

Flexible approach to exciton binding energies in type-I and type-II quantum wells

Z. S. Piao, M. Nakayama, and H. Nishimura

Department of Applied Physics, Faculty of Engineering, Osaka City University, Sugimoto 3-3-138, Sumiyoshi-ku, Osaka 558, Japan

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We propose a method to calculate the exciton binding energies in type-I and type-II quantum wells by expanding the electron and hole envelope functions into Gaussian-type functions separately. We calculate the binding energies, wave functions, and overlap integrals of the type-I and type-II excitons in an AlAs/GaAs/AlAs single quantum well as a function of the GaAs-layer thickness. This calculation model avoids predetermined "artificial" trial wave functions, and the results exhibit reasonable changes of the exciton properties from the quantum-confined states to the bulk states with the decrease of the GaAs-layer thickness to zero. It is suggested that the binding energy of the type-II exciton exceeds that of the type-I exciton when the GaAs-layer thickness is less than 2 ML because of the wave-function penetration and the large X -valley effective mass. In addition, it is obtained that the overlap integral between the electron and hole of the type-II exciton remarkably decreases with the increase of the GaAs-layer thickness, resulting from the spatial separation of the electron and hole.

I. INTRODUCTION

Excitons in semiconductor superlattices and quantum wells have received considerable attention theoretically and experimentally over the past two decades.¹ The most widely studied case is a type-I structure, where the electrons and holes are confined spatially in the same region. In this structure, it is well known that the confinement effects lead to the enhancement of the binding energies and oscillator strengths of excitons compared with those of the corresponding bulk materials. Recently, there has been growing theoretical interest in the excitons in type-II quantum well structures, where the electrons and holes locate in different regions.²⁻¹³ Actually, type-II structures are as common as type-I structures. As a typical example, for GaAs/AlAs quantum-well structures with the GaAs-layer thickness less than 13 monolayers (ML), the X -electron state of AlAs becomes lower than the Γ -electron state of GaAs, while the lowest-energy state in the valence band is the GaAs Γ -hole state.¹⁴ This results in the spatial separation of the X electron and Γ hole. In a GaAs/AlAs quantum well, the degenerate X valley is split into two states, resulting from the effective mass anisotropy: the X_z state along the growth direction z ($z=[001]$), and the X_{xy} state perpendicular to it. Consequently, there exist three types of excitons: the type-I Γ , type-II X_z , and type-II X_{xy} excitons.

Recent theoretical studies show that the type-II structures possess quite different excitonic properties compared to the bulk and type-I structures.²⁻¹³ Several calculation models have been proposed for the calculations of the binding energies of the type-II exciton states. Early models^{2,4,5,7-10} require prior determinations of the confined envelope functions of the electron and hole in the absence of the Coulomb attraction between them. This kind of method cannot be used to treat the problems like type-II excitons in a single quantum well, because only one particle, electron or hole, is confined in the well layer. Some of the recent works modified this method by considering an effective Coulomb potential created by one confined carrier.^{12,13} However, they used pre-

termined two-dimensional (2D) or three-dimensional (3D) hydrogenlike wave functions as the trial wave functions with some variable parameters. These kinds of trial wave functions seem to work well in some well-width range, but cannot produce correct results in the whole range of the well width, because the dimensionality of the exciton changes between the 2D state and 3D state as a function of the well thickness. As one of the reasonable gauges of the model calculations, it is plausible to expect that the resultant type-I and type-II exciton properties tend to their corresponding bulklike ones for the binding energies and the envelope functions, etc., when the well thickness approaches zero. From this viewpoint, the calculated binding energies in Refs. 12 and 13 for the type-II excitons in an AlAs/GaAs/AlAs single quantum well have been controversial: the tendency approaching the bulklike states in Ref. 12 is fully contrary to that in Ref. 13. Furthermore, there has been no report on the change of the envelope-function form of the type-II excitons from a spatially separated type to a bulklike type with the decrease of the well-layer thickness, which is a measure of reasonableness of the calculations.

In this paper, we propose a flexible approach to the calculations of the binding energies in type-I and type-II single quantum-well structures by separately expanding the electron and hole envelope functions to Gaussian-type functions. As an example, we calculate the binding energies, envelope functions, and overlap integrals of the type-I Γ , type-II X_z , and type-II X_{xy} excitons in an AlAs/GaAs/AlAs single quantum well. The calculations exhibit conceptually reasonable tendencies from the quantum confined states to the bulk states of the excitons with the decrease of the GaAs layer thickness to zero. In Sec. II, we describe the formalism, in which we express the electron and hole envelope functions in Gaussian-type functions separately, then determine the expansion coefficients variably. In Sec. III, we give the results of the exciton binding energies, envelope functions of the electron and hole, and overlap integrals, and discuss the relations of them with the GaAs-layer thickness. Conclusions are presented in Sec. IV.

