Calculation of the exciton binding energies in type-II GaAs/AlAs quantum-well structures: Application of the perturbation-variational expansion method

Spiros V. Branis and K. K. Bajaj
Department of Physics, Emory University, Atlanta, Georgia 30322
(Received 3 April 1991; revised manuscript received 4 November 1991)

We report a calculation of the binding energies of both the heavy-hole and the light-hole excitons in type-II GaAs/AlAs quantum wells as a function of the size of the AlAs layer (or GaAs layer). We use a perturbation-variational expansion method to the first order in perturbation expansion. The exciton binding energies are calculated assuming infinite potential barriers and results thus obtained are compared with those of a published variational calculation. We find that the values of the exciton binding energies that we calculate are somewhat lower than those obtained using a variational calculation, even for small wells similar to the perturbation-variational method results in type-I quantum wells. An explanation of this behavior in terms of the average spatial separation of electrons and holes is given.

The study of the excitonic properties in quantum-well (QW) structures has drawn considerable interest in recent years. Most of the work has been reported in the type-I QW structures, where an electron and a hole are confined spatially in the same well. For narrower well sizes [typically GaAs-layer thickness < 30 Å (Refs. 1 and 2)], the band-edge configuration at the GaAs-AlAs heterojunction may become "staggered" or type-II. In this case, an electron and a hole are confined in spatially separate wells (see Fig. 1). There have been a few studies in type-II QW systems. Duggan and Ralph[3] have calculated the exciton binding energies variationally in the type-II configuration using the infinite potential barrier model. Matsuura and Shinozuka[4] studied the same problem using the variational approach by incorporating slightly different trial wave functions. Salmasi and Bauer[5] studied the electron-hole exchange interaction in type-II QW's, while van Kesteren *et al.*[6] considered the order of the X conduction-band valleys in type-II GaAs/AlAs QW's. Recently, Degani and Farias[7] have calculated the exciton binding energy in type-II quantum wells in the presence of a static electric field using a variational approach.

In this Brief Report, we report a calculation of the binding energies of both the heavy-hole and light-hole excitons in type-II GaAs/AlAs QW's as a function of the size of the AlAs layer (or the GaAs layer). We use a perturbation-variational expansion method to the first order of perturbation expansion in our calculations. This approach was used by Mei and Lee[8] and Lee, Mei and Liu[9] in the study of the impurity states in anisotropic crystals, by Lee and Lin[10] for the study of Wannier excitons in a thin crystal film, and later by Jiang[11] and Ekenberg and Altarelli[12] in the calculations of the exciton binding energies in type-I GaAs/Al_xGa_{1-x}As QW's. These authors included only terms up to first order in perturbation expansion in their calculations. We calculate exciton binding energies assuming infinite potential barriers and compare our results with those of a variational calculation.[3]

The band-edge configuration for the type-II system and the corresponding coordinate system are shown in Fig. 1. The holes are confined in the GaAs layer while the electrons reside in the indirect-band-gap AlAs layer. Assuming perfect confinement for both electrons and holes (infinite-potential-barrier model), we replace the different dielectric constants of GaAs and AlAs materials with their average static dielectric constants $\epsilon_0=12.3$. This eliminates any image-charge corrections. Under the effective-mass approximation, and neglecting the off-diagonal terms of the Kohn-Luttinger Hamiltonian,[13] the total Hamiltonian of the exciton associated with either the heavy-hole or the light-hole band can be expressed as (in cylindrical polar coordinates)[14]

$$H=-\frac{\hbar^2}{2\mu_\pm}\left[\frac{1}{\rho}\frac{\partial}{\partial\rho}\rho\frac{\partial}{\partial\rho}+\frac{1}{\rho^2}\frac{\partial^2}{\partial\phi^2}\right]-\frac{\hbar^2}{2(m_e)_l}\frac{\partial^2}{\partial z_e^2}-\frac{\hbar^2}{2(m_\pm)_l}\frac{\partial^2}{\partial z_h^2}-\frac{e^2}{\epsilon_0[\rho^2+(z_e-z_h)]^{1/2}}+V_e(z_e)+V_h(z_h)\ , \tag{1}$$

