PHYSICAL REVIEW B VOLUME 36, NUMBER 11 15 OCTOBER 1987-I

Excitons and biexcitons in semiconductor quantum wires

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(Received 2 April 1987)

The exciton and biexciton ground-state binding energies are calculated for semiconductor quantum wires of radius R smaller than the bulk exciton radius a_0 assuming an infinite confining potential. Both the excitonic and biexcitonic (molecular) binding energies are enhanced by a factor greater than 5 for GaAs/$\mathrm{Ga}_{1-x}\mathrm{Al}_x\mathrm{As}$ quantum wires of radius approximately $a_0/2$. The simultaneous shrinking of the exciton size with the wire radius is shown to reduce the contribution (when compared with the quasi-two-dimensional case) of dielectric polarization effects which arise when the wire is embedded in a cladding with a lower dielectric constant.

I. INTRODUCTION

Following the success of semiconductor quantum-well structures in permitting the study of quasi-two-dimensional phenomena[1] there is a growing experimental[2] and theoretical[3,4] interest in similar quasi-one-dimensional structures. The Coulomb potential in one dimension has some pathological features[5] (e.g., the ground-state energy becomes infinite) so it is interesting to see how this affects the spectra of states in very slender semiconductor wires—so-called "quantum-well wires" (QWW's). Recently, Bryant[4] published a thorough study of the shallow impurity states in such QWW's, showing the strong increase of the ionization energy for small radii. Simple variational approaches for the excitonic binding energies in QWW's have been reported recently by several authors.[6]

In this paper we study the exciton and biexciton ground-state binding energies in such QWW's. We shall consider the electrons and holes to be confined within the wire for wire radii smaller than the bulk excitonic radius, a_0. The problem is reduced to a one-dimensional Schrödinger equation with an effective interaction between the charged particles which depends on the wire radius. The second section is devoted to the derivation of this effective one-dimensional potential accounting for the dielectric boundary conditions. In the next section we present a good analytic approximation to this potential (amounting to a simple regularization of the Coulomb interaction)[5] which simplifies the calculation in the biexcitonic case. Finally, in the fourth section the biexciton (excitonic molecule) is treated in the Heitler-London scheme[7] for the hole-hole potential and the resulting Schrödinger equation is solved numerically. Together with the large exciton binding energies for small radii we find relatively high molecular binding energies. However, unlike the exciton binding energy the biexciton energy does not blow up as the wire radius decreases to zero.

II. THE ONE-DIMENSIONAL EFFECTIVE COULOMB POTENTIAL

We shall consider here the ideal quantum confinement of a particle within an infinite circular potential of radius R. The ground-state wave function in such a well is

$$\phi_0(\rho)=\frac{J_0(\alpha_0\rho/R)}{\sqrt{\pi}RJ_1(\alpha_0)}\ , \tag{1}$$

where $\alpha_0=2.405$ is the first zero of the Bessel function[8] J_0 and ρ the radial coordinate. The corresponding ground-state energy is $E_0=\alpha_0^2\hbar^2/2\mu R^2$, where μ is the appropriate particle mass.

In the QWW's considered here we have charged electrons and holes which will interact via Coulomb forces. If the cladding medium outside the wire has a different dielectric constant ϵ_2 than that within the wire ϵ_1, the Coulomb interaction will be distorted. To compute this dielectric polarization effect we must find the associated variation in the electrostatic energy. As the first step towards this goal, we first calculate the potential $V(\mathbf{r},\mathbf{r}')$ at a point $\mathbf{r}$ in such a wire due to a charge e at $\mathbf{r}'$. This is achieved by solving the appropriate Poisson equations, i.e.,

$$\left.\begin{matrix}\epsilon_1\\ \epsilon_2\end{matrix}\right\}\times\nabla^2V(\mathbf{r},\mathbf{r}')=\begin{cases}-4\pi e\delta(\mathbf{r}-\mathbf{r}') & \text{for } |\rho|<R\ ,\\ 0 \text{ for } |\rho|>R\ ,\end{cases} \tag{2}$$

