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Two-dimensional charged-exciton complexes

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A simple method is used to calculate the binding energies of negatively and positively charged excitons (trions) in an ideal two-dimensional electron-hole system. The binding energy of the charged exciton is obtained as a function of the mass ratio $\sigma = m_e/m_h$, using a mass-weighted coordinate system, thus avoiding tedious variational calculations. Our calculated values of the binding energies of charged excitons agree well with existing experimental and theoretical results. The calculations are extended to determine the binding energy of the two-dimensional charged-biexciton complex. [S0163-1829(97)01112-0]

I. INTRODUCTION

Recently there has been considerable experimental interest[1–5] in the charged-exciton states of two-dimensional (2D) electron systems. Both negative and positive charged excitons have been observed and their binding energies have been measured to be in the range of 1 to 2 meV for III-V semiconductor materials. This is mainly the result of the relatively larger binding energies of charged-exciton complexes due to their reduced dimensionality in semiconductor quantum wells.

Theoretical calculations[6,7] of charged excitons of reduced dimensionality have shown good agreement with experimental results. However these calculations are variational and involve intensive computations. The accuracy of the results also depend on the parameters chosen to represent the trial wave functions. In this paper, we adopt a mass-weighted coordinate system to study the properties of a AAB-type Coulombic three-body system, where two of the three particles are similar in mass and carry charges of the same sign. The third particle is assumed to carry a charge of opposite sign. The use of mass-weighted coordinates was first proposed by Lin and Liu[8] in an attempt to present a unified approach for treating nonrelativistic three-body systems in a three-dimensional space. Chen and Lin[9] showed that the total binding energy of an AAB system of charged particles nearly scales with the reduced mass of A and B. However, mass-weighted coordinates have only been applied in the context of a hyperspherical coordinate space system[10] so far.

Our main result is an analytical expression of the binding energy of the charged exciton as a function of the mass ratio $\sigma = m_e/m_h$. Our aim is not to compete with variational calculations such as those of Stebe and Ainane[6], but to avoid tedious computations and to provide a rough estimate of the binding energy of the charged-biexciton complex—the simplest in the series of charged bound multiple-exciton complexes.

This paper is organized as follows. In Sec. II, we provide the theoretical basis used to tranform the Hamiltonian of three charged particles interacting via Coulombic forces and to obtain an analytical expression for the binding energy of the charged exciton. In Sec. III, we extend the method developed in Sec. II, to the case of the charged biexciton. In Sec. IV, we present some numerical results and compare our results with experimental results available in the literature.

II. BINDING ENERGY OF TWO-DIMENSIONAL CHARGED EXCITON

The two possible configurations for the charged exciton are X^- and X^+, for which, respectively, an electron is bound to an exciton and a hole is bound to an exciton. The Hamiltonian for a X^- charged exciton can be written as

$$\hat{H}_{X^-} = -\frac{\hbar^2}{2m_e^*}(\nabla_{e1}^2 + \nabla_{e2}^2) - \frac{\hbar^2}{2m_h^*}\nabla_h^2 + V_{X^-}, \tag{1}$$

where V_{X^-} is the interaction potential given by

$$V_{X^-} = -\frac{e^2}{\epsilon}\left(\frac{1}{|\mathbf{r}_{e1} - \mathbf{r}_h|} + \frac{1}{|\mathbf{r}_{e2} - \mathbf{r}_h|} - \frac{1}{|\mathbf{r}_{e1} - \mathbf{r}_{e2}|}\right), \tag{2}$$

where $\mathbf{r}_i$, $i = e1, e2$, and h, denote the position coordinates of the three charge carriers and ϵ is the dielectric constant of the quantum well material.

