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The mass dependence of the ground-state properties of the Wannier exciton in a quantum box

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Abstract. A simple variational study of the ground-state properties of an electron–hole system confined to a quantum box is presented. A two-parameter trial wavefunction which is asymmetric with respect to the coordinates of the two particles is introduced to account for both the localization of the heavy particle and the correlation between the two particles. The mass dependence of the ground-state energy and the average particle separation is discussed.

1. Introduction

Exciton dynamics in quantum-well (QW) structures have been intensively studied in recent years, both experimentally and theoretically [1]. These QW structures are systems of reduced dimensionality with properties different from bulk semiconductor properties; electrons in these systems are free to move in a plane, but motion perpendicular to the plane is confined by a depletion potential. Experimentally, the quantum confinement effect of Wannier excitons in QW structures is most directly observed as the high-energy shift of the interband absorption or luminescence peak as the size decreases. These size-dependent modifications of the optical and electronic properties suggest the potential applicability to the ultrafast non-linear optical devices, which seems to have motivated some of the recent investigations. There are at present several classes of new photonic devices which are all based on the excitonic properties of semiconductor QW structures. These include modulators [2], interferometers [3], self-electrooptic effect devices [4] and switches [3].

With the recent advances in the art of microfabrication, quantum microstructures can now be fabricated, that exhibit quantum carrier confinement in two dimensions (quantum well wires [5, 6]) and in all three dimensions (quantum boxes [7, 8] and microcrystallites [9, 10]). These structures provide new systems for the study of quantum confinement effects. The size quantization effect is due to the competition between the attractive two-body Coulomb force and the confining force at the boundary. When the characteristic length of the microstructure is large, the electron–hole pair will have the properties of a weakly confined exciton with effective Bohr radius a_B. As the size of the system decreases and becomes comparable to a_B, the electron and the hole will primarily be confined as individual particles with little spatial correlation [11–13]. In general, the properties of a Wannier exciton is strongly dependent on the shape or the dimensionality of the quantum microstructures [14]. Kayanuma recently studied the shape dependence of the quantum size

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effect using the variational approach [15]. He considered an exciton confined to a cylinder of variable radius and length. By varying the radius and length he could model a zero-, one-, two- and three-dimensional geometry. It is observed that quantum confinement effects are most severe in quantum boxes that are confined in all three dimensions. However, the computed excitonic binding energy depends only on the reduced mass of the exciton because of the symmetric form of the trial wavefunction under the exchange of the coordinates of the electron and hole. Clearly, in the cases where the electron and hole masses differ greatly, the symmetrical *ansatz* will no longer be adequate, and a more complicated trial wavefunction should be used instead. As shown in previous studies of Wannier excitons in quantum dots [16, 17], using a trial wavefunction with different parameters for the electron and hole will lead to an asymmetry in the optimal values of the variational parameters. This is due to the fact that the heavier particle is more localized and cannot approach the boundary since it should remain closer to the centre-of-mass. Thus, the effect of the confining wall has the biggest impact on the lighter particle.

In the present paper we study the ground-state properties of an electron–hole system confined to a quantum box using an asymmetrical trial wavefunction with respect to the electron and hole coordinates. In particular, we investigate the mass dependence of the ground-state properties for a fixed reduced mass. We carry out a variational calculation in the spirit of previous calculations on quantum dots [11, 16, 17] using a trial wavefunction with only two parameters. In this approach, one parameter describes the correlation between the two particles whereas the other parameter builds in the degree of localization of the heavier particle. Thus, this asymmetrical wavefunction has the ability to capture the mass dependence of the ground-state properties. In the transition regime the mass dependence becomes most transparent. It is found that the heavier the total mass is, the lower the energy and the smaller the average distance between the two particles will be. This dependence is important in quantum boxes constructed from some semiconductor materials like GaAs and AlGaAs, where the hole mass can be much larger than the electron mass [18]. Similar effects have also been studied in type-II quantum structures [19].