where $(m_e)_l$ and $(m_\pm)_l$ are the effective masses of electron and (heavy or light) holes, respectively, in the direction of growth [the transverse masses are $(m_e)_t$ and $(m_\pm)_t$]. All the other mass values can be expressed in terms of Luttinger parameters[13] γ_1 and γ_2 as in Ref. 15:

$$\frac{1}{(m_\pm)_l}=\frac{1}{m_0}(\gamma_1\mp2\gamma_2)\ ,\qquad \frac{1}{(m_\pm)_t}=\frac{1}{m_0}(\gamma_1\pm\gamma_2)\ ,\qquad \frac{1}{\mu_\pm}=\frac{1}{(m_e)_t}+\frac{1}{(m_\pm)_t}\ , \tag{2}$$

where m_0 is the free-electron mass and $\mu_\pm$ is the transverse reduced mass of the electron-hole pair in the x-y plane. The potential wells for electron $V_e(z_e)$ and for the

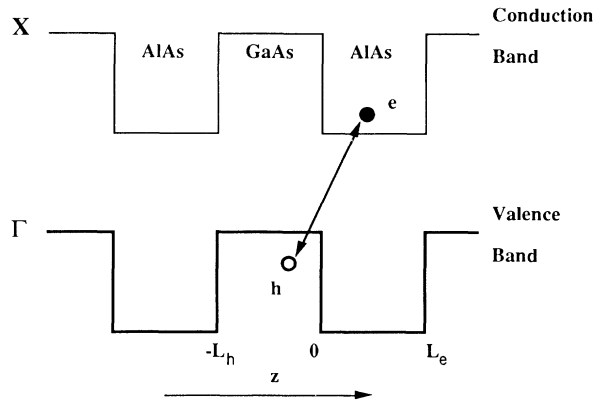

FIG. 1. The electron-hole pair in type-II QW structure.

hole $V_h(z_h)$ are assumed to be infinite, where the range of z coordinates are $0 \le z_e < L_e$ and $-L_h \le z_h \le 0$. For the perturbation-variational method, we rewrite the total Hamiltonian as follows:

$$H = H_0 + H' = H_0^e + H_0^h + H_0^{\rm exc} + H' \ , \tag{3}$$

where

$$H_0^e = -\frac{\hbar^2}{2(m_e)_l}\frac{\partial^2}{\partial z_e^2} + V_e(z_e) \ , \tag{4a}$$

$$H_0^h = -\frac{\hbar^2}{2(m_\pm)_l}\frac{\partial^2}{\partial z_h^2} + V_h(z_h) \ , \tag{4b}$$

$$H_0^{\rm exc} = -\frac{\hbar^2}{2\mu_\pm}\left[\frac{1}{\rho}\frac{\partial}{\partial\rho}\rho\frac{\partial}{\partial\rho} + \frac{1}{\rho^2}\frac{\partial^2}{\partial\phi^2}\right] - \frac{\lambda e^2}{\epsilon_0\rho} \ , \tag{4c}$$

and

$$H' = \frac{\lambda e^2}{\epsilon_0\rho} - \frac{e^2}{\epsilon_0[\rho^2 + (z_e - z_h)]^{1/2}} \ . \tag{4d}$$

We have added and subtracted the term $\lambda e^2/\epsilon_0\rho$, which corresponds to the Coulomb interaction between the electron-hole pair in the x-y plane. The term $H'(\lambda)$ in H can be taken as a small perturbation that physically represents the difference between 2D and 3D Coulomb interactions. For a general state, E_g, the energy perturbation series is $E_g = E_g^{(0)}(\lambda) + \Delta E_g^{(1)}(\lambda) + \cdots$, where $E_g^{(0)}(\lambda)$ is the unperturbed solution and $\Delta E_g^{(1)}(\lambda)$ is the first correction.

Since the total Hamiltonian in Eq. (1) does not depend on the parameter λ and only λ determines how much energy is divided between H_0 and H', we can choose λ in such a way to minimize the expectation value of the perturbation term H' to all orders of perturbation expansion. For the optimum value of λ we minimize $|\Delta E_g^{(1)}(\lambda)/E_g^{(0)}(\lambda)|$, which leads to fast convergence of the perturbation series.