II. CALCULATION MODEL

As an example, we consider the type-I Γ , type-II X_z , and type-II X_{xy} excitons in an AlAs/GaAs/AlAs single quantum well. The effective-mass Hamiltonian H for an electron of mass m_e and a hole of mass m_h is

$$H = -\frac{\mu}{m_h} \nabla_h^2 - \frac{\mu}{m_e} \nabla_e^2 - 2Q + V_e(z_e) + V_h(z_h), \quad (1)$$

where we take the z axis as the growth direction. We denote the electron and hole coordinates by the subscripts e and h . The unit of length is the exciton Bohr radius $a_B = \epsilon \hbar^2 / e^2 \mu$, and the unit of energy is the exciton Rydberg $R = e^2 / 2\epsilon a_B$. Here, ϵ is the static dielectric constant of the background materials and $\mu = m_e m_h / (m_h + m_e)$ is the reduced mass of the electron and hole. $V_e(z_e)$ and $V_h(z_h)$ are the potentials for the electron and hole, respectively, $Q = 1/r_{eh}$ is the Coulomb interaction, and r_{eh} is the distance between the electron and hole. For the sake of simplicity, we neglect the image charge effects and the interactions between the heavy hole and light hole. Because it is impossible to obtain analytically the binding energies of the excitons confined in type-I and type-II quantum wells, most of the calculations have used variational calculation techniques. In variational calculations, a main procedure is to choose appropriate trial wave functions, and this may remarkably influence all of the results. Because of the complexity of the geometric characteristics of the type-II excitons, it is unclear whether the normally used hydrogenlike wave functions with the quantum-well-confinement envelope functions or trivial modifications of them work well in the whole range of the well thickness. To avoid all these ambiguities, in our calculations we expand the unknown electron and hole envelope functions to Gaussian-type functions separately as follows (in cylindrical polar coordinates):

$$\Phi(\rho, z_h, z_e) = \sum_{i,j,k} S_{i,j,k} e^{-A_i \rho^2} e^{-B_j z_e^2} e^{-C_k z_h^2}, \quad (2)$$

where ρ is the relative coordinate of the electron and hole in the x - y plane, $S_{i,j,k}$ is the expansion coefficients to be determined, and A_i , B_j , C_k are variable parameters that will be discussed in detail in the next section. Aldrich and Greene applied a similar Gaussian basis to solve the problem of a hydrogen atom in a magnetic field.¹⁵ We calculate the eigenvalues and eigenfunctions of the Hamiltonian H by using a standard variational approach. It is expected that the flexible nature of the linear combination of the Gaussian-type functions produces reasonable envelope functions for the electron and hole, and may be very effective to treat complex exciton systems in which the well-working trial wave functions are unknown. The exciton binding energy E_b is defined as

$$E_b = E_e + E_h - E, \quad (3)$$

where E_e (E_h) is the quantum confinement energy in the potential well V_e (V_h) for the electron (hole) in the absence of the Coulomb interactions. In the AlAs/GaAs/AlAs single quantum well, we must let $E_e = 0$ for the type-II X_z and type-II X_{xy} excitons, because the electrons locate in the semi-infinite AlAs layers. The electron and hole probability functions in the z direction are given by

$$P(z_{e,h}) = \int_{-\infty}^{\infty} \int_0^{\infty} \Phi(\rho, z_h, z_e)^2 2\pi \rho d\rho dz_{h,e}. \quad (4)$$

From these probability functions, we can see the distributions of the electron and hole in the z direction. The squared overlap integral between the electron and hole is calculated by the following formula:

$$J = \left| \int_{-\infty}^{\infty} \Phi(0, z, z) dz \right|^2. \quad (5)$$

This overlap integral represents the probability of finding the electron and hole at the same point, which is closely related to the oscillator strength of the exciton.