together with the boundary conditions

$$\epsilon_1\left.\frac{\partial V}{\partial\rho}\right|_{\rho=R-}=\epsilon_2\left.\frac{\partial V}{\partial\rho}\right|_{\rho=R+}\ ,$$

$$V\,|_{\rho=R-}=V\,|_{\rho=R+}\ ,$$

$$V\,|_{\rho\to\infty}\to0 \quad\text{and}\quad V\,|_{\rho=0}<\infty\ .$$

with cylindrical coordinates (ρ,θ,z). These equations may be solved using a partial-wave expansion yielding the potential inside the wire as

$$V(\mathbf{r},\mathbf{r}')=-\frac{e}{\epsilon_1}\frac{1}{|\mathbf{r}-\mathbf{r}'|}-\frac{2e}{\pi}\left[\frac{1}{\epsilon_2}-\frac{1}{\epsilon_1}\right]\sum_{m=-\infty}^{\infty}\int_0^{\infty}dk\,\cos[k(z-z')]e^{im(\theta-\theta')}C_m(kR)I_m(k\rho)I_m(k\rho')\ , \tag{3}$$

where

$$C_m(kR)=\frac{K_m(kR)K_m'(kR)}{I_m(kR)K_m'(kR)-(\epsilon_1/\epsilon_2)I_m'(kR)K_m(kR)} .$$

Here I_m, K_m are the modified Bessel functions.[8]

The electrostatic energy W for an electron-hole pair at positions $\mathbf{r}_e=(z_e,\rho_e,\theta_e)$ and $\mathbf{r}_h=(z_h,\rho_h,\theta_h)$ is defined as

$$W=\tfrac{1}{2}\int \rho(\mathbf{r})[V(\mathbf{r},\mathbf{r}_h)-V(\mathbf{r},\mathbf{r}_e)]d\mathbf{r} , \tag{4}$$

where

$$\rho(\mathbf{r})=e\delta(\mathbf{r}-\mathbf{r}_e)-e\delta(\mathbf{r}-\mathbf{r}_h) .$$

Due to the electrostatic self-energy this quantity is not well defined (i.e., infinite); however, the variation δW between the $R=\infty$ value and that for finite R is finite and gives the change in the energy due to the introduction of the dielectric boundaries. Hence,

$$\delta W=\frac{2e^2}{\pi}\left[\frac{1}{\epsilon_2}-\frac{1}{\epsilon_1}\right]\sum_{m=-\infty}^{\infty}\int_0^{\infty}dk\, C_m(kR)\{I_m^2(k\rho_e)+I_m^2(k\rho_h)-2\cos[k(z_e-z_h)]\cos[m(\theta_e-\theta_h)]I_m(k\rho_e)I_m(k\rho_h)\}. \tag{5}$$

It may be seen that as expected there is no contribution to the dielectric polarization when $\mathbf{r}_e=\mathbf{r}_h$, as the charges neutralize one another. To this energy we must add the usual Coulomb interaction energy

$$-\frac{e^2}{\epsilon_1}\frac{1}{|\mathbf{r}_e-\mathbf{r}_h|} . \tag{6}$$

By restricting the motion in the transverse plane via the quantum confinement the motion along the z axis will be governed by an effective interaction energy

$$U(z_e-z_h)=e\int_0^R\rho_e d\rho_e\int_0^R\rho_h d\rho_h\int_0^{2\pi}d\theta_e\int_0^{2\pi}d\theta_h V(\mathbf{r}_e,\mathbf{r}_h)\,|\phi_0(\rho_e)|^2\;|\phi_0(\rho_h)|^2 \tag{7}$$

apart from a constant energy shift

$$\Delta=4e^2\left[\frac{1}{\epsilon_2}-\frac{1}{\epsilon_1}\right]\int_0^{\infty}dk\int_0^R d\rho\sum_{m=-\infty}^{\infty}C_m(kR)\,|\phi_0(\rho)|^2 I_m^2(k\rho) , \tag{8}$$