The solutions for the unperturbed part H_0 are exactly known. The electron and hole motion in the z-direction is a one-dimensional problem with infinite potential barriers, while the exciton part is equivalent to the 2D hydrogen problem. The 2D problem has been discussed by Ralph[16] and Shinada and Sugano.[17] The eigenvectors for the unperturbed Hamiltonian H_0 are:

$$\Psi_g^{(0)} = \psi_0^e(z_e)\psi_0^h(z_h)\psi_{n,m}^{\rm exc}(\rho,\phi) = \left[\frac{4}{L_eL_h}\right]^{1/2}\sin\left[\frac{n_e\pi z_e}{L_e}\right]\left[\frac{n_h\pi z_h}{L_h}\right]\psi_{n,m}^{\rm exc}(\rho,\phi) \ , \tag{5}$$

where

$$\Psi_{n,m}^{\rm exc}(\rho,\phi) = \frac{1}{\sqrt{2\pi}}\exp[im\phi]\left[\frac{2\lambda}{a_\beta^*(n+1/2)}\right] \times \left[\frac{(n-|m|)!}{(2n+1)[(n+|m|)!]^3}\right]^{1/2} \times \exp[-\tilde\rho/2]\tilde\rho^{|m|}L_{n-|m|}^{2|m|}(\tilde\rho) \ , \tag{6}$$

with

$$\tilde\rho = \frac{2\rho\lambda}{a_\beta^*(n+1/2)} \ ,$$

and the eigenenergies are given by

$$E_g^{(0)} = E_e + E_h + E_n = \frac{\hbar^2\pi^2 n_e^2}{2(m_e)_l L_e^2} + \frac{\hbar^2\pi^2 n_h^2}{2(m_\pm)_l L_h^2} - \frac{{\rm R}^*\lambda^2}{(n+1/2)^2}, \quad n = 0,1,2,\cdots \ , \tag{7}$$

where $n_e, n_h = 1,2,\ldots$ and $L_{n-|m|}^{2|m|}(\tilde\rho)$ are the associated Laguerre polynomials, ${\rm R}^* = \mu_\pm e^4/2\epsilon_0^2\hbar^2$ is the 3D effective rydberg, and $a_\beta^* = \epsilon_0\hbar^2/\mu_\pm e^2$ is the transverse effective Bohr radius, while $|m| \le n$.

For the ground state (lowest subband exciton) ($n_e = n_h = 1, n = m = 0$), the first-order perturbation to the ground-state energy is:

$$\Delta E_g^{(1)}(\lambda) = \frac{e^2}{\epsilon_0 a_\beta^*}(4\lambda)^2\left[\frac{1}{4} + \frac{1}{L_eL_h\lambda}\int_0^{L_e}dz_e\int_{-L_h}^{0}dz_h\sin^2\left[\frac{\pi z_e}{L}\right]\sin^2\left[\frac{\pi z_h}{L}\right]\beta_1\left[1 - \frac{\pi}{2}[H_1(\beta_1) - N_1(\beta_1)]\right]\right] \ , \tag{8}$$

where $\beta_1 = 4\lambda(z_e - z_h)/a_\beta^* \ge 0$, while $H_k(\cdots)$ is the Struve function of order k and $N_k(\cdots)$ is the Neumann function of order k.[18]

Based on the first-order perturbative energy correction, the optimum condition requires $\Delta E_g^{(1)}(\lambda) = 0$, and thus determines the variational parameter $\lambda = \lambda_0$. Therefore, the binding energy for the ground-state exciton is $E_B = 4{\rm R}^*\lambda_0^2$.