We now consider the transformation from space-fixed coordinates to relative coordinates for the three-body system of the charged exciton. This is done by requiring that the relative coordinates and their conjugate momenta satisfy the fundamental commutation relations. We assume that the particles are constrained to move in a plane and define the center of mass position $\overline{\mathbf{R}}$ as

$$\overline{\mathbf{R}} = \frac{m_h^*\mathbf{r}_h + m_e^*\mathbf{r}_{e1} + m_e^*\mathbf{r}_{e2}}{M}, \tag{3}$$

where $M = 2m_e^* + m_h^*$ is the total effective mass of the charged exciton and we define the relative separation $\mathbf{R}$ between the hole and one of the electrons as

$$\mathbf{R} = \mathbf{r}_h - \mathbf{r}_{e1}. \tag{4}$$

The momenta conjugate to $\overline{\mathbf{R}}$ and $\mathbf{R}$ can be easily obtained as

$$\mathbf{P}_{\overline{\mathbf{R}}} = \mathbf{P}_h + \mathbf{P}_{e1} + \mathbf{P}_{e2} \tag{5}$$

and

$$\mathbf{P}_\mathbf{R} = \frac{m_e^*}{m_e^* + m_h^*}\mathbf{P}_h - \frac{m_h^*}{m_e^* + m_h^*}\mathbf{P}_{e1}, \tag{6}$$

where $\mathbf{P}_i$, $i = e1$, $e2$, and h, are the momenta of the three charge carriers.

The third relative coordinate $\mathbf{r}$ is expressed in terms of $\mathbf{r}_h$, $\mathbf{r}_{e1}$, and $\mathbf{r}_{e2}$ and unknown coefficents a_1 and a_2 as

$$\mathbf{r} = a_1\mathbf{r}_h + a_2\mathbf{r}_{e1} + \mathbf{r}_{e2}. \tag{7}$$

Likewise, the momenta of $\mathbf{r}$ can be obtained in terms of the unknown coeffiecients, b_1, b_2, b_3 as

$$\mathbf{P}_\mathbf{r} = b_1\mathbf{P}_h + b_2\mathbf{P}_{e1} + b_3\mathbf{P}_{e2}. \tag{8}$$

The five unknown coefficients are then determined using the commutation relations:

$$[\mathbf{r},\mathbf{P}_\mathbf{r}] = Ih, \quad [\mathbf{R},\mathbf{P}_\mathbf{r}] = 0, \tag{9}$$

$$[\overline{\mathbf{R}},\mathbf{P}_\mathbf{r}] = 0, \quad [\mathbf{r},\mathbf{P}_\mathbf{R}] = 0, \quad [\mathbf{r},\mathbf{P}_{\overline{\mathbf{R}}}] = 0. \tag{10}$$

With the known values of a_1, a_2, b_1, b_2, b_3, and (3)–(8), the Hamiltonian in (1) can be transformed as

$$\hat{H}_{X^-} = H_\text{cm} - \frac{\hbar\nabla_R^2}{2M_R} - \frac{\hbar\nabla_r^2}{2M_r} + V_{X^-}, \tag{11}$$

where M_R is the reduced mass of m_{e1}^* and m_h^* and M_r is the reduced mass of charge carrier, $e2$ relative to the center of mass of charge carriers $e1$ and h:

$$M_r = \frac{(m_{e1}^* + m_h^*)m_{e2}^*}{M}. \tag{12}$$

H_cm in (11) is the Hamiltonian of the kinetic energy of the center of mass motion. This term will be discarded since it does not contribute to the binding energy of the system. It is to be noted that the third relative coordinate r in (7) is obtained as the relative position of the center of mass position of charge carriers, $e1$ and h, with respect to the position of charge carrier $e2$ as shown in Fig. 1.

Using a linear model of the charged exciton (e-h-e), which provides the optimal binding in an exact two-dimensional space, we define a relation between the Jacobi coordinates, $\mathbf{R}$ and $\mathbf{r}$ as

$$\mathbf{r} = \sqrt{1 + \frac{M_R}{M_r}}\,\mathbf{R}. \tag{13}$$

It is to be noted that (13) is used within the context of a two-dimensional mass-weighted coordinate system. It establishes a crucial relationship between the two possible modes of relative motion in a two-dimensional three-body system. As such, the final results of the exciton binding energy are strongly dependent on the form of (13).