2. Model

Microfabricated quantum boxes are constructed from narrow two-dimensional quantum wells by processing the wells to laterally confine the two-dimensional motion. Typically, the width w of the two-dimensional quantum well is an order of magnitude less than the length L of the side of the box, so the box is a thin plate or disk. We shall here model the quantum box as a square plate with sides of length L and width w. The effective-mass Hamiltonian for an electron and a hole in the quantum box is given by

$$H = \sum_{i=\mathrm{e,h}} \left\{ -\frac{\hbar^2}{2m_{\parallel,i}} \left(\frac{\partial^2}{\partial x_i^2} + \frac{\partial^2}{\partial y_i^2} \right) - \frac{\hbar^2}{2m_{\perp,i}} \frac{\partial^2}{\partial z_i^2} + V_i \right\} - \frac{e^2}{\epsilon |\boldsymbol{r}_\mathrm{e} - \boldsymbol{r}_\mathrm{h}|} \tag{1}$$

where

$$V_i = \begin{cases} 0 & \text{if } |x_i|, |y_i| < L/2 \text{ and } |z_i| < w/2 \\ \infty & \text{otherwise.} \end{cases} \tag{2}$$

Here, for simplicity, we have assumed the effective masses in the plane to be isotropic and an infinite confining potential. The exciton variational ground-state wavefunction Ψ is chosen to be of the following form

$$\Psi(\boldsymbol{r}_\mathrm{e}, \boldsymbol{r}_\mathrm{h}) = \mathcal{N} \phi_\mathrm{e}(\boldsymbol{r}_\mathrm{e}) \phi_\mathrm{h}(\boldsymbol{r}_\mathrm{h}) s(\boldsymbol{r}_\mathrm{e}, \boldsymbol{r}_\mathrm{h}) \tag{3}$$

where

$$\begin{aligned}
\phi_e(r_e) &= \cos(kx_e)\cos(ky_e)\cos(qz_e)\exp(-\eta_e r_e^2)\\
\phi_h(r_h) &= \cos(kx_h)\cos(ky_h)\cos(qz_h)\exp(-\eta_h r_h^2)\\
s(r_e, r_h) &= \sum_n c_n \exp\{-\alpha_n[(x_e - x_h)^2 + (y_e - y_h)^2]\}
\end{aligned} \tag{4}$$

with $k = \pi/L$ and $q = \pi/w$. Here $\mathcal{N}$ is a normalization constant determined by requiring the norm of Ψ (with proper measure) to be one. The parameters η_e and η_h describe the localization of the electron and hole, respectively. Following Bryant [13], we have (for computational convenience) written the correlation function $s(r_e, r_h)$ as a linear combination of Gaussian functions, with the parameters α_n regulating the strength of the correlation between the electron and the hole. Accurate energies are obtained by use of 5–10 Gaussians. Since our objective in this study is a basic understanding of the mass dependence of the ground-state properties of the system, we shall, for the sake of simplicity, confine ourselves to only one Gaussian. The corresponding wavefunction is therefore less accurate, but still good enough to capture the essential physics. Furthermore, it should be noted that no correlation of z_e and z_h has been included in the trial wavefunction because typical well widths w are so small that in the z-direction the electron and the hole behave like individual particles with little spatial correlation.

According to the variational principle the best estimate for the ground-state energy is obtained by minimizing $E = \langle H \rangle$ with respect to the variational parameters. After some algebra we can write the energy in the following form

$$E = \int_{-L/2}^{L/2} dx_e \int_{-L/2}^{L/2} dy_e \int_{-w/2}^{w/2} dz_e \int_{-L/2}^{L/2} dx_h \int_{-L/2}^{L/2} dy_h \int_{-w/2}^{w/2} dz_h |\Psi|^2 \varepsilon(r_e, r_h) \tag{5}$$

with

$$\varepsilon(r_e, r_h) = \sum_{i=e,h} \left\{ \frac{\hbar^2(k_i^2 + 2\eta_i + 2\alpha)}{m_{\parallel,i}} + \frac{\hbar^2 q^2}{2m_{\perp,i}} - \frac{\hbar^2}{2m_{\parallel,i}}\Big(F_i(x_i) + F_i(y_i)\Big)\right\} - \frac{e^2}{\epsilon |r_e - r_h|} \tag{6}$$

and

$$F_i(x_i) = 4k\tan(kx_i)(\eta_i x_i \pm \alpha(x_e - x_h)) + 4\eta_i^2 x_i^2 \pm 8\eta_i \alpha x_i (x_e - x_h) + 4\alpha^2 (x_e - x_h)^2 \tag{7}$$

where the 'minus' sign holds for $i = \mathrm{e}$ (the electron coordinate), and the 'plus' sign holds for $i = \mathrm{h}$ (the hole coordinate). (Note, to derive the energy of the trial wavefunction the following formulae are useful:

