Energy levels of Wannier excitons in GaAs-$Ga_{1-x}Al_xAs$ quantum-well structures

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Energy levels of Wannier excitons in a quantum-well structure consisting of a single slab of GaAs sandwiched between two semi-infinite layers of $Ga_{1-x}Al_xAs$ are calculated with the use of a variational approach. Owing to lowering of symmetry along the axis of growth of this quantum-well structure and the presence of energy-band discontinuities at the interfaces, the degeneracy of the valence band of GaAs is removed, leading to two exciton systems, namely, the heavy-hole exciton and the light-hole exciton. The values of the binding energies of the ground state and of a few low-lying excited states of these two exciton systems are calculated as a function of the size of the GaAs quantum well for several values of the heights of the potential barriers and their behavior is discussed. The results thus obtained are also compared with the available experimental data. The reliability of the various approximations made in this calculation is discussed.

INTRODUCTION

The study of the behavior of Wannier excitons in a quantum-well structure consisting of a single layer of GaAs sandwiched between two semi-infinite (generally greater than 200 Å in practice) layers of $Ga_{1-x}Al_xAs$ has attracted considerable attention in recent years.[1-5] The conduction- and the valence-band discontinuities at the interface between the two semiconductors at the Γ point have been shown to be about 85% and 15%, respectively, of the total energy-band-gap difference.[1] Owing to a reduction in symmetry along the axis of growth of this quantum-well structure and the presence of energy-band discontinuities, degeneracy of the valence band of GaAs is removed, leading to the formation of two exciton systems, namely, the heavy-hole exciton and the light-hole exciton.

In this paper we present a calculation of the energies of the ground state and of a few low-lying excited states of heavy- and light-hole excitons associated with the lowest electron and hole subbands in this quantum-well structure for finite values of the potential barrier heights. We follow a variational approach and calculate the energy levels as a function of the GaAs layer thickness for several different values of the potential barrier (or equivalently, for several values of Al concentration x). In addition, we calculate the energy levels for infinite potential barriers and compare our results for the $1s$ state with those of Bastard *et al.*[4] and for the $1s$ and $2s$ states with those of Miller *et al.*[3] Both of these groups calculate the energies of these states for infinite potential barriers only. We find that both the magnitude and the qualitative behavior of the energy levels calculated using finite barrier heights are quite different from those obtained using infinite potential barriers, especially for thin ($L < 100$ Å) wells. This is not unusual in view of the fact that for most commonly used values of x ($x \leq 0.4$) the band discontinuities cannot be treated as infinite. This is especially true for the valence bands where the discontinuity, as mentioned earlier, is only about 15% of the total band-gap difference. A brief report of the calculation of the energies of the ground state of the two exciton systems has already been presented.[5] In this paper we provide some additional details and present calculations of the energies of several low-lying excited states of heavy- and light-hole excitons. Both even- and odd-parity states are examined as a function of the quantum-well size for several different values of the potential barrier heights. For the sake of completeness, we mention briefly our results on the ground-state energies of the two exciton systems. We also compare our results with the available experimental data. Finally, we comment on the validity of the exciton Hamiltonian used and discuss the various approximations made in the present calculation.

THEORY

The Hamiltonian of an exciton associated with either the heavy-hole or the light-hole band in a GaAs slab sandwiched between two semi-infinite layers of $Ga_{1-x}Al_xAs$ grown along the (001) direction can be expressed (within the framework of an effective-mass approximation) using cylindrical coordinates as[6]

$$H=\frac{-\hbar^2}{2\mu_{\pm}}\left[\frac{1}{\rho}\frac{\partial}{\partial\rho}\rho\frac{\partial}{\partial\rho}+\frac{1}{\rho^2}\frac{\partial^2}{\partial\phi^2}\right]-\frac{\hbar^2}{2m_e}\frac{\partial^2}{\partial z_e^2}-\frac{\hbar^2}{2m_{\pm}}\frac{\partial^2}{\partial z_h^2}-\frac{e^2}{\epsilon_0|\vec{r}_e-\vec{r}_h|}+V_{ew}(z_e)+V_{hw}(z_h)\ . \tag{1}$$