which should be added to the confinement energy shift E_0. In Ref. 4 the shallow impurity case was considered, with the charged impurity fixed on the cylinder axis and with the averaging performed only over the electronic motion. More explicitly, we have, after a change of integration variable,

$$\begin{aligned}U(Z)=-\frac{e^2}{\epsilon_1 R\pi^2 J_1^4(\alpha_0)}\int_0^1\rho_e d\rho_e\int_0^1\rho_h d\rho_h J_0(\alpha_0\rho_e)J_0(\alpha_0\rho_h)\\ \times\left[\frac{2}{\pi}\left[\frac{\epsilon_1}{\epsilon_2}-1\right]\int_0^{\infty}\cos(kZ)C_0(k)I_0(k\rho_e)I_0(k\rho_h)dk\right.\\ \left.+\int_0^{2\pi}d\theta_e\int_0^{2\pi}d\theta_h\frac{1}{[Z^2/R^2+(\rho_e\cos\theta_e-\rho_h\cos\theta_h)^2+(\rho_e\sin\theta_e-\rho_h\sin\theta_h)^2]^{1/2}}\right] ,\end{aligned} \tag{9}$$

where $Z=(z_e-z_h)$.

It may be seen from Eq. (1) that this potential, apart from the factor $e^2/\epsilon_1 R$ in front is a universal function of Z/R, i.e., it depends only on the ratio of the electron-hole separation and the wire radius and not on each individually. We denote this universal potential as $\bar{U}(Z)=R\epsilon_1 U(RZ)/e^2$. This universal feature is convenient as it allows us to calculate the potential $\bar{U}(Z)$ once and then apply it for any value of the wire radius simply by scaling the Z coordinate accordingly. We have accordingly evaluated $\bar{U}(Z)$ and show the results in Fig. 1, for $\epsilon_1=\epsilon_2$ and $\epsilon_1=1.3\epsilon_2$, the latter corresponding to the extreme case of $x=1$ of a GaAs/Ga$_{1-x}$Al$_x$As QWW. The dashed curve is a pure $1/|Z|$ potential plotted for comparison. The important feature to note is that the effective one-dimensional potential no longer diverges at $Z=0$, attaining a value of $\simeq -3$. For $Z\gg 1$ the ratio of the two curves is naturally ϵ_1/ϵ_2 (here 1.3), as at separations much larger than the wire radius one should have a simple Coulomb potential with most of the force lines going through the cladding medium. On the other hand, for $Z=0$ the ratio of the two potentials

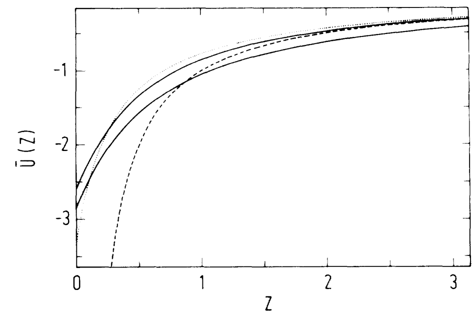

FIG. 1. $\bar{U}(Z)$ as a function of electron-hole separation Z for two different dielectric constant ratios: $\epsilon_1=\epsilon_2$ for the upper solid curve and $\epsilon_1=1.3\epsilon_2$ for the lower solid curve. The dashed curve is a Coulomb potential plotted for comparison, and the dotted line is the regularized Coulomb potential for $\alpha=0.3$.

is 1.08—much less than ϵ_1/ϵ_2.

For $\epsilon_1=1.3\epsilon_2$ the numerical calculation gave an energy shift $\Delta=0.27E_R a_0/R$.