$$\begin{aligned}
\partial\Psi/\partial x_i &= -[k\tan(kx_i) + 2\eta_i x_i \pm 2\alpha(x_e - x_h)]\Psi\\
\partial\Psi/\partial y_i &= -[k\tan(ky_i) + 2\eta_i y_i \pm 2\alpha(y_e - y_h)]\Psi\\
\partial\Psi/\partial z_i &= -q\tan(qz_i)\Psi
\end{aligned} \tag{8}$$

where the same sign convention holds.) It is clear that for a general box of size $L \times L \times w$, to evaluate the energy E one needs to perform a six-dimensional integration, and this, of course, constitutes a formidable numerical task if one is to use an adequate grid spacing. Thus, in the following we shall confine ourselves to the most relevant situation for which $w \ll L$. In that case the integral in the z-direction drops out. In other words, we model our quantum box to be effectively of *zero* thickness, i.e. $w = 0$, and neglect the variations in the z-direction. As a result, the energy $E(\alpha, \eta_i)$ reduces to the form

$$E(\alpha, \eta_i) = \int_{-L/2}^{L/2} \mathrm{d}x_\mathrm{e} \int_{-L/2}^{L/2} \mathrm{d}y_\mathrm{e} \int_{-L/2}^{L/2} \mathrm{d}x_\mathrm{h} \int_{-L/2}^{L/2} \mathrm{d}y_\mathrm{h} \, |\Psi|^2 \varepsilon(\alpha, \eta_i) \tag{9}$$

where $\varepsilon(\alpha, \eta_i)$ can be written as

$$\varepsilon(\alpha, \eta_i) = \sum_{i=\mathrm{e,h}} \left\{ \frac{\hbar^2(k^2 + 2\eta_i + 2\alpha)}{m_{\parallel,i}} - \frac{\hbar^2}{2m_{\parallel,i}} \Big(F_i(x_i) + F_i(y_i) \Big) \right\} - \frac{e^2}{\epsilon |\boldsymbol{r}_\mathrm{e} - \boldsymbol{r}_\mathrm{h}|} \tag{10}$$

and the functions $F_i(x_i)$ are defined in equation (7). Hence, we are then left with a four-dimensional integral over the electron- and hole-coordinates in the x- and y-direction.

To simplify the problem further, we shall choose $\eta_\mathrm{e} \equiv 0$ (or equivalently $\exp(-\eta_\mathrm{e} r_\mathrm{e}^2) = 1$) and let $\eta_\mathrm{h} \equiv \eta$. Thus, the wavefunction Ψ is reduced to a two-parameter trial wavefunction, and the energy depends only on the two parameters η and α: $E = E(\alpha, \eta)$. These two variational parameters η and α incorporate the localization of the heavy hole and the correlation between the electron and the hole, respectively. Finally, the variational principle requires the energy E to be minimized with respect to the two variational parameters η and α to obtain the best estimate for the ground-state energy. Although our variational *ansatz* is quite simple, it is able to capture the most relevant physics of the confined electron–hole system in which the hole is much heavier than the electron.

3. Results and discussion

For the units of length and energy we shall take the effective Bohr radius $a_\mathrm{B} = \epsilon\hbar^2/\mu e^2$ and the effective Rydberg energy Ryd $= \mu e^4/2\epsilon^2\hbar^2$, respectively. Also, we shall let m_1 be the mass of the heavier hole and m_2 the electron mass. In figure 1 we show the minimum energy plotted against the size of the system for mass ratios $\sigma \equiv m_1/m_2 = 1$ and 10. The overall behaviour is independent of the mass ratio and shows the familiar quantum size effect; a transition at around $L/a_\mathrm{B} \approx 2 - 3$ between a regime of exciton confinement (for large L) and a regime of individual particle confinement (for small L). We have also calculated the electron–hole separation r:

$$r = \langle (x_\mathrm{e} - x_\mathrm{h})^2 + (y_\mathrm{e} - y_\mathrm{h})^2 \rangle^{1/2}. \tag{11}$$

In figure 2 we have plotted r against L for the same set of mass ratios as in figure 1. Again, the overall behaviour is familiar and r goes linearly to zero as the system size decreases.