Using (13) and the relation $[\mathbf{r},\mathbf{P}_\mathbf{r}] = [\mathbf{r},\alpha\mathbf{P}_\mathbf{R}] = Ih$, where α is a scaling factor, the Hamiltonian in (11) can be simplified as

$$\hat{H}_{X^-} = -\frac{\hbar\nabla_R^2}{2M_R}\left[1 + \frac{M_R}{M_r}\frac{1}{1 + \dfrac{M_R}{M_r}}\right] + V_{X^-}. \tag{14}$$

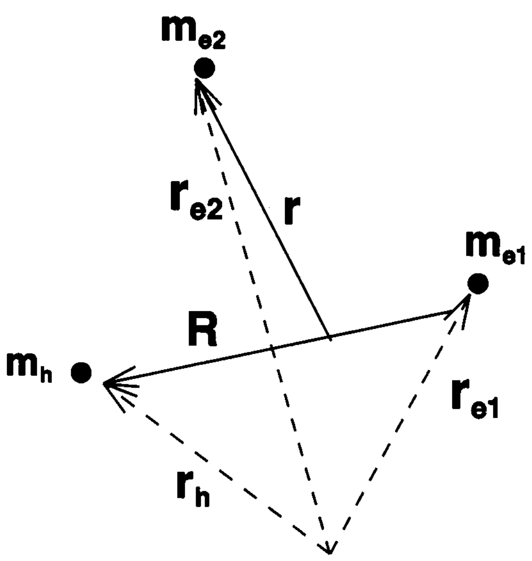

FIG. 1. Relative coordinates of a three particle system. $\mathbf{r}$ originates from the center of mass of m_{e1} and m_h.

The above Hamiltonian can then be rewritten as

$$\hat{H}_{X^-} = -\frac{\hbar^2}{2M_{X^-}}\nabla_R^2 - \frac{e^2}{\epsilon_{X^-}}\frac{1}{R}, \tag{15}$$

where

$$M_{X^-} = \left(1 + \frac{2\sigma + 1}{\sigma^2 + 4\sigma + 2}\right)^{-1} M_R \quad \text{and} \quad \epsilon_{X^-} = \frac{2}{3}\epsilon, \tag{16}$$

where $\sigma = m_e^*/m_h^*$.

The energy eigenvalue of an isolated exciton, E_ex, can be expressed as[11,12]

$$E_\text{ex} = E_g - Eb_\text{ex}, \quad Eb_\text{ex} = \frac{\gamma M_R R_H}{\epsilon^2 m_e}, \tag{17}$$

where Eb_ex denotes the binding energy of the exciton. Likewise the energy eigenvalue, E_{X^-}, of the two-dimensional charged-exciton Hamiltonian in (15) can be obtained as

$$E_{X^-} = 2E_g - \frac{\gamma M_{X^-} R_H}{\epsilon_{X^-}^2 m_e}, \tag{18}$$

where γ is a measure of dimensionality, E_g is the energy band gap, R_H is the Rydberg constant, and m_e the electronic rest mass.

The binding energy, Eb_{X^-}, of a negatively charged exciton can be written[6] as

$$Eb_{X^-} = E_g + E_\text{ex} - E_{X^-}. \tag{19}$$

Substituting (18) and (17) in (19), we get

$$Eb_{X^-} = \left(\frac{M_{X^-}}{M_R}\frac{\epsilon^2}{\epsilon_{X^-}^2} - 1\right)Eb_\text{ex}. \tag{20}$$

Using (16) in (20), we get the ratio of the binding energy of a negatively charged exciton to that of a three-dimensional neutral donor as

