# Exciton binding energies and oscillator strengths in a symmetric $Al_xGa_{1-x}As/GaAs$ double quantum well

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Within the framework of the effective-mass approximation, we have used an improved three-parameter trial function to calculate the exciton binding energy for both the ground state and the excited states in a symmetric double quantum well. When comparing our results with those derived with the commonly used one-parameter trial function, we found that the binding energies are improved significantly, especially for the excited states. Our calculated exciton transition energies agree well with the measured photoluminescence-excitation (PLE) spectra. We have also calculated the oscillator strengths. By comparing our results with the oscillator strength derived from the PLE spectra, we find good agreement for heavy holes, but less satisfactory agreement for light holes.

The physical properties of superlattices and single quantum wells have attracted much attention in the literature. Recently, the electronic and optical properties of the double-quantum-well (DQW) have received increasing interest.[1,2] One of the central themes concerns the enhancement of the exciton binding energy[3] due to the confinement of the electron-hole pair wave function in a heterostructure. If the binding energy becomes comparable to the thermal energy at room temperature, such a situation will be of great interest to electro-optical devices.[3]

Various approaches have been used to study the exciton in quantum well systems.[4–14] For typical well widths of experimentally investigated single quantum well samples, the energy separation between subbands is much larger than the exciton binding energy, and therefore the binding energy of the exciton ground state is hardly affected by the higher lying subband states.[6,7] However, for DQW's the situation can be quite different.

Let the interfaces of the DQW system be parallel to the $xy$ plane. Within the effective-mass approximation (EMA), the potential along the $z$ axis for electrons (or holes) in the conduction (or valence) band is shown schematically in the inset of Fig. 1. If the central barrier is sufficiently wide the excitons in different wells are practically uncoupled. When the barrier width is reduced the excitons in the two wells begin to interact, and the theoretical analysis in the present paper is restricted to symmetric structures of such kind.

We start from a single quantum well which has one electron energy level $\epsilon_e$, one heavy hole level $\epsilon_{hh}$, and one light hole level $\epsilon_{lh}$. When two such wells are separated by a finite barrier, the splits between the pairs of hybridized energy levels $\epsilon_{e2}$-$\epsilon_{e1}$, $\epsilon_{hh1}$-$\epsilon_{hh2}$, and $\epsilon_{lh1}$-$\epsilon_{lh2}$ depend on the barrier width. One can tailor-make a structure such that these energy splits are comparable to the exciton binding energy in a single quantum well. Consequently, the excited states in a DQW can be of great importance to the exciton binding energy.

It has been recognized that the spherically symmetric single-exponential envelope function commonly used for the excitonic ground state in a bulk semiconductor is not suitable for the excited states. Due to the symmetry

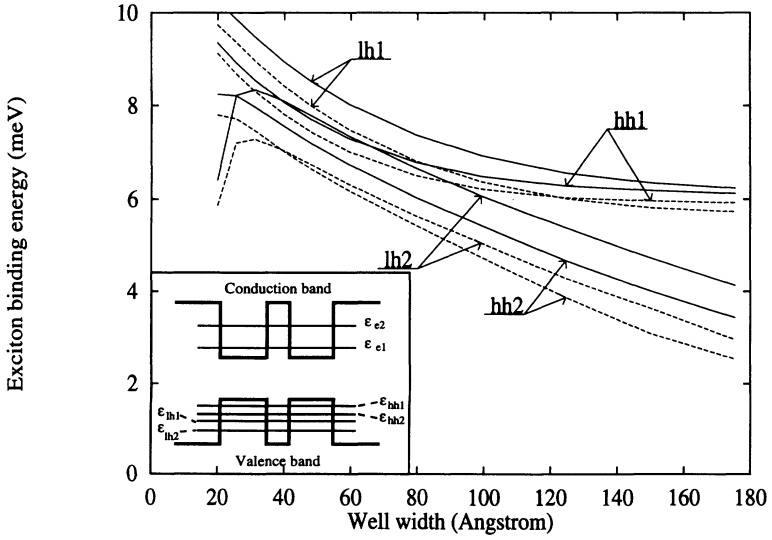


FIG. 1. Exciton binding energy as a function of well width for fixed barrier width of 14.15 Å. Solid curves are three-parameter trial function results, and dashed curves are one-parameter trial function results.