It is instructive to study how the variational parameters α and η vary with the system size as well. In figure 3 we show α/a_B^{-2} for the mass ratios $\sigma = 1$ and 10. When the length L is decreased from a very large value $L \gg a_\mathrm{B}$, α decreases, and the correlation between the electron and the hole increases. However, as the system reaches a size of $L \simeq 2.5a_\mathrm{B}$, α increases rapidly, and the interparticle correlation decreases. For small systems the electron and hole are confined as individual particles with little spatial correlation. For instance, the characteristic correlation length is $1/\sqrt{2\alpha} = 0.22a_\mathrm{B}$ for $L = 0.1a_\mathrm{B}$; in other words, the Gaussian-type correlation function s is essentially equal to unity within the quantum box. Also, note that there is a slight mass dependence of α in the transition region. In figure 4 we show how η changes with the size of the quantum box. In the case of equal masses η is equal to zero. For mass ratio $\sigma = 10$ we see a monotonic decrease of η to the limiting value of zero as L increases. The decay of η is most rapid for $0 < L \leqslant 3a_\mathrm{B}$.

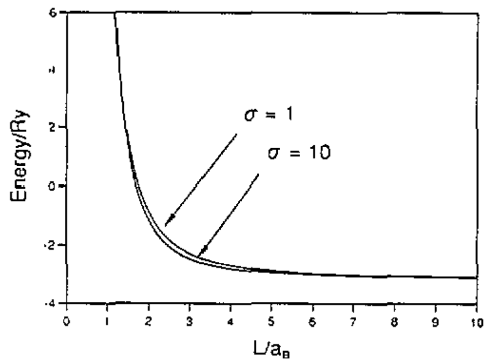

Figure 1. Size dependence of the ground-state energy for mass ratios $\sigma = m_1/m_2 = 1$ and 10.

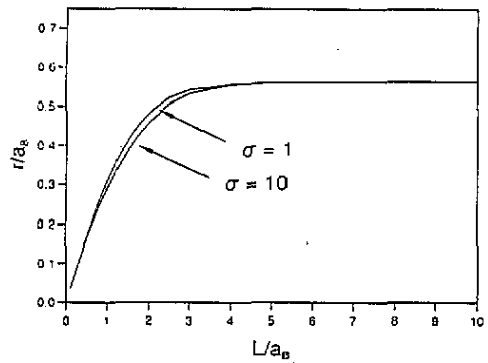

Figure 2. Size dependence of the electron–hole separation r for the same set of mass ratios as in figure 1.

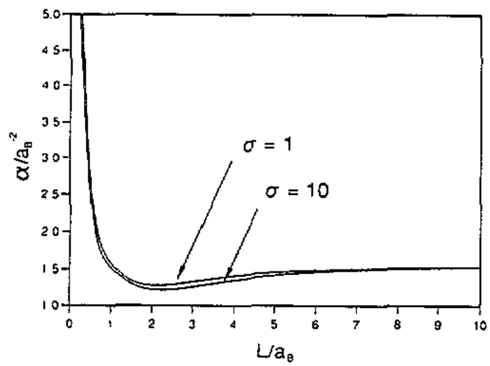

Figure 3. Plot of $\alpha/a_{\mathrm{B}}^{-2}$ versus length L for mass ratios $\sigma = 1$ and 10.

So far we have concentrated on the quantum size effects and the overall L-dependence. However, one can also see a dependence on the mass ratio between the particles, especially in the transition region $L/a_{\mathrm{B}} \approx 2-3$. Here we choose the value $L/a_{\mathrm{B}} = 2.5$ for our study, but qualitatively the mass dependence will not be much different for other values of L in this region. In figure 5 we show the ground-state energy plotted against the mass ratio σ. As the mass ratio increases, the energy first decreases and then flattens out. This behaviour is mainly attributed to the decrease of kinetic energy of the heavy particle. In figure 6 we

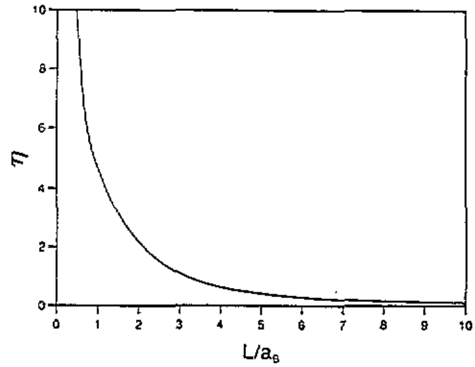

Figure 4. Variational parameter η versus L. The curve is for the mass ratio $\sigma = 10$. For equal masses η is identically zero.