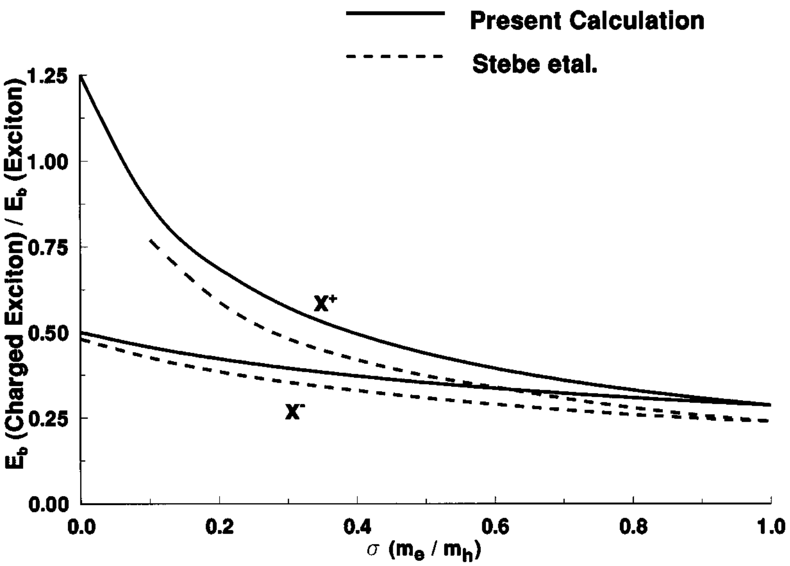

FIG. 2. Ratio of binding energies, Eb_{X^-}/D_N and Eb_{X^+}/D_N, as functions of mass ratio, σ. The dashed lines refer to results of variational calculations of Stebe and Ainane (Ref. 6).

$$\frac{Eb_{X^-}}{D_N}=\left[\left(\frac{3}{2}\right)^2\left(1+\frac{2\sigma+1}{\sigma^2+4\sigma+2}\right)^{-1}-1\right]\frac{1}{1+\sigma}, \qquad (21)$$

where $D_N = m_e^* R_H/\epsilon^2 m_e$ is the binding energy of the neutral donor.

In the hydrogenic limit, $\sigma = 0$, Eq. (21) gives $Eb_{X^-}/D_N = 0.5$, which agrees very well with the numerical result of about 0.48.[6] In the positronium limit, $\sigma = 1$, $Eb_{X^-}/B = 0.2875$, which compares well with the value of 0.25 obtained from the variational calculations of Stebe and Ainane[6]. The binding energy of a positively charged exciton (h-e-h) (Eb_{X^-}) can be obtained from Eq. (21) using

$$Eb_{X^+}(\sigma)=\frac{Eb_{X^-}\left(\frac{1}{\sigma}\right)}{\sigma}. \qquad (22)$$

In Fig. 2, we have compared our results of Eb_{X^-}/B and Eb_{X^+}/B as functions of σ with that of Stebe and Ainane.[6]

III. TWO-DIMENSIONAL CHARGED-BIEXCITON COMPLEX

Among three-electron atomic systems the H_3^+ ion, which is barely bound, is perhaps the most perculiar ion. Theoretical calculations[13] have shown that the equilibrium H_3^+ configuration is an equilateral triangle with interatomic distances comparable to that in the H_2 molecule. The semiconductor analogs of the H_3^+ ion are the charged multiple-exciton states, X_3^- and X_3^+, for which, respectively, an electron is bound to a single biexciton[14–16] and a hole is bound to a single biexciton. It is difficult to describe a structure for the charged biexciton, as it is equally possible for the unpaired electron to be bound to two excitons which interact weakly, and thus less likely to be existing in a biexcitonic state. A possible configuration of the X_3^+ state is shown in Fig. 3, which can be considered as an average structure arising out of the individual resonance structure of an extra hole interacting with a biexciton. Binding can be viewed as resulting from the quantum mechanical sharing of two electrons among the three holes. A similar structure can be conceptualized for the X_3^- complex. The exotic complex structure as shown in Fig. 3 is convenient in extending the theory established in Sec. II to the case of the charged-biexciton complex.

We use the definition of the binding energy, $Eb_{X_3^-}$, of a negatively charged biexciton as

$$Eb_{X^-}=E_g+E_{2\mathrm{ex}}-E_{X_3^-}, \qquad (23)$$

where $E_{2\mathrm{ex}}$ and $E_{X_3^-}$ are, respectively, the energy eigenvalues of the biexciton and charged-biexciton Hamiltonian.