Here m_e is the effective mass of the conduction electron, ϵ_0 is the static dielectric constant, $m_{\pm}$ is the heavy- (+)

or light- (−) hole mass along the z direction, and $\mu_\pm$ is the reduced mass corresponding to heavy- (+) or light- (−) hole bands in the plane perpendicular to the z axis. Both $\mu_\pm$ and $m_\pm$ can be expressed in terms of the well-known Kohn-Luttinger[7] band parameters γ_1 and γ_2 as[6]

$$\frac{1}{\mu_\pm}=\frac{1}{m_e}+\frac{1}{m_0}(\gamma_1\pm\gamma_2) \tag{2}$$

and

$$\frac{1}{m_\pm}=\frac{1}{m_0}(\gamma_1\mp 2\gamma_2)\ , \tag{3}$$

where m_0 is the free-electron mass. In these equations the upper sign refers to the $J_z=\pm\frac{3}{2}$ (heavy-hole) band and the lower sign to the $J_z=\pm\frac{1}{2}$ (light-hole) band. The positions of the electron and the hole are designated by $\vec{r}_e$ and $\vec{r}_h$, respectively, and ρ, ϕ, and z are the relative electron-hole coordinates in the cylindrical coordinate system. In our expression for the Hamiltonian we have used the same values for the conduction- and the valence-band mass parameters in GaAs and $Ga_{1-x}Al_xAs$. In addition, we have assumed the same values for the static dielectric constant in the two semiconductors, thus neglecting the effects of the image charges. For most practical quantum-well structures these should be good approximations, as for the most commonly used values of x, these physical parameters are not too different in the two materials.[8] The potential wells for the conduction electron $V_{ew}(z_e)$ and for the holes $V_{hw}(z_h)$ are assumed to be squre wells of width L,

$$V_{ew}(z_e)=\begin{cases}0, & |z_e|<L/2\\ V_e, & |z_e|>L/2\end{cases} \tag{4a}$$

and

$$V_{hw}(z_h)=\begin{cases}0, & |z_h|<L/2\\ V_h, & |z_h|>L/2\ .\end{cases} \tag{4b}$$

Here we have chosen, without any loss of generality, the origin of the coordinate system to be the center of the GaAs well. The values of the potential-well heights V_e and V_h are determined from the Al concentration in $Ga_{1-x}Al_xAs$, using the following recently proposed expression[8] for the total energy-band-gap discontinuity:

$$\Delta E_g=1.155x+0.37x^2\ , \tag{5}$$

in units of electron volts. The values of V_e and V_h are assumed to be about 85% and 15% of ΔE_g, respectively. Fairly accurate values of the Kohn-Luttinger parameters γ_1 and γ_2 for GaAs have recently been determined[9] using magnetoabsorption data.

An exact solution of the Schrödinger equation corresponding to the exciton Hamiltonian [Eq. (1)] is clearly not possible. We shall therefore follow a variational approach and use the following from of the trial wave function:

$$\psi=f_e(z_e)f_h(z_h)g(\rho,z,\phi)\ . \tag{6}$$

The functions $f_e(z_e)$ and $f_h(z_h)$ are taken to be (unnormalized) ground-state solutions for the finite square-well potentials. For example, for the electron,[10]

$$f_e(z_e)=\begin{cases}\cos(k_e z_e), & |z_e|<L/2\\ B_e e^{-\kappa_e|z_e|}, & |z_e|>L/2\ .\end{cases} \tag{7}$$

The parameter k_e is determined for the energy of the first electron subband, and B_e and κ_e are obtained from k_e by requiring continuity of f_e and its first derivative at the interface. The hole wave function f_h is obtained in a similar fashion. The validity of the interface connection rules for these effective-mass wave functions for GaAs-$Ga_{1-x}Al_xAs$ heterojunctions is discussed briefly later in this paper.