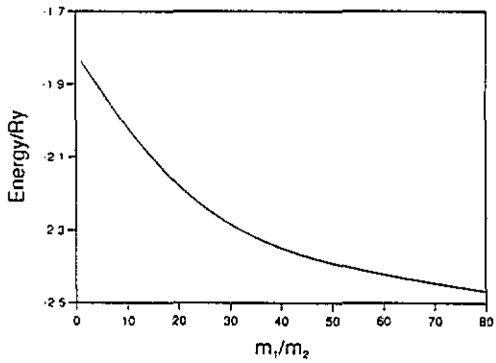

Figure 5. Minimum energy versus mass ratio for length $L/a_B = 2.5$.

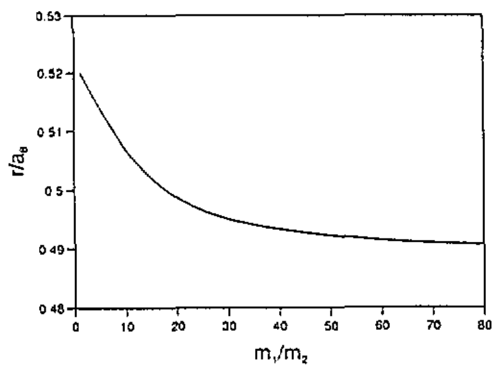

Figure 6. The electron–hole separation r versus mass ratio for the same length as in figure 5.

plot r versus mass ratio. For large mass ratios the average distance becomes smaller. This is intuitively correct since the heavy particle is more localized and thereby exerts a more singular attractive force.

We have also plotted the mass dependence of the variational parameters upon the mass ratio. In figures 7 and 8 we show η and α as a function of σ, respectively. The variational parameter η in figure 7 describes the localization of the heavy hole. Since the characteristic length of localization of the hole is given by $1/\sqrt{2\eta}$, a large value of η means a large degree

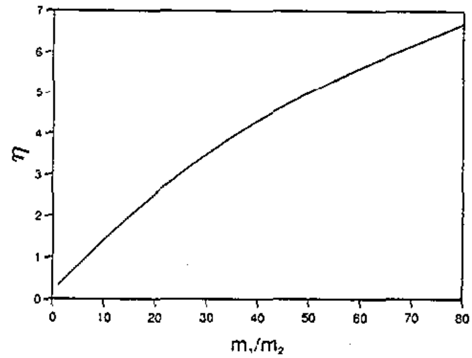

Figure 7. Variational parameter η versus mass ratio, for the same length as in figure 5.

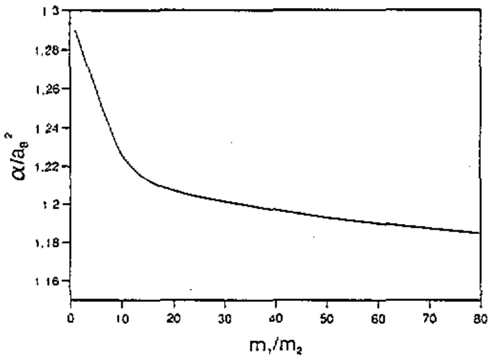

Figure 8. Mass dependence of the variational parameter α/a_B^{-2} for the same length as in figure 5.

of localization of the hole for a given L. From figure 7 we can see how this localization depends upon the mass of the heavy hole. When σ increases, η increases, and the hole is strongly localized. In figure 8 we show the variational parameter α which governs the correlation between the electron and hole. It is clear that α is not as sensitive to the mass ratios as η since the Coulomb interaction is independent of the mass. In the transition region there is only a slight mass dependence of α as a result of localization of charge associated with the heavy hole.

In summary we have studied the mass dependence of the ground-state energy and the average particle separation for an electron–hole system confined to a quantum box. By using a simple two-parameter trial wavefunction we could explain the behaviour as an effect of the localization of the heavy particle. In this variational approach one parameter describes the correlation between the two particles whereas the other parameter builds in the degree of localization of the heavier particle. In the present model the thickness of the quantum box is assumed to be small. A quantum box of finite thickness will be of interest for future studies and should lead to modifications of the properties.

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