 

breaking along the direction perpendicular to the interfaces, such $s$-like envelope functions are also improper even for the excitonic ground state in quantum wells systems.[4,12,14] For example, in a strongly coupled DQW, a sizable contribution to the exciton ground state binding energy comes from the configuration in which the electron and hole are confined in different wells. In this paper we will use an improved trial function to calculate the exciton binding energy for both the ground state and excited states. We further calculate the oscillator strengths.

The DQW has translational symmetry in the $xy$ plane. According to the envelope function theory the normalized *single particle* wave function can be written as a product of a plane wave in the $xy$ plane, and the envelope $\chi_{\nu n}(z)$, in the growth direction $z$. In our case we will limit ourselves to three bands ($\nu$=$e$, hh, lh); $e$ for electrons in the conduction band, and hh (or lh) for heavy holes (or light holes) in the valence band. Since in a heterostructure the heavy and light hole bands are split, we will ignore the mixing of heavy and light hole states. The two excitonic states of electron–heavy hole and electron–light hole are therefore decoupled and can be treated separately. To simplify the notation without causing any confusion, we can therefore use the subscript $\nu$=$h$ for either heavy hole or light hole. The several subbands in the DQW system are labeled by the subscript $n$. The subband states $\chi_{\nu n}(z)$ are found with the transfer matrix method.

Including the Coulomb interaction between the electron and the hole, the excitonic Hamiltonian, $H$=$H_e$+$H_h$+$H_{eh}$, consists of the single-particle part

$$H_\nu = \frac{-\hbar^2}{2}\frac{\partial}{\partial z_\nu}\frac{1}{m_{\nu z}}\frac{\partial}{\partial z_\nu} + V_\nu(z_\nu)$$

for $\nu$=$e$ and $h$, and the electron-hole interaction part

$$H_{eh} = \frac{-\hbar^2}{2\mu_\perp}\left(\frac{1}{\rho}\frac{\partial}{\partial\rho}\rho\frac{\partial}{\partial\rho}\right) - \frac{e^2}{\varepsilon r} ,$$

where $m_{\nu z}$ is the longitudinal effective mass and $\mu_\perp$ the reduced transverse effective mass of the electron-hole pair. The electron and hole coordinates are $\vec{r}_e = (\vec{\rho}_e, z_e)$ and $\vec{r}_h = (\vec{\rho}_h, z_h)$, with the relative distances $\vec{r}$=$\vec{r}_e - \vec{r}_h$ and $\vec{\rho}$=$\vec{\rho}_e - \vec{\rho}_h$. The potential $V_e(z_e)$ [or $V_h(z_h)$] for electrons (or holes) in the conduction (or valence) band is schematically illustrated in inset of Fig. 1. The kinetic energy operator of $H_\nu$ is expressed in its proper form, and the corresponding matching conditions[15–18] will be used when solving the eigenvalue equation $H_\nu\chi_{\nu n}(z_\nu) = \epsilon_{\nu n}\chi_{\nu n}(z_\nu)$.

To study the characteristic features of excitons in a DQW, it is sufficient to have only two bound states $\chi_{\nu 1}(z_\nu)$ and $\chi_{\nu 2}(z_\nu)$. Let us define $\phi_1(z_e,z_h)$= $\chi_{e1}(z_e)\chi_{h1}(z_h)$, $\phi_2(z_e,z_h)$=$\chi_{e1}(z_e)\chi_{h2}(z_h)$, $\phi_3(z_e,z_h)$= $\chi_{e2}(z_e)\chi_{h1}(z_h)$, and $\phi_4(z_e,z_h)$=$\chi_{e2}(z_e)\chi_{h2}(z_h)$. With proper trial envelope functions

$$F(\rho,z) = \left\{1+\left(\frac{z}{\alpha}\right)^2\right\}\exp\left\{-\sqrt{\left(\frac{\rho}{\lambda_1}\right)^2+\left(\frac{z}{\lambda_2}\right)^2}\right\} , \quad (1)$$

the variational wave functions for both the electron–heavy hole and the electron–light hole excitonic state can be constructed as

$$\psi(\vec{r}_e,\vec{r}_h) = F(\rho,z)\sum_{m=1}^{4} C_m\phi_m(z_e,z_h) . \quad (2)$$