We have solved numerically the corresponding Schrödinger equation for the electron-hole relative motion:

$$\left[-\frac{\hbar^2}{2m}\frac{\partial^2}{\partial Z^2}+U(Z)-\varepsilon\right]\psi_x=0\ , \tag{10}$$

where

$$\frac{1}{m}=\frac{1}{m_e}+\frac{1}{m_h}\ .$$

The binding energies for the two values of ϵ_1/ϵ_2 considered above are shown in Fig. 2 [with R scaled to the bulk excitonic Bohr radius $a_0=\hbar^2\epsilon_1/me^2$ and the binding energy $(-\epsilon)$ scaled to the bulk excitonic Rydberg energy $E_R=\hbar^2/2ma_0^2$]. It can be seen that the binding energy diverges as $R\rightarrow 0$, exceeding the limiting two-dimensional ground-state binding energy of $4E_R$ for $R<a_0/2$ which corresponds to the two-dimensional exciton Bohr radius.

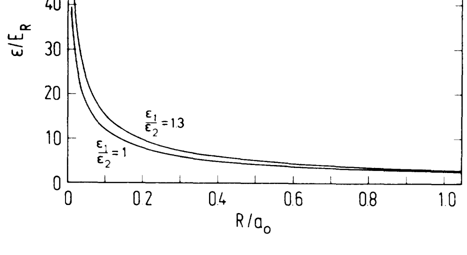

FIG. 2. Excitonic binding energy as a function of wire radius for two different dielectric constant ratios: $\epsilon_1=\epsilon_2$ for the lower curve and $\epsilon_1=1.3\epsilon_2$ for the upper curve.

In quantum wires it can be seen that the effect of the dielectric polarization on the binding energy is small in comparison to the corresponding two-dimensional case, where the ratio of the binding energies should be $(\epsilon_1/\epsilon_2)^2$, a factor of 1.69 compared with our enhancement of $\simeq 1.2$. This is attributable to the differing behavior in the limit of small thicknesses. For two-dimensional systems the wave-function width tends to a constant $a_0/2$ while in the QWW case the wave-function width collapses to zero.[5] Since the materials currently being proposed for the manufacture of QWW's have dielectric constant ratios under the 1.3 case considered here, we shall concentrate now exclusively on the case $\epsilon_1=\epsilon_2$.

For very small radii one must return to a bulk problem in the cladding material a limit not present within our model as we assumed an infinite cylindrical potential for the wire. For GaAs/Ga$_x$Al$_{1-x}$As QWW's the estimated minimal radius below which such effects should be taken into account is about $R=a_0/2$. This estimate is based on potential depths of $1.06x$ eV for electrons and $0.187x$ eV for holes.[1] The results of Bryant,[4] who dealt with the similar problem of shallow impurities in QWW's and accounted for this finite well depth, suggest that the excitonic binding energy will continue to increase for radii beneath this value before the bulk effect sets in. In other materials this limitation may be less stringent.

III. ANALYTIC APPROXIMATIONS

In the treatment of the excitonic molecule (see the next section) it is useful to have an analytic approximation to the potential energy $U(Z)$ as well as for the excitonic wave functions. Loudon[5] has considered analytically the bound-state problem for a regularized Coulomb potential $1/(|Z|+\alpha)$ instead of $1/|Z|$. The wave functions are then given in terms of the Whittaker function[8]

$$\psi_x(|Z|)=W_{\lambda,1/2}(2(|Z|+\alpha R)/\lambda a_0)\ , \tag{11}$$

where

$$\lambda=\frac{1}{\sqrt{|\varepsilon|}}\ .$$

The energy eigenvalues for the symmetrical states, which include the ground state, are to be determined from the transcendental equation for ε:

$$\left.\frac{d\psi_x(Z)}{dZ}\right|_{Z=0}=0\ . \tag{12}$$

For the ground state this equation has the asymptotic solution[5] as $R\rightarrow 0$

$$\frac{R}{a_0}\simeq\frac{1}{2\alpha\sqrt{|\varepsilon|}}e^{-\sqrt{|\varepsilon|}/2}\ . \tag{13}$$

One sees that as $R\rightarrow 0$ the ground-state binding energy diverges and from the asymptotic expansion of the Whittaker function[8] one may see that the normalized, squared wave function tends to a δ function in this limit. All the