III. RESULTS AND DISCUSSION

In the numerical calculations, we use the same values of the band discontinuities in Refs. 12 and 13: $\Delta_{e,\Gamma} = 1.068$ eV, $\Delta_{e,X} = -0.197$ eV, and $\Delta_{h,\Gamma} = 0.457$ eV. The effective masses for the Γ and X electrons are $m_{e,\Gamma} = 0.067m_0$, $m_{e,Xz} = 1.1m_0$, and $m_{e,Xxy} = 0.19m_0$. The heavy-hole effective mass is $m_h = 0.37m_0$. These values are also the same as those in Refs. 12 and 13, except for $m_{e,Xz} = 1.1m_0$,¹⁶ because an extremely large value of $4.1m_0$ is used in the calculations. The static dielectric constant is $\epsilon = 12.53$.¹² One-monolayer thickness of GaAs and AlAs is taken to be 2.83 \AA . The choice of the appropriate basis functions is important to obtain reasonable results in the numerical calculations. In our calculations, we use three sets of Gaussian-type functions for z_e , z_h , and ρ coordinates as represented by Eq. (2), and each set consists of five Gaussian-type functions. The parameters of A_i , B_j , and C_k ($i, j, k = -2, -1, 0, 1, 2$) have the forms of geometrical progressions as follows:

$$A_i = A_0 q^i, \quad B_j = B_0 q^j, \quad C_k = C_0 q^k, \quad (6)$$

where A_0 , B_0 , C_0 are variational parameters used to minimize the total energy E , and q is the proportional constant for the geometrical progressions. Here, unlike the expansion of a hydrogenlike envelope function, the basis wave functions of the excitons are separable for the electron and hole in the z direction, and their forms will be determined by the minimization procedure of the total energy E . The wave functions determined in this way are very flexible, and free from predetermined stereotypes of the trial wave functions. It is noted that in the absence of the Coulomb attraction Q between the electron and hole, the results agree with the quantum confinement energies of the electron and hole calculated using an ordinary envelope-function approximation.¹⁷ In addition, when we neglect the quantum potential V_e and V_h , the exciton binding energies in the bulk crystals are obtained. The above results in the two limit cases support the correctness and flexibility of the present calculations. Using $q = 3$, we obtain satisfying results of the binding energies and envelope functions. Other values of q around 3 give similar results. The values of A_0 , B_0 , and C_0 are varied between 0 and 10^4 to minimize the total energy E . The further increase of the number of basis functions affects little the numerical results.

Figure 1 shows the calculated binding energies of the type-I Γ , type-II X_z , and type-II X_{xy} excitons in the AlAs/

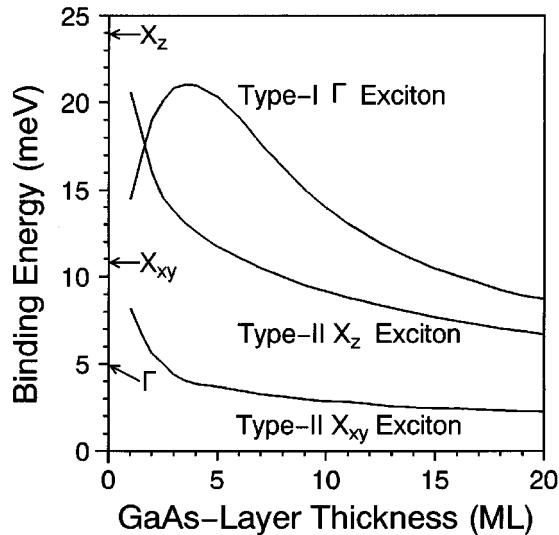


FIG. 1. Calculated binding energies of the type-I Γ , type-II X_z , and type-II X_{xy} excitons in an AlAs/GaAs/AlAs single quantum well vs the GaAs-layer thickness.

GaAs/AlAs single quantum well versus the GaAs-layer thickness, where the arrows at the left side indicate the binding energies of the corresponding bulk excitons. From Fig. 1, it is evident that the exciton binding energies of all three excitons exhibit reasonable tendencies to the values of the bulk excitons with the decrease of the GaAs-layer thickness. Comparing the present results with those of Refs. 12 and 13, it is noted that the calculated binding energies show similar behavior when the GaAs-layer thickness is larger than 5 ML. This suggests that the calculation models scarcely affect the exciton binding energies in such a thickness range. However, when the GaAs-layer thickness is less than 5 ML, the exciton binding energies obtained in the present work are remarkably different from those in Refs. 12 and 13, which do not exhibit reasonable tendencies to approach to the values of the corre-