The function $g(\rho,z,\phi)$ of Eq. (6) is assumed to depend only on the relative electron-hole coordinates. We have chosen the following form for $g(\rho,z,\phi)$, depending on the projection of the angular momentum along the z axis (quantum number m):

$$g(\rho,z,\phi)=\rho^{|m|}e^{im\phi}\sum_j a_j g_j(\rho,z)\ , \tag{8}$$

where the basis functions g_j are taken to be

$$g_1(\rho,z)=e^{-\alpha(\rho^2+z^2)^{1/2}}\ , \tag{9a}$$

$$g_2(\rho,z)=z^2e^{-\alpha(\rho^2+z^2)^{1/2}}\ , \tag{9b}$$

$$g_3(\rho,z)=\rho e^{-\beta(\rho^2+z^2)^{1/2}}\ . \tag{9c}$$

The quantities α and β are nonlinear variational parameters which are adjusted to minimize the energy. The coefficients a_j are determined in the usual way by solving the matrix eigenvalue equation

$$H\psi=EU\psi\ . \tag{10}$$

The Hamiltonian and overlap matrices are formed using the complete basis described above,

$$\psi_j=f_e(z_e)f_h(z_h)\rho^{|m|}e^{im\phi}g_j(\rho,z)\ . \tag{11}$$

For $m=0$ ($1s$- and $2s$-like states) we use all three g_j functions of Eqs. (9a)–(9c); for $m=\pm1$ ($2p_\pm$-like states) only the first two g_j functions are used. Note that the $1s$ and $2s$ energies are determined simultaneously, so that the trial wave functions are orthogonal to each other.

The variational binding energies of the $1s$, $2s$, and $2p_\pm$ exciton states are obtained by subtracting from the lowest electron and hole subband energies (E_e and E_h) the eigenvalues of Eq. (10). These subband energies are determined by numerically solving the transcendental equations for the finite square wells,[10]

$$\left[\frac{E_e}{V_e}\right]^{1/2}=\cos\left[\left[\frac{m_e E_e}{2\hbar^2}\right]^{1/2}L\right] \tag{12a}$$

and

$$\left[\frac{E_h}{V_h}\right]^{1/2}=\cos\left[\left[\frac{m_\pm E_h}{2\hbar^2}\right]^{1/2}L\right]\ . \tag{12b}$$

This procedure yields values which are lower bounds for the true binding energies.

RESULTS AND DISCUSSION

We have calculated the values of the binding energies of the $1s$, $2s$, and $2p_{\pm}$ states of the heavy-hole exciton and the light-hole exciton as a function of L for values of Al concentration $x=0.15$ and 0.30. In order to compare our results with those of Miller *et al.*[3] and Bastard *et al.*[4] we have also calculated the binding energies of these levels as a function L for an infinite potential well. The values of the various physical parameters pertaining to GaAs,[9] used in our calculations are $m_e=0.067m_0$, $\epsilon_0=12.5$, $\gamma_1=7.36$, and $\gamma_2=2.57$. The values of the heavy-hole mass (m_+) and of the light-hole mass (m_-) obtained using these values of γ_1 and γ_2 are $0.45m_0$ and $0.08m_0$, respectively. The reduced mass in the x-y plane for the heavy-hole $(J_z=\pm\frac{3}{2})$ exciton is $0.04m_0$ and for the light-hole $(J_z=\pm\frac{1}{2})$ exciton, the reduced mass is $0.051m_0$. The reduced mass associated with $J_z=\pm\frac{3}{2}$ band is smaller than that associated with $J_z=\pm\frac{1}{2}$. This is due to the anisotropic nature of the kinetic energy expressions in the diagonal terms of the Kohn-Luttinger Hamiltonian for an exciton.[7]