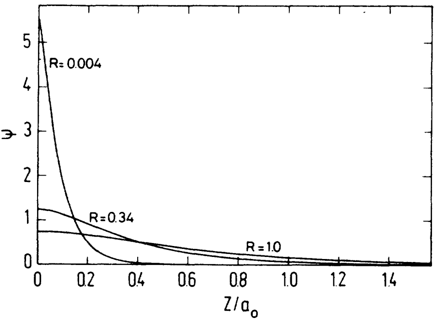

FIG. 3. Excitonic wave functions for various wire radii.

excited states become pairwise degenerate; however, their eigenvalues remain finite in this limit. The reader is referred to Loudon[5] for more details.

The integral representation of the Whittaker function[8]

$$W_{\lambda,1/2}(x)=\frac{e^{-x/2}}{\Gamma(1-\lambda)}\int_0^\infty e^{-t}\left[\frac{x}{t}+1\right]^\lambda dt \tag{14}$$

was used in our calculations. Some typical normalized, squared excitonic wave functions are given in Fig. 3 for various radii showing the shrinking of the wave function for small radii. The binding energies calculated using this approximation for $\alpha=0.3$ were so close to those of the $\epsilon_1=\epsilon_2$ curve in Fig. 2 that they could not be distinguished on the same graph. The corresponding analytic potential is shown as the dotted curve in Fig. 1. We have used the corresponding analytical wave functions and energies for the biexciton calculations in the next section.

IV. THE BIEXCITON

Consideration of the biexciton problem in QWW's leads one to the one-dimensional effective Hamiltonian,

$$\begin{aligned}H=&-\frac{\hbar^2}{2m_e}\left[\frac{\partial^2}{\partial z_{e_1}^2}+\frac{\partial^2}{\partial z_{e_2}^2}\right]-\frac{\hbar^2}{2m_h}\left[\frac{\partial^2}{\partial z_{h_1}^2}+\frac{\partial^2}{\partial z_{h_2}^2}\right]\\&+U(z_{e_1}-z_{e_2})+U(z_{h_1}-z_{h_2})\\&-U(z_{e_1}-z_{h_1})-U(z_{e_2}-z_{h_2})\\&-U(z_{e_1}-z_{h_2})-U(z_{e_2}-z_{h_1})\end{aligned} \tag{15}$$

or in the new coordinates

$$y_1=z_{e_1}-z_{h_1},\quad y_2=z_{e_2}-z_{h_2},$$

$$x=z_{h_1}-z_{h_2},\quad X=\frac{m_e(z_{e_1}+z_{e_2})+m_h(z_{h_1}+z_{h_2})}{2M},$$

with

$$M=m_e+m_h .$$

In units of E_R and a_0 the Hamiltonian is

$$\begin{aligned}H=&-\frac{\partial^2}{\partial y_1^2}-\frac{\partial^2}{\partial y_2^2}+\frac{2m}{m_h}\left[\frac{\partial}{\partial y_1}-\frac{\partial}{\partial y_2}\right]\frac{\partial}{\partial x}\\&-\frac{2m}{m_h}\frac{\partial^2}{\partial x^2}-\frac{m}{2M}\frac{\partial^2}{\partial X^2}+2u ,\end{aligned} \tag{16}$$

where the potential u is given by

$$\begin{aligned}u=&U(y_1-y_2+x)+U(x)-U(y_1)-U(y_2)\\&-U(y_1+x)-U(y_2-x)\end{aligned}$$

with the effective one-dimensional Coulomb potential

$$U(z)=\frac{1}{|z|+\alpha R/a_0} .$$

We wish to solve for the ground-state biexcitonic binding energy, and we look for a solution of the form[9]

$$\Psi(y_1,y_2,x)=\psi_b(|x|)\Phi(y_1,y_2,x) , \tag{17}$$

which consists of the biexciton wave function $\psi_b(|x|)$ for the relative motion of the two holes and $\phi(y_1,y_2,x)$, which describes the exciton correlations:

$$\Phi(y_1,y_2,x)=\frac{1}{S(x)}\phi(y_1,y_2,x)$$

with the normalization function

$$S^2(x)=\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}dy_1dy_2\phi^2(y_1,y_2,x) .$$