It has been shown[17,18] recently that the biexciton has an anisotropic character in two-dimensional systems and reduces to the hydrogenic form of a single exciton, with a modified reduced mass and dielectric constant, in exact 2D systems. Using this result, we rewrite (23) as

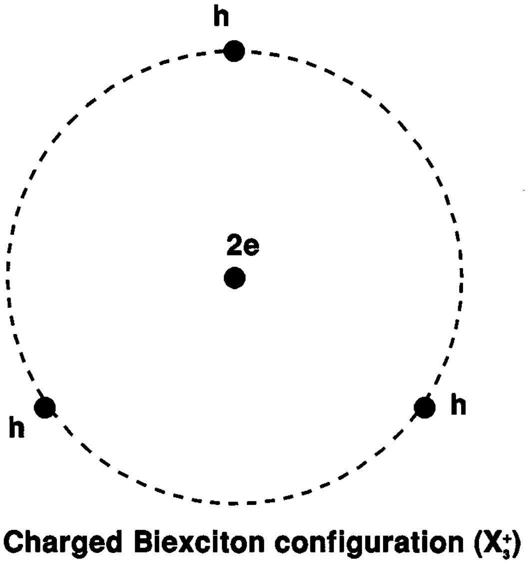

FIG. 3. Schematic representation of the charged-biexciton complex, X_3^+.

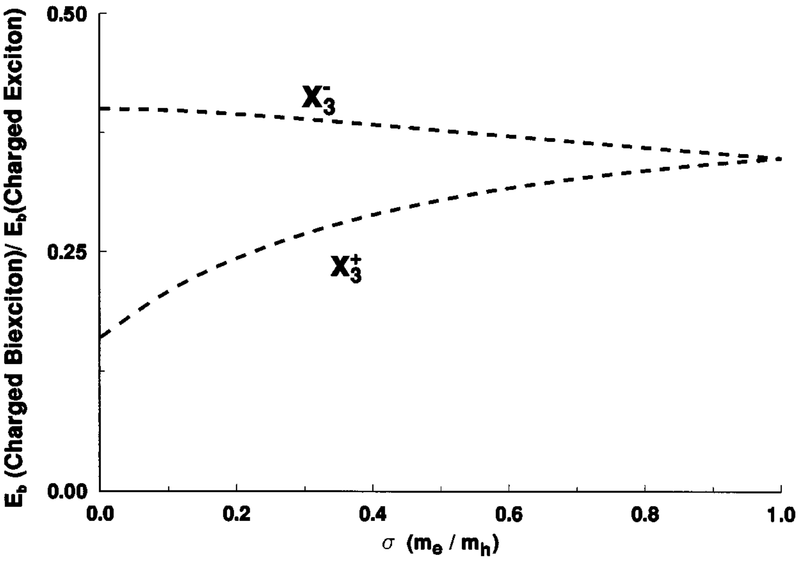

FIG. 4. Ratio of binding energies, $Eb_{X_3^-}/Eb_{X^-}$ and $Eb_{X_3^+}/Eb_{X^+}$ as functions of mass ratio, σ.

$$Eb_{X_3^-}=\left(\frac{M_{X_3^-}}{M_{R'}}\frac{\epsilon_{2\mathrm{ex}}^2}{\epsilon_{X_3^-}^2}-1\right)Eb_{2\mathrm{ex}},\tag{24}$$

where $Eb_{2\mathrm{ex}}$ is the binding energy of the biexciton, $M_{R'}$ and $M_{X_3^-}$ are the reduced masses of the biexciton and charged biexciton, respectively. $\epsilon_{2\mathrm{ex}}$ denotes the dielectric constant of the biexciton while $\epsilon_{X_3^-}$ denotes that of the charged biexciton.

Using the experimental[19,20] value of ≈ 0.2 for the ratio of the binding energy of a biexciton to that of an exciton, which is shown to be independent of the width of the quantum well,[18] and (21) and (24), we get the upper bound of the ratio of the binding energy of the charged exciton to charged biexciton as

$$\frac{Eb_{X_3^-}}{Eb_{X^-}}=\frac{0.2}{\left[\left(\frac{3}{2}\right)^2\left(1+\frac{2\sigma+1}{\sigma^2+4\sigma+2}\right)^{-1}-1\right]}.\tag{25}$$

When $\sigma=0$, Eq. (25) gives $Eb_{X_3^-}/Eb_{X^-}=0.4$, and when $\sigma=1$, $Eb_{X_3^-}/Eb_{X^-}=0.35$. The binding energy of a positively charged biexciton (h-h-e-e-h-h) (X_3^+) is obtained from Eq. (25) by replacing σ with $1/\sigma$. Figure 4 shows results of $Eb_{X_3^-}/Eb_{X^-}$ and $Eb_{X_3^+}/Eb_{X^+}$ as functions of σ.