We have calculated the values of the binding energies E_B of the heavy-hole and the light-hole exciton as a function of AlAs-layer thickness (or GaAs-layer thickness) up to first-order perturbative correction. The values of the various physical parameters involved in the GaAs/AlAs QW's that have been used in our calculations are $(m_e)_l=1.1m_0$, $(m_e)_t=0.19m_0$,[19] $\gamma_1=6.93,\gamma_2=2.15$;[20] for heavy-hole mass $[(m_+)_l=0.38m_0,(m_+)_t=0.11m_0]$; for light-hole mass $[(m_-)_l=0.089m_0,(m_-)_t=0.21m_0]$. The reduced mass in the x-y plane for the heavy-hole exciton is $\mu_+=0.07m_0$ and for the light-hole exciton $\mu_-=0.1m_0$.

In Fig. 2 we display the heavy-hole and light-hole exciton binding energies versus the AlAs-layer thickness, for the GaAs-layer width $L_h=20$ Å. The exciton binding energies calculated with a variational approach[3] using the above-mentioned material parameters along with the results from the first-order perturbation-variational expansion are displayed. The first-order perturbation expansion does not give results close to variational results even for small AlAs widths; a second-order perturbation term was calculated and the additional correction was less than 0.1 meV. For heavy-hole excitons, the percentage change in the binding energy from the first- to second-order correction in perturbation expansion is about 0.25% for $L_h=20$ Å and $L_e=40$ Å and increases for larger AlAs wells (at $L_e=20$ Å and $L_e=80$ Å, the percentage correction is about 0.41%). Regarding the difference between the variational and the perturbation-variational results, we observe that for $L_h=20$ Å and $L_e=40$ Å the difference is 0.71 meV, while for $L_h=20$ Å and $L_e=80$ Å the difference increases up to 0.92 meV. As the AlAs well thickness increases, the exciton binding energy E_B reduces from the two-dimensional value $4\mathcal{R}^*$ to the three-dimensional result $\mathcal{R}^*$. Similar results hold for the light-hole exciton binding energies. The percentage change in the binding energy from the first- to second-order correction in the perturbation expansion is about 0.35% for $L_h=20$ Å and $L_e=40$ Å, and 0.57% for $L_h=20$ Å and $L_e=80$ Å. The variational results are different from the perturbation-variational results as follows: at $L_h=20$ Å and $L_e=40$ Å the difference is 1.31 meV, while at $L_h=20$ Å and $L_e=80$ Å it is about 1.47 meV. For both the heavy-hole and the light-hole excitons, the binding energies are decreasing for increasing GaAs widths, due to the increase of the separation of electrons and holes that produces a loose exciton quasiparticle.

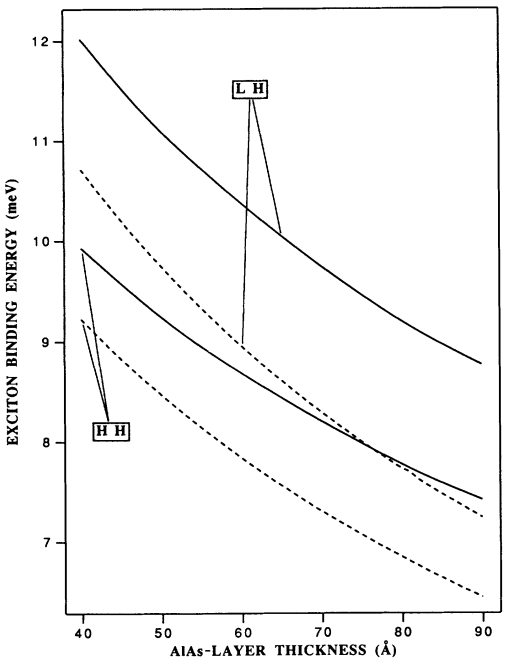

FIG. 2. Variation of the exciton binding energy of the heavy-hole (HH) exciton and the light-hole (LH) exciton as a function of AlAs-layer thickness for GaAs-layer thickness $L_h=20$ Å. Perturbation-variation expansion (— — —); variational calculation (Ref. 3) (——).

For comparison, we tested the perturbation-variational method in another QW system; we calculated the exciton binding energies associated with heavy and light holes for type-I QW's. The difference between the perturbation-variational method and the variational results[14] is the following: for heavy-holes at $L_{e\,(h)}=50$ Å it is about 0.23 meV, while for $L_{e\,(h)}=250$ Å, it is about 0.80 meV; for light holes the results for the same well widths are 0.11 and 0.77 meV, respectively. Similar to Jiang's[11] results, which were derived for type-I QW's for finite barriers, our results show that the first-order perturbation correction is not enough to approach the upper bound results of the variational calculation.[14] Higher-order corrections are needed to approach the variational upper bound.