In (1), the *plus* sign in front of $(z/\alpha)^2$ is not obvious, but our calculation shows that this is the correct sign. The excitonic state (2) contains seven variational parameters $\alpha$, $\lambda_1$, $\lambda_2$, and $C_m$'s, which are determined by minimizing the energy $E = \langle\psi(\vec{r}_e,\vec{r}_h)|H|\psi(\vec{r}_e,\vec{r}_h)\rangle$ , subject to the normalization constraint $\langle\psi(\vec{r}_e,\vec{r}_h)|\psi(\vec{r}_e,\vec{r}_h)\rangle$=1. If we minimize $E$ first with respect to the coefficients $C_m$, we get the eigenvalue problem

$$\sum_{m=1}^{4}[H_{lm} - E\,S_{lm}]C_m = 0, \qquad l = 1,...,4,$$

where $H_{lm}$=$\langle\phi_l\,F|H|F\,\phi_m\rangle$ and $S_{lm}$=$\langle\phi_l\,F|F\,\phi_m\rangle$. If the matrix elements $H_{lm}$ and $S_{lm}$ are known, the four eigensolutions can be easily derived. However, these matrix elements depend on $\lambda_1$, $\lambda_2$, and $\alpha$, the optimum values of which need to be determined numerically. Without going into details we will just briefly sketch how this is done. By use of the symmetry of the DQW, the eigenvalue problem can be decoupled into a symmetric and an antisymmetric part, which can be treated further in equivalent ways. Let us consider the two symmetric eigenstates with energies $E_{s1}$ and $E_{s2}$. In principle, the minima of $E_{s1}$ and $E_{s2}$ correspond to two different sets of ($\lambda_1$, $\lambda_2$, $\alpha$) values, but we have approximated them equal for the two states. However, the parameter-set should minimize both $E_{s1}$ and $E_{s2}$ simultaneously, which we have obtained by minimizing the object function $0.9\,E_{s1}$+$0.1\,E_{s2}$. This gives results very close to the true minima.

Our theoretical results will be compared with the measured photoluminescence-excitation (PLE) spectra, for which the transition from the initial state to an antisymmetric exciton state is forbidden. Hence, we will present only the calculated energies $E_{\rm hh1}$, $E_{\rm hh2}$, $E_{\rm lh1}$, and $E_{\rm lh2}$ for symmetric exciton states. PLE experiments have been performed on two series of symmetric $Al_{0.3}Ga_{0.7}As/GaAs$ DQW samples with varying well width but fixed barrier width 14.15 Å (five monolayers), and 19.81 Å (seven monolayers).[2] We will only show results for the 14.15-Å samples, while the results for the others are similar.

The calculated exciton binding energies $E_{B,\rm hh1}$=$\epsilon_{\rm hh1}$+$\epsilon_{e1}$ − $E_{\rm hh1}$, $E_{B,\rm hh2}$= $\epsilon_{\rm hh2}$+$\epsilon_{e2}$ − $E_{\rm hh2}$, $E_{B,\rm lh1}$=$\epsilon_{\rm lh1}$+$\epsilon_{e1}$ − $E_{\rm lh1}$, and $E_{B,\rm lh2}$=$\epsilon_{\rm lh2}$+$\epsilon_{e2}$ − $E_{\rm lh2}$ are shown as solid curves in Fig. 1. $E_{B,\rm hh1}$ and $E_{B,\rm lh1}$ correspond to states where we have a high probability for the electron and hole to be in the same well, thus for infinite well width the binding energies will approach the values of bulk GaAs. On the other hand, for $E_{B,\rm hh2}$ and $E_{B,\rm lh2}$ the electron and hole are in opposite wells, hence the binding energy approaches zero. These features are clearly indicated in Fig. 1. In the opposite limit of very narrow well width, the decrease in the binding energy of $E_{B,\rm hh2}$ and $E_{B,\rm lh2}$ is due to the reduced confinement

