Study of excitons in an arbitrarily shaped GaAs/$Al_{0.3}Ga_{0.7}As$ single quantum well in the presence of static transverse electric field

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A method is developed to calculate exciton parameters in an arbitrarily shaped quantum well by using numerical methods in conjunction with the variational method. We solve the Schrödinger equation of the relevant system both by the Monte Carlo method and by the ordinary differential equation solving method, and find that the two approaches yield similar results. Our technique is readily applied to arbitrarily shaped GaAs/AlGaAs quantum wells, and allows us to study the variation of exciton parameters with respect to transverse electric field. We report the results for exciton binding energies, electron and hole tunneling rates, and exciton volumes, which are essential to predicting electro-optical properties of quantum well. Results are presented for the square, graded, and asymmetric quantum wells, and the effects of the shape of the well on exciton properties are discussed. We find that there is only small dependence of the exciton binding and emission energy on the well shape, but a strong dependence of the electron and hole tunneling rates and hence of the dynamical aspects of excitonic properties on the well shape.

I. INTRODUCTION

Epitaxial crystal growth techniques such as molecular-beam epitaxy have made it possible to grow heterostructures with controlled thickness and sharp interfaces. The ability to confine carriers in quantum-well structures has generated considerable interest due to novel physics of quasi-two-dimensional systems. The GaAs/AlGaAs quantum wells have been extensively studied from the point of view of exciton physics.[1,2] Considerable effort has recently focused on understanding the excitonic phenomenon in presence of a transverse electric field due to potential applications for light modulation and optical switching.[3–8]

The confining effect in a quantum-well structure gives not only the remarkable persistence of exciton transitions to very high electric field (~ 50 times the classical ionization field in bulk material), but also several times as strong excitonic transitions as that of bulk material.[9] Because of the enhanced exciton binding energy and the electric field dependence of excitonic transitions, the quantum well has important applications as a modulator and an optical switch. In electro-optical device application the speed and the efficiency of device operation are critical factors. The device speed is directly related to the quenching speed of luminescence, which depends on the electron and hole tunneling rates in the presence of the applied field. The electron and hole tunneling rates can be tailored independently by use of asymmetric quantum-well structures. The efficiency of these devices is dependent on the exciton absorption coefficient and linewidth. In considering inhomogeneous line broadening, exciton volume plays an important role.[10] This parameter is also useful in getting nonlinear optical effects, exciton bleaching effects at high photon intensity, and calculating absorption coefficents.

The exciton parameters in quantum wells can be tailored by changing the shapes and sizes of the wells. One needs to develop a formalism that can handle "arbitrarily shaped quantum wells" with ease. Previous attempts have focused on understanding the exciton problem in the square well and have not dealt with arbitrarily shaped quantum wells. Bastard and Green *et al.* have used the variational method to solve the exciton problem in a square well in the absence of the electric field.[11,12] Miller *et al.* have considered an exact solution for a one-dimensional infinite well with static transverse electric field in terms of resonances. Very recently, the infinite barrier well problems even with strong electric field are solved numerically by considering resonances represented by Airy functions.[13]

We have addressed the exciton problem using the following approach: (i) The solution of the electron (hole) subband levels are obtained by either solving differential equation numerically or a variational technique based on Monte Carlo methods; (ii) the solution of the exciton problem is obtained by a variational method. This approach is readily applied to an arbitrarily shaped quantum well in the presence of electric field. In this paper, we apply this numerical technique to electron and hole quasibound state in GaAs/$Al_{0.3}Ga_{0.7}As$ quantum wells in the presence of a transverse electric field. Since graded and asymmetric quantum wells are promising candidates for their tailoring potential, we deal with these two typical cases as well as the abrupt quantum well structure. In Sec. II we describe the exciton problems and the relevant exciton parameters, and in Sec. III show the result of our calculation. We conclude in Sec. IV.