In Fig. 1 we display the variation of binding energy of the ground state of a heavy-hole exciton $E_{1s}(h)$ (solid lines) and a light-hole exciton $E_{1s}(l)$ (dashed lines) as a function of L for three different values of potential barrier heights. The variation of the binding energy of the $2p_{\pm}$ state of the heavy-hole exciton $E_{2p_{\pm}}(h)$ (solid lines) and of the light-hole exciton $E_{2p_{\pm}}(l)$ (dashed lines) are displayed in Fig. 2. In Fig. 3 we show the variation of the binding energy of the $2s$ state of the heavy-hole exciton $E_{2s}(h)$ (solid line) and of the light-hole exciton $E_{2s}(l)$ (dashed lines). There are several interesting features to be noted in these plots. In Fig. 1, for instance, we find that for a given value of x, the value of $E_{1s}(h)$ increases as L is reduced until it reaches a maximum, and then decreases quite rapidly. The value of L at which $E_{1s}(h)$ reaches a maximum is smaller for larger x. Essentially the same behavior is exhibited by $E_{1s}(l)$. The binding energies of the excited states $2p_{\pm}$ and $2s$ as a function of L have the same type of behavior. The reason for this is quite simple. As L is reduced, the exciton wave function is compressed in the quantum well, leading to increased binding. However, beyond a certain value of L the spread of the exciton wave function into the surrounding $Ga_{1-x}Al_xAs$ layers becomes more important. This makes the binding energy

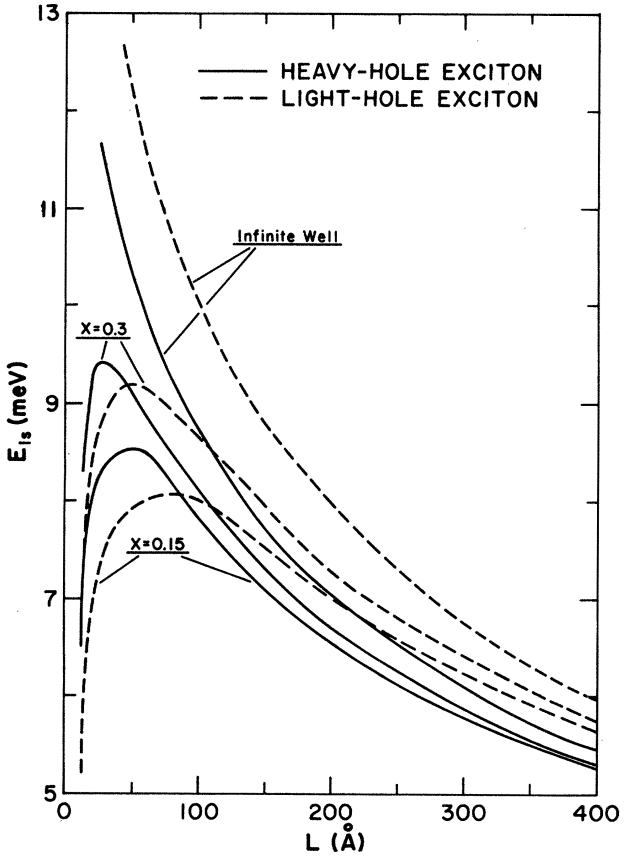

FIG. 1. Variation of the binding energy of the ground state, E_{1s}, of a heavy-hole exciton (solid lines) and a light-hole exciton (dashed lines) as a function of the GaAs quantum-well size (L) for Al concentrations $x=0.15$ and 0.3, and for an infinite potential well.