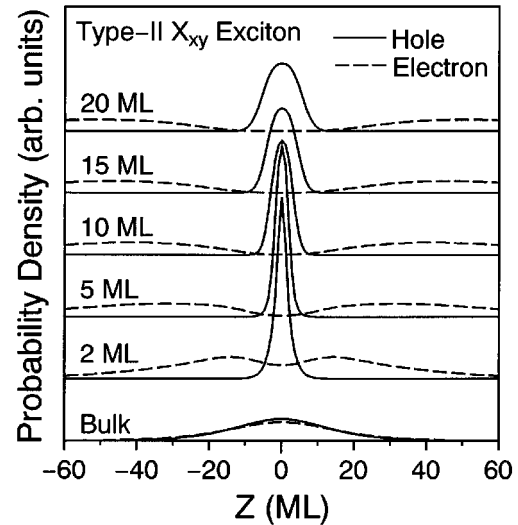


FIG. 3. Hole and electron probability functions of the type-II X_{xy} exciton in an AlAs/GaAs/AlAs single quantum well at various GaAs-layer thicknesses.

sponding bulk excitons. The difference seems to be caused by the adiabatic approximation¹⁸ of the effective Coulomb potential due to the confined hole used in Refs. 12 and 13. As the GaAs-layer thickness approaches zero, the envelope function of the hole confined in the GaAs layer tends to penetrate into the AlAs layers; therefore, the influence of the electron Coulomb potential on the hole envelope function cannot be neglected. At the limit of the bulk state, there exists no quantum-confinement state, so that the adiabatic approximation of the effective Coulomb potential itself has no meaning. Thus, the models used in Refs. 12 and 13 are inadequate to calculate the exciton binding energies in the case of the ultrathin GaAs layer.

Next, we discuss the envelope functions of the electron and hole. Figures 2, 3, and 4 show the probability density of the envelope functions of the type-II X_z , type-II X_{xy} , and

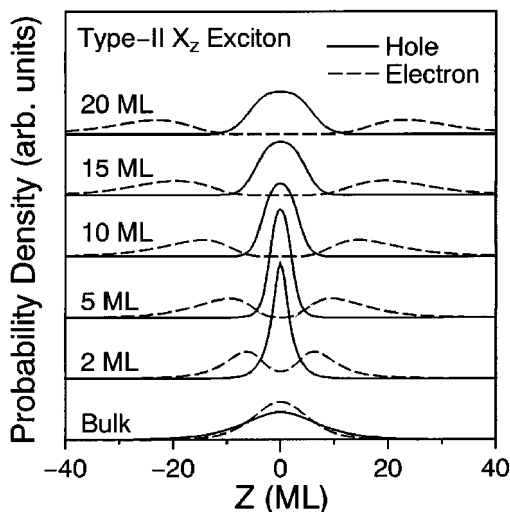


FIG. 2. Hole and electron probability functions of the type-II X_z exciton in an AlAs/GaAs/AlAs single quantum well at various GaAs-layer thicknesses.

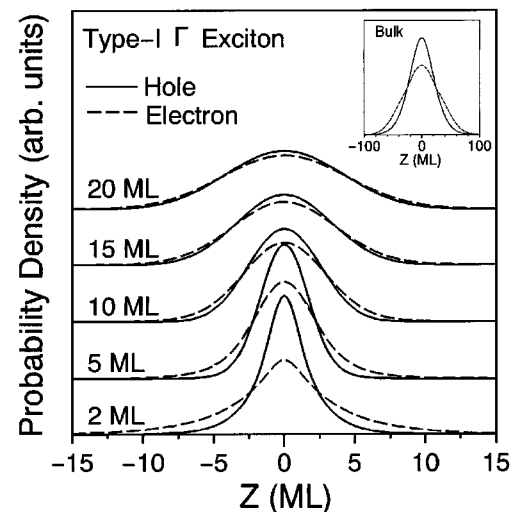


FIG. 4. Hole and electron probability functions of the type-I Γ exciton in an AlAs/GaAs/AlAs single quantum well at various GaAs-layer thicknesses. The inset represents the probability functions of the bulk Γ exciton.