In order to compare the results between type-I and type-II QW's, we point out the location of electrons and holes in these QW's: in type-I, electrons and holes are confined in the GaAs well, while in type-II, electrons and holes are separated in different wells. This implies that the average relative separation between electrons and holes $(\langle|z_e-z_h|\rangle/a^*_{\mathrm{I(II)}})$ is different for both cases; for example, in the type-I case, $\langle|z_e-z_h|\rangle/a^*_{\mathrm{I}}\sim 0.21L_{e(h)}/a^*_{\mathrm{I}}$ (both electrons and holes reside in the GaAs well), while for type-II QW's , $\langle|z_e-z_h|\rangle/a^*_{\mathrm{II}}\sim(L_e+L_h)/2a^*_{\mathrm{II}}$ (where a^*_{I} and a^*_{II} are the Bohr radii for type-I and type-II QW's, respectively). The magnitude of the perturbation term H' depends on the ratio of the average separation of the electron-hole pair over the corresponding exciton Bohr radius. The value of the relative separation, namely $\langle|z_e-z_h|\rangle/a^*_{\mathrm{I(II)}}$, is an indication of the magnitude of H'. Le us estimate for which well widths for both cases the quantity $\langle|z_e-z_h|\rangle/a^*_{\mathrm{I(II)}}$ is the same: if we take $L_e=20$ Å and $L_h=40$ Å, and $a^*_{\mathrm{II}}\sim 93$ Å for the type-II heavy-hole exciton case, $\langle|z_e-z_h|\rangle/a^*_{\mathrm{II}}\sim 0.32$. For the type-I case, $a^*_{\mathrm{I}}\sim 165$ Å, and this corresponds to $L_{e\,(h)}\sim 250$ Å. Our results indicate that in the type-I case, the difference between the variational and perturbation results, especially around $L_{e\,(h)}\sim 250$ Å (type-I), is almost the same as in the type-II case for $L_e=20$ Å and $L_h=40$ Å (about 0.75 meV). Thus, the difference between the two-dimensional and the three-dimensional Coulomb interaction (H') depends on the relative separation $\langle|z_e-z_h|\rangle/a^*_{\mathrm{I(II)}}$; its value is

determined by the exciton Bohr radius and the average separation of the electron-hole pair. For type-I QW's, widths about 50 Å can be considered small due to the large Bohr radius ($\sim$165 Å for heavy hole), while for type-II QW's, the same widths are large in magnitude, compared with the corresponding Bohr radius. As QW thickness increases for both types, the three-dimensional Coulomb interaction becomes dominant, and summation over high-order corrections is required to calculate the exciton binding energies. Our results indicate that for the overall picture of the perturbation-variational method, corrections up to first order are not sufficient to reproduce the variational results, and higher-order terms have to be calculated.

In conclusion, we have presented a calculation of the binding energies of both the heavy-hole and the light-hole excitons in type-II GaAs/AlAs QW's as a function of the size of the AlAs layer (or the GaAs layer). We have used a perturbation-variational expansion method and included terms up to first order in perturbation expansion in our calculations. The exciton binding energies are calculated assuming infinite potential barriers and results thus obtained are slightly less in magnitude with those of a variational calculation.[3] We have found that summing over all higher-order corrections is essential to deriving the exciton binding energies for type-II QW's; the same result applies to type-I QW's. We have demonstrated that the spatial separation for the electron-hole system and the corresponding Bohr radius influences the perturbation expansion parameter not in the same way for type-I and type-II QW's; the expansion parameter $\langle |z_e - z_h| \rangle / a^*_{\mathrm{I(II)}}$ has the same value for large type-I wells and small type-II wells.

We thank X. Zhang for useful discussions. This work was supported by the Air Force Office of Scientific Research under Grant No. AFOSR-91-0056.

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