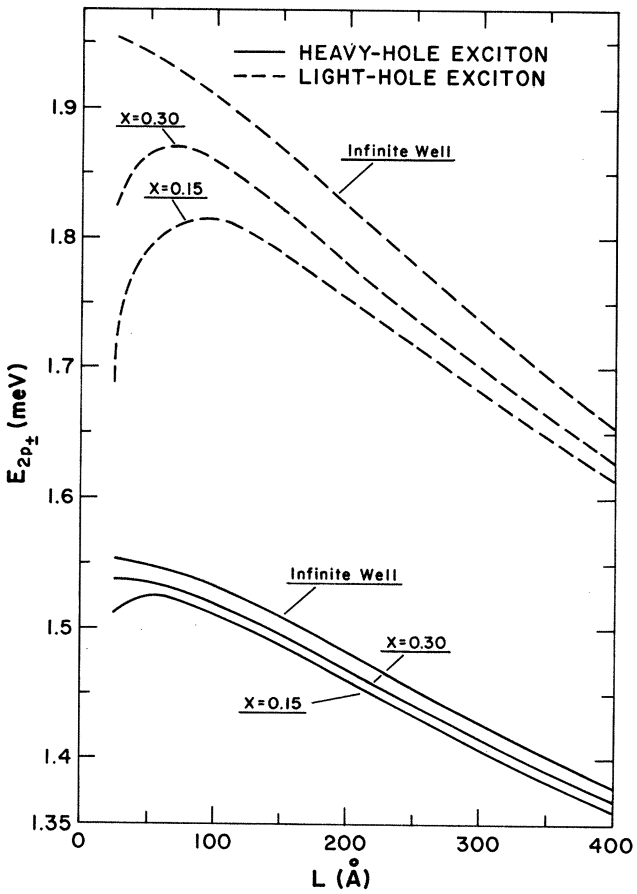

FIG. 2. Variation of the binding energy of the $2p_{\pm}$ state, $E_{2p_{\pm}}$, of a heavy-hole exciton (solid lines) and a light-hole exciton (dashed lines) as a function of the GaAs quantum-well size (L) for Al concentrations $x=0.15$ and 0.30, and for an infinite potential well.

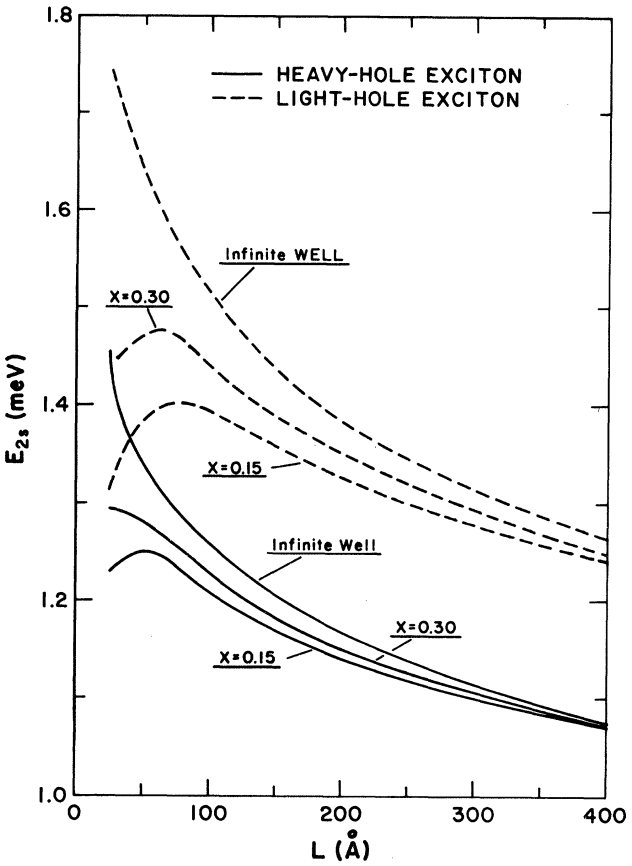

FIG. 3. Variation of the binding energy of the $2s$ state, E_{2s}, of a heavy-hole exciton (solid lines) and of a light-hole exciton (dashed lines) as a function of the GaAs quantum-well size (L) for Al concentrations $x=0.15$ and 0.30, and for an infinite potential well.