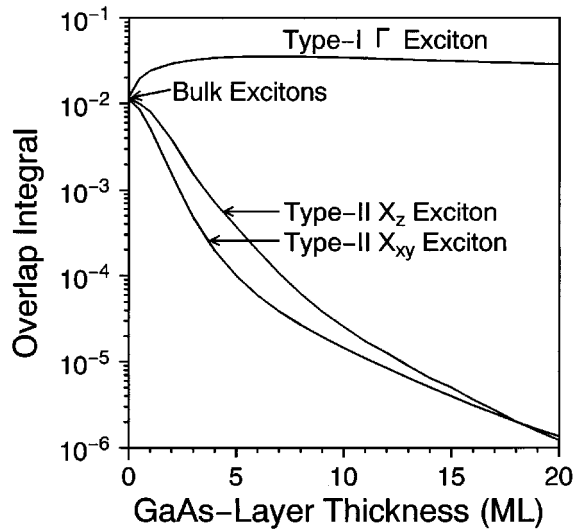


FIG. 5. Squared overlap integrals between the hole and electron envelope functions of the type-I Γ , type-II X_z , and type-II X_{xy} excitons in an AlAs/GaAs/AlAs single quantum well vs the GaAs-layer thickness.

type-I Γ excitons, respectively, at various GaAs-layer thicknesses. From Figs. 2 and 3, it is found that the electron spreads broadly in the AlAs layers. Since the effective mass of m_{e,X_z} is approximately 6 times $m_{e,X_{xy}}$, the electron envelope function of the type-II X_z exciton locates more closely to the interfaces in comparison with that of the type-II X_{xy} exciton. These results are similar to the results of Bellabchara *et al.*¹³ Furthermore, the systematic results of the envelope functions show that the electron and hole in the type-II excitons are distributed symmetrically in both sides of the GaAs layer. The symmetric properties of the electron envelope functions shown in Figs. 2 and 3 are similar to the results reported by Zimmermann and Bimberg.¹² However, it should be noted that when the GaAs layer becomes very thick, the distributions of the electron and hole are expected to be asymmetric: The electron locates preferably in one side, and the envelope function of the hole inclines to the side of the electron to form a minimum energy state. A careful discussion of this problem is beyond the scope of this paper.

Figure 5 shows the squared electron-hole overlap integrals of the type-I Γ , type-II X_z , and type-II X_{xy} excitons as a function of the GaAs-layer thickness in the AlAs/GaAs/AlAs single quantum well. It is evident that the overlap integrals of the type-II X_z and type-II X_{xy} excitons change from 10^{-6} to 10^{-2} with the decrease of the GaAs-layer

thickness, while that of the type-I Γ exciton changes slightly. It is noted that the remarkable changes of the overlap integrals of the type-II excitons are a typical aspect of the spatial separation of the electron and hole envelope functions. When the GaAs-layer thickness approaches zero, the overlap integrals of the three excitons approach the values of the corresponding bulk excitons, which are close to each other. On the other hand, in Ref. 13, the calculated overlap integral of the type-II X_z exciton is one order larger than that of the type-II X_{xy} exciton when the GaAs-layer thickness tends to zero. This indicates that the appropriate choice of the trial wave function is very important to obtain accurate results, and shows the considerable improvement of the present calculations to earlier ones. The overlap integrals have close relations with the oscillation strengths of the excitons. The Bloch-function term of the oscillator strength of the type-II X_z exciton is finite because of a Γ -X mixing in the conduction band,¹⁹ while the type-II X_{xy} exciton is almost pure indirect because the X_{xy} wave vector does not match the growth direction. The resultant overlap integrals suggest that the type-II X_z exciton in the single quantum well may be detectable in very thin GaAs-layer thickness.

IV. CONCLUSIONS

In the calculations for the binding energies of the type-I Γ , type-II X_z , and type-II X_{xy} excitons in an AlAs/GaAs/AlAs single quantum well, we expand the electron and hole envelope functions into Gaussian-type functions separately to avoid artificially predetermined trial wave functions. When the GaAs-layer thickness approaches zero, the calculated results of the type-I Γ , type-II X_z , and type-II X_{xy} excitons show excellent agreement with the corresponding bulk states for the binding energies, wave functions, and overlap integrals between the electron and hole. This demonstrates the validity of our calculations for the type-I and type-II excitons. We note that for a system where “well-working” variational wave functions are still unknown, it is more plausible to use flexible trial wave functions like those used in the present calculations. In the AlAs/GaAs/AlAs single quantum well, the binding energy of the type-II exciton may be larger than the type-I Γ exciton for narrow GaAs-layer thickness, because of the large X-valley effective mass of the electron. The overlap integrals between the electron and hole envelope functions for the type-II excitons decrease drastically with the increase of the GaAs-layer thickness, resulting from the spatial separation of the electron and hole. This model can be applied to calculate type-II multiple quantum wells and superlattice systems, and also can include external perturbations such as a magnetic field.

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