For ϕ we use the Heitler-London[7] approximation, i.e.,

$$\phi(y_1,y_2,x)=\psi_x(y_1)\psi_x(y_2)+\psi_x(y_1+x)\psi_x(y_2-x) . \tag{18}$$

Each term describes a product of exciton wave functions, ψ_x, one exciton formed by e_1,h_1 and the other by e_2,h_2 in the first term and by e_2,h_1 and e_1,h_2 in the second term. The wave function Ψ, for an even ψ_b, is invariant under the permutations of electrons, holes, and a common reflection of both. As usual the ground state is assumed to be a singlet spin state for both electrons and holes, and therefore the antisymmetry required by the exclusion principle is contained in the spin part of the wave function. Within the Heitler-London approximation that we use in this first calculation no polarization of one exciton by the other is accounted for. In three dimensions this polarization can be easily included variationally due to the simple exponential nature of the exciton wave functions, and leads to a significant enhancement of the molecular binding energy. Indeed, for equal electron and hole masses such a polarization is necessary to obtain a bound state.[9] In the one-dimensional problem due to the complicated nature of the exciton wave functions an analogous scheme is computationally much more involved. Nevertheless, we will show that even without polarization one obtains a strong binding for all mass ratios in the one-dimensional case.

The separation of the normalization integral $S(x)$ enables one to write[9] a Schrödinger equation for the relative hole-hole wave function ψ_b,

$$\left[-\frac{2m}{m_h}\frac{\partial^2}{\partial x^2}+v(x)-\varepsilon_b\right]\psi_b=0 , \tag{19}$$

with the effective-hole potential

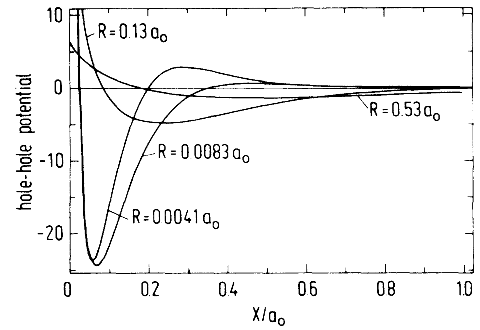

FIG. 4. Hole-hole potentials for various wire radii for $m_e/m_h=0.48$.

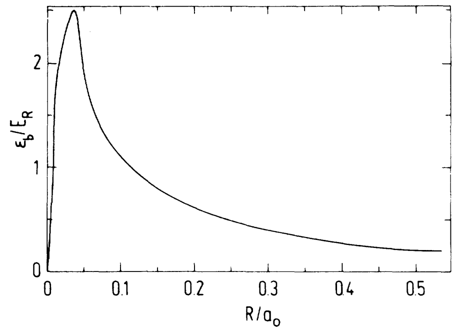

FIG. 5. Biexcitonic binding energy as a function of wire radius for $m_e/m_h=0.48$.

$$
\begin{aligned}
v(x) &= -2\varepsilon+\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\Phi(y_1,y_2,x)H\Phi(y_1,y_2,x)dy_1dy_2 \\
&= -2\varepsilon+\frac{2m}{m_h}\frac{1}{S(x)}\frac{\partial^2 S(x)}{\partial x^2}+\frac{1}{S^2(x)}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\phi(y_1,y_2,x)H\phi(y_1,y_2,x)dy_1dy_2 \; . \qquad (20)
\end{aligned}
$$

Here ε is the ground state excitonic energy and the molecular binding energy $(-\varepsilon_b)$ is defined as the difference between the energy of the system and the energy of two infinitely separated excitons therefore $v(\infty)=0$.