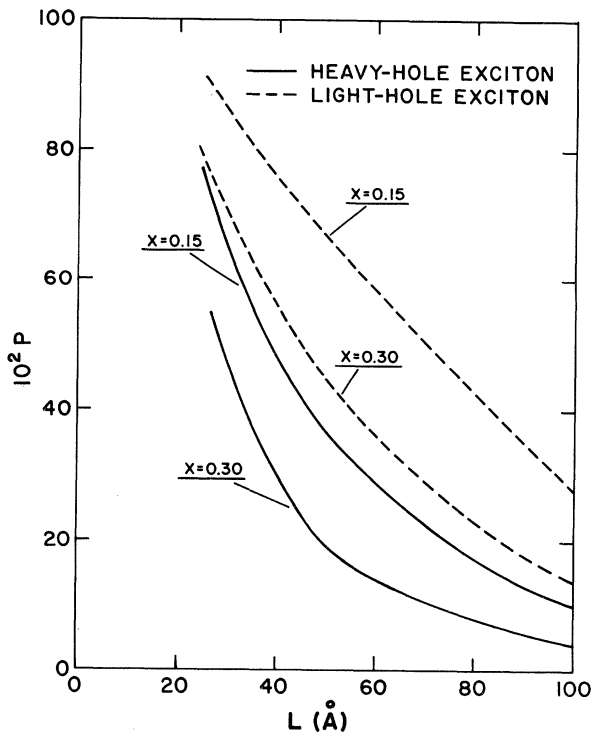

FIG. 4. Probability P of finding the heavy-hole exciton (solid lines) and the light-hole exciton (dashed lines) outside the GaAs layer as a function of the quantum-well size (L) for Al concentrations $x=0.15$ and 0.30.

approach the $Ga_{1-x}Al_xAs$ value for infinite thickness in the case of that exciton system as L is reduced further. It should be pointed out that in our model there are two types of excitons in bulk $Ga_{1-x}Al_xAs$ associated with two different valence bands, whereas in reality there is only one exciton system associated with the fourfold-degenerate valence band. We should note that for a given value of x, the maximum in the binding energy of the $1s$, $2s$, and $2p_{\pm}$ states of a heavy-hole exciton is reached at about the same value of L. The same is true for those states associated with the light-hole exciton, although the value of L at maximum binding energy is larger. This is due to the fact that the probability of finding an exciton outside the well, P, defined as

$$P=1-\int_0^{\infty}\int_{-L/2}^{L/2}\int_{-L/2}^{L/2}|\psi|^2\rho\,d\rho\,dz_e\,dz_h\ , \qquad (13)$$

is the same for all the internal states of a given exciton system. Here we have put the integral of $|\psi|^2$ over all space equal to 1. This probability, however, is larger for a light-hole exciton, which accounts for the larger value of L at which the maximum is reached. In Fig. 4 we plot the variation of $P(h)$ (heavy-hole) and $P(l)$ (light-hole) exciton as a function of L for $x=0.15$ and 0.30. As expected, for a given value of x and L, $P(l)$ is larger than $P(h)$. In addition, for those values of x, which are commonly used in quantum-well structures, $P(h)$ and $P(l)$ are negligible for $L>100$ Å. We also find, that for a given value of x and L the binding energy of the $2p_{\pm}$ state is somewhat larger than that of the $2s$ state. These states, for each exciton system, however, again become degenerate in the two-dimensional limit (V_e and $V_h\rightarrow\infty$ and $L=0$) and in the bulk $Ga_{1-x}Al_xAs$ limit (V_e and V_h finite and $L=0$). Our results for E_{1s} and E_{2s} for infinite potential barriers are very close to those of Miller *et al.*[3] and are also very similar to those of Bastard *et al.*[4] (these authors calculate only E_{1s} using an exciton Hamiltonian with isotropic masses).