IV. RESULTS AND DISCUSSION

We have used a method of determining the binding energy of two-dimensional charged-exciton complexes without performing any tedious numerical calculations. Though this approach is based on a two-dimensional space, the results obtained here may also be applied to quantum wells of small well widths. Charged exciton and biexciton complexes have large diameters,[21] and hence have limited degrees of freedom in thin quantum wells.

It may be desirable to compare our calculated values with recent experimental results. Binding energy values of 1.2 to 2 meV have been reported[1,3,4,22] for the charged exciton. The binding energy of the positively charged exciton has been measured[1] as 1.3 meV. Taking the binding energy of the neutral donor, D_N, as 5.5 meV, for GaAs, Eb_{X^-}/D_N and Eb_{X^+}/D_N are expected to be in the range of 0.22–0.36 for the GaAs/Al$_x$Ga$_{1-x}$As system. These experimental values are consistent with our calculated results of 0.32 and 0.37 for the X^- and X^+ charged excitons, respectively, at $\sigma=0.68$. It is important to note that the ratio in (25) is independent of γ which is related[12] to the quantum well width. This is due to the assumption of an ideal two-dimensional limit for the electron and hole wave functions in an infinite well potential. As is well known in the theory of finite quantum wells, an exact two-dimensional structure of exciton is never attained. Due to the effect of the spreading of the electron and hole wave function into the barrier regions, the diameter of excitons decrease as the quantum well thickness decreases and pass through a minimum point[23] before increasing again. Thus we expect a similar qualitative result in the ratio of our calculated binding energies as a function of the well width, when applied to finite wells.

It is to be noted that precise quantitative calculations of the ratio of binding energies as a function of the well width is difficult. Apart from the numerical difficulties linked to an accurate calculation of the exciton binding energy at fixed well widths, there is a lack of knowledge of a suitable relation of the form given in (13) as a function of the well width. However, it is likely that solving the existing problem within the framework of a fractional dimensional space[11] may circumvent some of the difficulties mentioned here.

We are unable to obtain experimental values of the charged biexciton at this moment in time to compare with Fig. 4 which shows that the binding energies of $Eb_{X_3^-}$ and $Eb_{X_3^+}$ range from 1.25 meV to 0.6 meV for $0 \leqslant \sigma \leqslant 1$. Thus the charged-biexciton complex is stable for any value of the mass ratio σ, in the range from 0 to 1. It is interesting to note that the ratio, $Eb_{X_3^-}/Eb_{X^-}$ is a monotonic decreasing function of σ, whereas $Eb_{X_3^+}/Eb_{X^+}$ is a monotonic increasing function of σ.

It is to be noted that in the limit $\sigma=0$, the hole mass is relatively heavy and the ground state of the charged-exciton complex becomes comparatively high. When the electron and hole masses are equal, $\sigma \sim 1$, the zero-point motion of holes in the exciton complexes can decrease the binding between the charges. This may explain the decrease of the binding energies with σ in Figs. 2 and 4. However, the role of the mass of the hole can only partly explain the change in binding with σ between the various charges forming the exciton complexes. The spin structure[24,25] of the exciton complex may be a crucial factor in explaining the optimization of binding in charged-exciton complexes. In this work, we have ignored the role of spins in exciton, in order to obtain analytical expressions for binding energies. Another assumption worth mentioning is the neglect of correlation terms[26] arising out of electron-electron and electron-hole interactions. The inclusion of such terms is expected to yield higher values for the binding energies of the charged-exciton complexes. Though this has not been significant in the case of the charged exciton, we expect our numerical values calculated using (25) to vary by some factors (10–30 %) from those calculated with the inclusion of electron-electron and electron-hole interactions.

In conclusion, we have presented in this paper, the binding energies of two-dimensional charged-exciton complexes. It is expected that our results may be important in the quantitative understanding of future experimental work involving charged-exciton complexes.

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