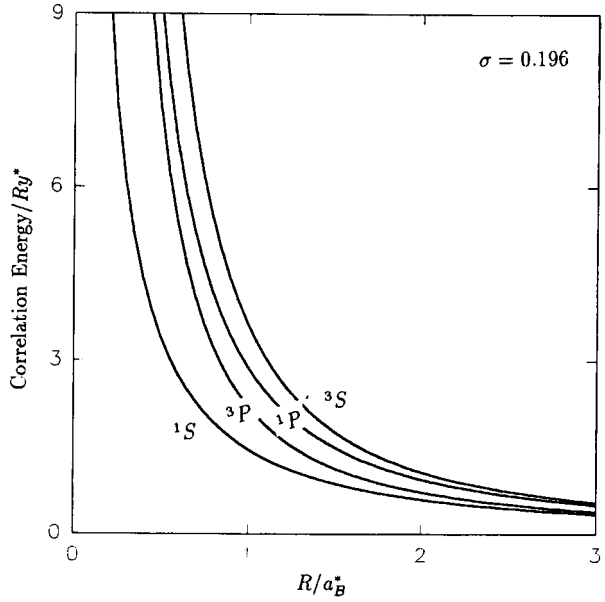

Fig. 3. Variations of the correlation energies of a negatively charged trion X^- in a double-layer quantum dot ($d = 1.0a_{\rm B}^*$) as a function of the dot radius with a fixed value of $\sigma = 0.196$.

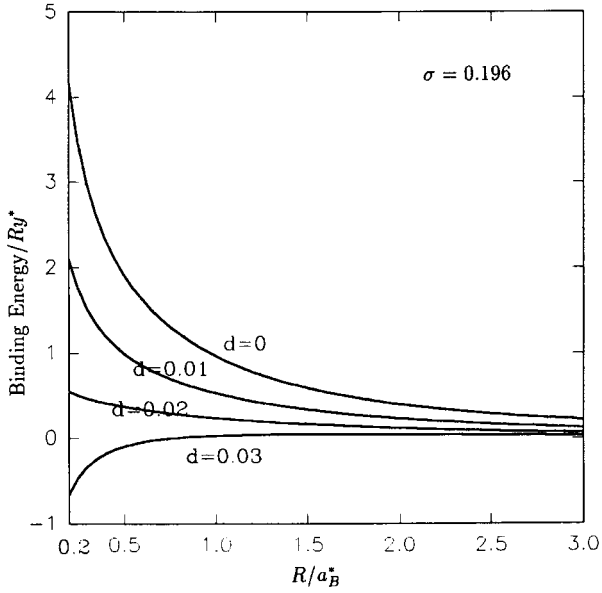

Fig. 4. Dependence of the binding energy W on the QD radius R, normalized by the bulk exciton Bohr radius $a_{\rm B}^*$, is plotted for a few values of the distance d between the double-layer dots with a fixed value of $\sigma = 0.196$.

hole, respectively, in the quantum dot. Thus, $E^{\rm c}$ is the energy due to the Coulomb interaction between the charged particles. In Figure 3, we plot the correlation energies $E^{\rm c}$ of $L = 0$ and $L = 1$ states of the trion as a function of the QD radius R for $\sigma = 0.196$ with $d = 1.0a_{\rm B}^*$. Two different values of the spin are considered. From Figure 3 we can find the following results. (1) The correlation energies decrease when the dot radius increases. Evidently, all three charged particles would prefer to be close to the common axis due to parabolic confinement. However, electrons 1 and 2 in the same layer would not be close to each other. Therefore, as the radius increases, the physical origin of the decrease of correlation energies is related to the decrease of the electron-hole attraction. (2) The ground state is the ^1S state. (3) The second state of the trion is the ^3P state and the energy of the ^3S state is the highest. These results can be easily explained from an analysis of symmetry [19]. Obviously, the equilibrium configuration of the present system is an isosceles triangle (IST) with its height parallel to the common axis called an upstanding IST (or an UIST). Though in quantum mechanics, a system cannot possess a definite geometrical shape as its classical correspondent does, the distribution of the wavefunction of low-lying states should be smoothly (without nodal lines) peaked at the UIST in order to minimize the interacting energy. However, for an UIST configuration a rotation by 180° is equivalent to an interchange of the electrons 1 and 2. This implies that the UIST configuration would be completely prohibited by symmetries unless the values of L takes the numbers fulfilling $(-1)^L = (-1)^{S_{12}}$, *i.e.* $L =$ even if $S_{12} = 0$, and $L =$ odd if $S_{12} = 1$. In the case of $(-1)^L \neq (-1)^{S_{12}}$, an inherent nodal line appears for the UIST, and the binding of the state is spoiled.