It should be pointed out that for a given value of x, $E_{1s}(l)$ is larger than $E_{1s}(h)$ for L greater than a certain critical value L_c, at which they become equal. For values below L_c, $E_{1s}(l)$ is smaller than $E_{1s}(h)$. The value of L_c, of course, depends on the magnitude of x; the larger the magnitude of x, the smaller the value of L_c. For $x=0.3$, for instance, $L_c=50$ Å. The reason for this behavior is fairly easy to understand. The value of $E_{1s}(l)$ is greater than the value of $E_{1s}(h)$ for large L. Both of these values increase as L is reduced. The value of $E_{1s}(l)$ increases less rapidly than the value of $E_{1s}(h)$ as proportionally more of the light-hole–exciton wave function tends to spill over into the surrounding $Ga_{1-x}Al_xAs$ layers, thus reducing the increase in $E_{1s}(l)$. For a certain value of L, which depends on x, the two values become equal and then $E_{1s}(l)$ becomes smaller as L is reduced further. This is in contrast to the behavior of $E_{1s}(h)$ and $E_{1s}(l)$ for infinite potential barriers where $E_{1s}(l)$ is always larger than $E_{1s}(h)$.

Binding energies of $2s$ and $2p_{\pm}$ levels do not show this behavior. The light-hole—exciton binding energies of these two levels are always larger than those of the heavy-hole exciton for values of x and L studied in this work. It may be that the effect occurs at values of $L < 25$ Å .

We shall now compare our results with the existing experimental data. The first determination of the binding energy of the ground state of a heavy-hole exciton in these quantum-well structures was made by Dingle.[1] His absorption measurements for $x=0.2$ suggest a value of about 9 meV for quantum-well size $L=100$ Å. We find that our calculated value of $E_{1s}(h)$, for $x=0.2$ and $L=100$ Å is quite close to the experimental value. Vojak *et al.*[2] studied the photoluminescence spectra of these quantum-well systems ($x=0.3$) grown by metal-organic chemical vapor deposition (MOCVD) and proposed values of 13 and 20 meV for $E_{1s}(l)$ and $E_{1s}(h)$, respectively, for $L=120$ Å. According to these authors, the binding energy of the ground state of the heavy-hole exciton is larger than that of the light-hole exciton, a result contrary to the findings of Miller *et al.*[3] and ourselves. These values are, however, approximately 4 times the bulk values of $E_{1s}(l)$ and $E_{1s}(h)$ (two-dimensional limit) obtained by assuming isotropic masses for the light hole and the heavy hole of $0.080m_0$ and $0.45m_0$, respectively. Recently Miller *et al.*[3] have carried out a systematic detailed study of the excitation spectra of the photoluminescence for both single and multiple quantum wells with $42 < L < 145$ Å for $x=0.37$ at 6 K. They have been able to determine, for the first time, the differences in energy between the ground state and the first excited state ($2s$) of heavy-hole and light-hole excitons in these quantum-well structures. The transition associated with the ground state ($1s$) of the exciton is fairly sharp, whereas the transition associated with the $2s$ state cannot be resolved from the continuum. Miller *et al.*[3] assume that the apparent continuum edge corresponds to the $2s$ state of the exciton. The differences in energy between the $1s$ and $2s$ state Δ_{12} that we calculated for $x=0.37$ for the heavy-hole exciton and for the light-hole exciton agree fairly well with the measured values of Miller *et al.*[3] The experimental values, however, are always somewhat larger than the calculated values, especially for thin wells (less than 100 Å). Miller *et al.* also plot the experimental variation of $E_{1s}(h)$ and $E_{1s}(l)$ as a function of L. Again we find that these values are somewhat larger than the calculated values, especially for thin wells. The largest difference between the calculated values and the experimental values is for the thinest well used (50 Å), and that difference is about 2 meV. This, incidently, is considerably smaller than the line widths of these transitions for this well and therefore may not be regarded as significant. The values of $E_{1s}(h)$, $E_{1s}(l)$, $\Delta_{12}(h)$, and $\Delta_{12}(l)$ calculated using infinite barrier heights, however, agree better with the experimental data for thin wells. This is a little confusing since the potential barrier heights, especially for the light hole, cannot be treated as infinite.