In Fig. 4 we illustrate this effective hole-hole potential with an electron-hole mass ratio of 0.48 for various wire radii. This mass ratio corresponds to that used in the bulk GaAs. In fact quantum confinement effects lift the valence-band degeneracy giving a heavy and light hole with mass ratios of 0.335 and 0.67, respectively. We see that the potential becomes deeper and narrower for smaller radii until a maximum depth is reached at $R\simeq 0.083$, after which the depth decreases. In fact $v(x)\rightarrow 0$ as $R\rightarrow 0$ for $x\neq 0$ while $v(0)\rightarrow\infty$ for $R\rightarrow 0$. In other words, at any finite hole separation, as the excitons shrink there will eventually be no overlap of their wave functions, while if the holes are already at the same point before the excitons shrink to a point then the Coulomb repulsion will be infinite.

The corresponding Schrödinger equation for these potentials was solved numerically and the molecular binding energy is given as a function of wire radius in Fig. 5. One may see that the binding energy has a maximum at $R=0.03a_0$ and vanishes for $R=0$ as the potential is zero everywhere except $x=0$. Notwithstanding this down turn for small radii, the actual value of the exciton binding energies are relatively large—at $R<0.5a_0$ they exceed $0.2E_R$, more than a factor of 5 larger than the bulk molecular binding energy[9] of $\simeq 0.04E_R$. As they stand these energies are roughly the same as those obtained in two-dimensional quantum wells;[10] however, one should realize that our calculations attained such large binding energies without the inclusion of the excitonic polarization and deformation, effects which will certainly enhance them further.

Figure 6 shows the squares of the normalized ground-state wave functions corresponding to the potentials given in Fig. 4. An interesting connection to the bulk is the electron-hole mass ratio dependence of the molecular binding energy. This dependence is given in Fig. 7 with the binding energy scaled to the coresonding exciton

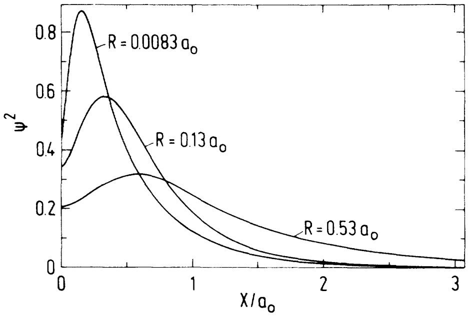

FIG. 6. Biexciton hole-hole wave functions for various wire radii for $m_e/m_h=0.48$.

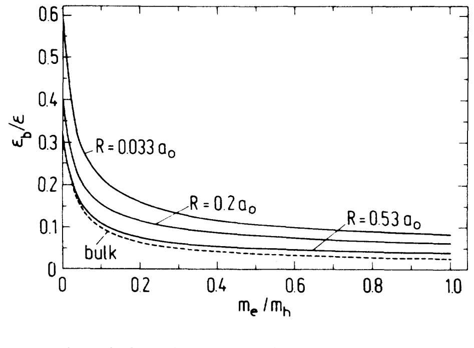

FIG. 7. Ratio of the biexciton binding energy to the exciton binding energy as a function of the electron-hole mass ratio. The equivalent bulk values are given by the dashed curve (after Ref. 9).

binding energy for three different wire radii. Note that (in these relative units) the curves are qualitatively the same as the bulk curve as given by the dashed curve,[9] indeed our curve for $R = 0.53$ coincides almost exactly with the bulk values.

Our results for the excitonic and biexcitonic spectra in QWW's show that for small wire radii one may obtain a strong enhancement of the binding energies. The optical properties of QWW's will therefore exhibit large excitonic and biexcitonic nonlinearities even at room temperature. The question of whether or not condensation into plasma clusters will occur remains an open one.

ACKNOWLEDGMENTS

L.B. wishes to thank J. Weidmann, U. Scharffenberger, and F. Constantinescu for useful discussions pertaining to the mathematical aspects of the problem. I.G. acknowledges receipt of European Community Grant No. STI-0168-D(CD). This work was supported by the Deutsche Forschungsgemeinschaft through the Sonderforschungsbereich 185 Frankfurt-Darmstadt.

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