of electron and hole as the energy levels approach the continuum above the barriers. We have also used a one-parameter trial function[2] where we have set $\lambda_2=\lambda_1$ and $1/\alpha = 0$. The so-obtained binding energies shown in Fig. 1 as dashed curves exhibit a decrease of about 0.5 to 1 meV (6–12%) for the hh1 and lh1 states, and about 1 to 1.5 meV (12–35% depending on well width) for the hh2 and lh2 states. It is reasonable to believe that our improved wave functions, which give much larger binding energies, should also lead to improved calculations of the oscillator strengths.

Figure 2 shows the PLE experimental results[2] for the series of samples of barrier width 14.15 Å. The well width of each sample is indicated next to the corresponding data curve. The peak at the lowest photon energy is the measured $E_{\rm hh1}$, which is larger than our calculated $E_{\rm hh1}$ by an amount between 1 meV and 8 meV. Since the variational ground state energy cannot be lower than the true ground state energy, this discrepancy, remarkably small compared to the band gap 1955 meV of $Al_{0.3}Ga_{0.7}As$, is most likely caused by the sample structure dependence of the band gap and the band-offset parameter $Q$. Therefore, when comparing with experimental results, for each sample we have shifted the zero reference energy by the amount indicated in parentheses, in order to bring the calculated $E_{\rm hh1}$ in agreement with the measured one. Our calculated energies $E_{\rm hh1}$, $E_{\rm lh1}$, $E_{\rm hh2}$, and $E_{\rm lh2}$, marked in Fig. 2 by vertical bars, agree well with the measurement. With increasing well width, all energies approach the bulk GaAs values, and $E_{\rm lh1}$ and $E_{\rm hh2}$ get very close to each other. We should mention that for the samples of 19.81 Å barrier width, $E_{\rm lh1}$ and $E_{\rm hh2}$ even cross at well width about 70 Å.

We will now turn to the oscillator strength calculations. The initial state $\Psi_i$ is the ground state of an intrinsic semiconductor at zero temperature with a filled valence band and an empty conduction band. $\Psi_f$ is the final state after absorption of a photon. $\Psi_i$ and $\Psi_f$ are $N$-particle states. Then, the absorption coefficient can be calculated from the golden rule as

$$\alpha(\omega) = \frac{\hbar\omega}{I_0}\frac{N}{S}\frac{2\pi}{\hbar}\sum_{i,f}|\langle\Psi_i|V|\Psi_f\rangle|^2\,\delta(E_f - E_i - \hbar\omega)\,.$$

Here $I_0$ is the incident light-intensity, and $N/S$ is the two-dimensional particle density in the wells. From the theory of interband absorption[19,20] the matrix element has the form

$$\langle\Psi_f|V|\Psi_i\rangle = \sum_{\vec{k}_e\vec{k}_h} A(\vec{k}_e,\vec{k}_h) \times \int d\vec{r}\,\phi^*_{v,-\vec{k}_h}(\vec{r})\,\frac{e}{m_0}\vec{A}\cdot\vec{p}\,\phi_{c,\vec{k}_e}(\vec{r})\,,$$

where $\phi$ is the single-particle Bloch function and $A(\vec{k}_e,\vec{k}_h)$ the Fourier component of the exciton wave function (2). Then, following the standard theory of excitons, we calculate the matrix elements. It is important to point out that although the final states in general have different energies, for a large range of well widths the measured $E_{\rm hh2}$ peak and the $E_{\rm lh1}$ peak coincide, causing uncertainty in the interpretation of the experimental data. In order to be able to compare with experimental data, we first calculate the oscillator strengths $\alpha_{\rm hh1}$, $\alpha_{\rm hh2}$, $\alpha_{\rm lh1}$, and $\alpha_{\rm lh2}$ of the four corresponding exciton states. Then, the calculated ratios $\alpha_{\rm hh2}/\alpha_{\rm hh1}$ and $\alpha_{\rm lh2}/\alpha_{\rm lh1}$ are compared with the corresponding measured values. Our theoretical results are shown in Fig. 3 as solid curves for samples of barrier width 14.15 Å, together with the experimental results derived by Westgaard *et al.*[2] from the spectra in Fig. 2.