We now briefly comment on several significant features of our calculation. First we discuss the validity of the exciton Hamiltonian described by Eq. (1). Owing to reduction in symmetry along the axis of growth and the presence of energy-band discontinuities at the interfaces, the degeneracy of the valence band at the Γ point is removed. We have assumed that this leads to the formation of two independent exciton systems, the heavy-hole exciton and the light-hole exciton. This is a valid assumption when the contribution of the off-diagonal terms in the exciton Hamiltonian as described by Kohn and Luttinger[7] is small. In addition, the energy separation between the light-hole subband and the heavy-hole subband is required to be much larger than the exciton binding energy in the quantum well. This is clearly a good approximation for large values of x and small values of L. In order to determine how good this approximation is for arbitrary values of x and L one needs to solve for the full Kohn-Luttinger exciton Hamiltonian in a quantum-well situation. This obviously is not an easy task.

The exciton Hamiltonian is derived with the use of the effective-mass approximation. For small values of L (less than 100 Å) and large values of the potential barrier, the energies of the first subbands of electrons and holes (especially the light hole) can be quite significant. This requires including the contributions due to nonparabolicity of the energy bands, especially of the conduction band and the light-hole band. The larger the energy of the first subband, the more important this contribution becomes. Also, as discussed earlier, for small values of L (less than 50 Å) and for small values of x, a considerable part of the exciton wave function is located in the surrounding $Ga_{1-x}Al_xAs$ layers, thus necessitating an appropriate use of the physical parameters pertaining to $Ga_{1-x}Al_xAs$. In our calculations, we have used the same values of m_e, ϵ_0, γ_1, and γ_2 for both semiconductors. This is expected to be a good approximation for most practical quantum-well structures. For the range of values of x (less than or equal to 0.4) the values of these physical parameters are not very different in these two semiconductors. The use of the different physical parameters for $Ga_{1-x}Al_xAs$ will change the values of E and the subband energies. Since the binding energies are the differences between these quantities, they are not expected to be significantly modified.

Finally we briefly comment on the use of the standard connection rules for the envelope functions f_e and f_h at the GaAs-$Ga_{1-x}Al_xAs$ interface. Recently Ando and Mori,[11] and Zhu and Kroemer,[12] have examined this problem for the conduction-band envelope functions and have suggested that for small values of x, the use of these connection rules is justified. The situation with regard to the valence bands is much more complicated. In the GaAs quantum well, the heavy-hole and the light-hole bands are assumed to have sufficient energy separation so that they may be treated as isolated bands from the point of view of exciton formation. In the surrounding $Ga_{1-x}Al_xAs$ layers, the valence band is fourfold degenerate, and thus there is only one exciton system in these layers. In our calculation we have used the standard connection rules for the heavy-hole band and for the light-hole band, thus implying the existence of two isolated bands in $Ga_{1-x}Al_xAs$. For thin wells this is clearly not a good approximation. How this affects the accuracy of our results is not clear. Connection rules for envelope functions associated with valence bands for the GaAs-$Ga_{1-x}Al_xAs$ interface have not yet been studied.

CONCLUDING REMARKS

We have studied the $1s$-, $2s$-, and $2p_{\pm}$-like states of heavy-hole and light-hole excitons in a quantum-well structure consisting of a single slab of GaAs sandwiched between two semi-infinite layers of $Ga_{1-x}Al_xAs$. With the use of a variational approach we have calculated the binding energies as a function of the size of the GaAs quantum well for several values of the heights of the potential barriers. We find that for finite values of the potential barriers the variation of the binding energies of $1s$, $2s$, and $2p_{\pm}$ states as a function of L is essentially very similar; namely, the values of the binding energies increase as L is reduced until they reach their respective maximum values and then begin to decrease. For an infinite barrier, the binding energies increase monotonically as L is reduced, approaching their two-dimensional values at $L=0$. We compare our results with the available experimental data and find good agreement. Finally we have discussed the validity of the approximations made in this calculation.

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