The possibility of observing a negatively charged exciton depends on its stability against dissociation into an exciton and a free electron. It may be verified that the corresponding sufficient stability condition reads

$$W = E_X^{\rm c} - E^{\rm c} \geq 0 \tag{18}$$

where W denotes the charged exciton "binding energy", while the $E_X^{\rm c}$ of the exciton is defined by

$$E_X^{\rm c} = E_X - E_{\rm e} - E_{\rm h} \tag{19}$$

where E_X is the exciton ground-state "relative" energy.

The dependence of W on the QD radius R is plotted in Figure 4 for a few values of the distances d with $\sigma = 0.196$. When $d < 0.03a_{\rm B}^*$, the binding energy increases as the radius is reduced. However, when $d \geq 0.03a_{\rm B}^*$, as the dot size is reduced further, the binding energy begins to decrease and eventually becomes negative, *i.e.*, there exists a critical radius $R^{\rm c}$, such that if $R > R^{\rm c}$ ($R < R^{\rm c}$) the configuration of the negatively exciton is stable (unstable). From $W = 0$, we can obtain the value of $R^{\rm c}$, this critical position is dependent of the distance d between the double-layer QD's. On the other hand, we note that the binding energy decreases when the d increases. The reason for this is related to the decrease of the electron-hole attraction with the displacement of the hole from the center of the QD when compared to the constant electron-electron repulsion.

In order to see more clearly the importance of the mass ratio in enhancing the trion binding energy, we plot W as a function of the QD radius R in Figure 5 for several values of the electron-to-hole mass ratio with a fixed value of d ($= 0.02a_{\rm B}^*$). The binding energy increases with decreasing $\sigma = m_{\rm e}^*/m_{\rm h}^*$ and takes a maximum value at the hydrogen

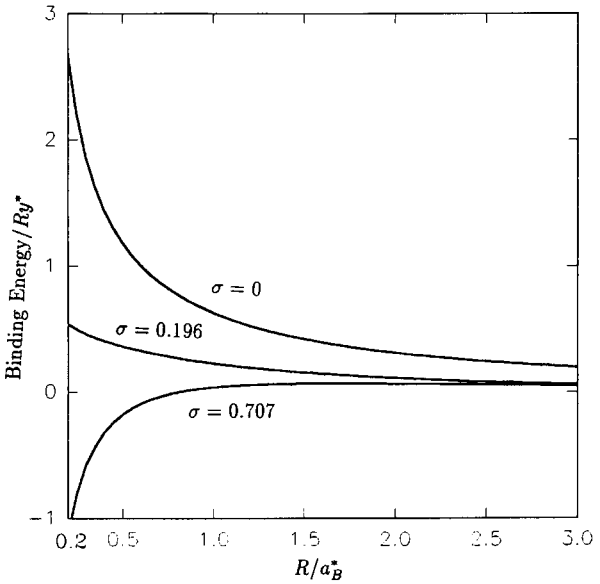

Fig. 5. Dependence of the binding energy W on the QD radius R, normalized by the bulk exciton Bohr radius a_{B}^*, is plotted for a few values of the mass ratio σ with a fixed value of $d = 0.02a_{\mathrm{B}}^*$.

limit ($\sigma = 0$). For a sufficiently large σ, we find that the binding energy has a maximum as a function of R. The binding energy starts to decrease after this maximum, and for sufficiently small R it can even become negative, indicating an unbinding of the X^- state.

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