Westgaard *et al.* have also calculated the ratios of the oscillator strengths which we reproduce in Fig. 3.

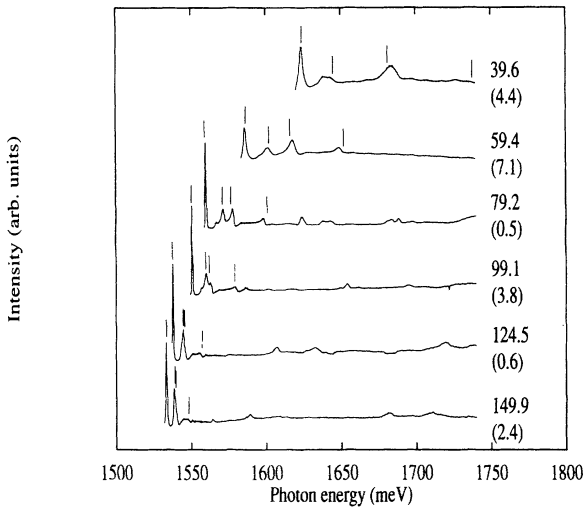


FIG. 2. PLE spectra of a set of samples with fixed barrier width of 14.15 Å. The well width of each sample is indicated at the right-hand side. Vertical bars mark the calculated exciton transition energies, with a shift of each spectrum to fit the $E_{\rm hh1}$ peak. The amount of shift is given by a number in parentheses.

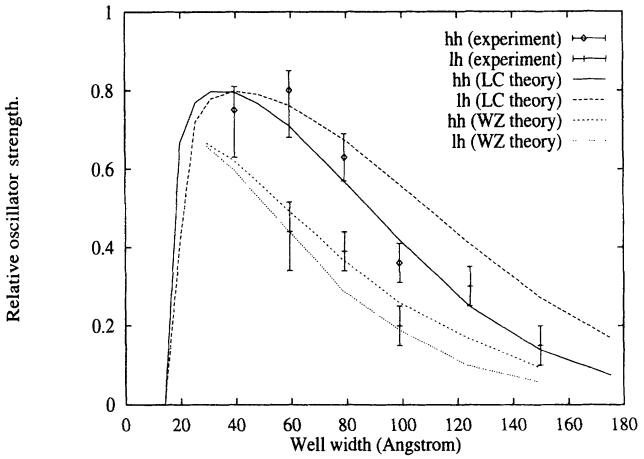


FIG. 3. Relative oscillator strengths (hh=$\alpha_{\rm hh2}/\alpha_{\rm hh1}$ and lh=$\alpha_{\rm lh2}/\alpha_{\rm lh1}$) as functions of well width for fixed barrier width of 14.15 Å. Our theoretical calculations are indicated by (LC theory) and the results of Westgaard *et al.* (Ref. 2) by (WZ theory). The measurements are also from Westgaard *et al.*

In their calculation they have approximated the initial states with a constant and the final states with a one-parameter trial function. The difference between the two sets of theoretical curves must be ascribed to the different wave functions and methods of oscillator-strength calculations. The $\alpha_{hh2}/\alpha_{hh1}$ ratios of our calculation are within the error bars and agree with the experimental data better than the theoretical results of Westgaard *et al.* However, our $\alpha_{lh2}/\alpha_{lh1}$ ratios do not fit as well with experiment as the results of Westgaard *et al.*, contrary to expectation, since we use a more advanced calculation. We have no definite explanation for this, but we suspect that Westgaard *et al.* have underestimated the error in the oscillator strengths due to the effect of the background level of the data. By a visual inspection of the experimental data (Fig. 2) we see that the light-hole peaks are generally smaller than the heavy-hole peaks, and a background level will therefore have a larger influence on light-hole than heavy-hole oscillator strengths.

In conclusion, we find a good agreement with experiments for the heavy-hole oscillator strengths but a poor agreement for light holes.

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