II. THE EXCITON PROBLEM

The Hamiltonian of an exciton in an electric field in a GaAs slab surrounded by two GaAlAs layers grown along the (001) direction can be expressed as[14,15]

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$$H = -\frac{\hbar^2}{2\mu_\pm}\left(\frac{1}{\rho}\frac{\partial}{\partial\rho}\rho\frac{\partial}{\partial\rho} + \frac{1}{\rho^2}\frac{\partial^2}{\partial\phi^2}\right) - \frac{\hbar^2}{2m_e}\frac{\partial^2}{\partial z_e^2} - \frac{\hbar^2}{2m_h}\frac{\partial^2}{\partial z_h^2} - \frac{e^2}{\epsilon|r_e - r_h|} + V_{ew}(z_e) + V_{hw}(z_h) + eEz_e - eEz_h . \quad (1)$$

Here we use the effective mass approximation, and neglect the band structure effects as well as the effective mass mismatch between well and barriers, which is valid when well size is relatively large (> 50 Å). Here, m_e is the effective mass of the conduction-band electron and $m_\pm$ is the heavy (+) or light (−) hole mass along the z direction. These two exciton systems are nondegenerate since the degeneracy of the valence band of GaAs is removed due to reduction in symmetry along the z axis. $\mu_\pm$ 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 band parameters.[16] In Eq. (1) we have ignored the kinetic energy of the exciton center of mass since we are only interested in the state which can be accessed optically. We have also assumed the same values for the static dielectric constant in the two semiconductors. The appropriate dielectric constant is taken to be $\epsilon = \sqrt{\epsilon_\omega \epsilon_b}$, where $\epsilon_\omega, \epsilon_b$ are the dielectric constants of the well and barrier, respectively. This comes from a consideration of the equivalent dielectric continuum for thin wells and barriers of equal width. The potential wells for conduction electron $V_{ew}(z_e)$ and for holes $V_{hw}(z_h)$ can be arbitrarily confining potentials, which give rise to bound or quasibound states. The quantities eEz_e and eEz_h are the additional potential energy terms for the electron and hole due to the electric field. We solve the problem for the graded potential well and asymmetric wells in addition to the square-well case, since the graded and asymmetric cases cannot only show the versatility of the numerical techniques but also offer comparision with the square-well case where much of the theoretical and experimental effort has focused. It is clearly impossible to solve directly for the Schrödinger equation associated with exciton Hamiltonian [Eq. (1)] either analytically or numerically. Therefore, we assume the trial wave function can be factored as[11]

$$\psi = \phi_e(z_e)\phi_h(z_h)\exp(-\rho/\lambda) , \quad (2)$$

where $\phi_e(z_e)$, $\phi_h(z_h)$ are achieved exactly by applying numerical methods to one-dimensional potential wells, which describe the z-axis directional motion of electron and holes. The exponential factor is an envelope function; we choose a simple 1-s like orbital for in-plane radial motion. Previous calculations have indicated that this simple trial function is accurate for the thickness of our interest (~ 100 Å).[11] Here, λ is trial parameter of variational method for calculating exciton binding energy. To determine $\phi_e(z_e)$ and $\phi_h(z_h)$, we use numerical techniques. Since we have already described the Monte Carlo approach,[17] we briefly outline the ordinary differential equation solving approach.

Initial and final conditions are chosen in a region − L to + L surrounding the well [$\phi(-L) = 0, \phi'(-L) = 0$ and $\phi(+L) = 0, \phi'(+L) = 0$, respectively]. Here L has a value larger than the extent of the function (we use $L \sim 2.5$ W). A quasibound level is then determined by solving the Shrödinger equation:

$$-\frac{\hbar^2}{2m}\frac{\partial^2}{\partial x^2}\phi + [V(x) \pm eEx]\phi = E\phi . \quad (3)$$

It is important to realize that both these methods are straightforward and do not assume any starting wave functions.

We have applied these techniques to calculate the electron and hole states and the associated wave functions. We assume that the total band discontinuity of 360 meV (for $Al_{0.3}Ga_{0.7}As$/GaAs system) across the interface is distributed in conduction and valence bands in the two extreme ratios of 60% and 40% or 85% and 15%, respectively. In our calculations, we assume the electron, heavy-hole, and light-hole masses to be $0.067m_0$, $0.35m_0$, and $0.08m_0$ (m_0 is free electron mass).[2] The reduced masses in the x-y plane for heavy hole and light hole are $0.04m_0$ and $0.05m_0$, respectively. Due to anisotropic nature of the kinetic energy, the reduced mass associated with the heavy-hole band ($J_z = \pm 3/2$) is smaller than that of light-hole band ($J_z = \pm 1/2$). The variation of the electron and hole subband energies ($E_e + E_h$) with electric field, is also calculated and used to get the exciton emission energy.

A. Exciton binding energy

If a quantum well is exposed to light with energy above the effective band gap, electron-hole pairs are produced. The absorption energy is given by

$$E_{ph} = E_e + E_h + E_g - E_b , \quad (4)$$

where E_e and E_h are electron and hole subband energies in a quantum well, respectively, E_g is the band gap of the well material (i.e., GaAs), and E_b is exciton binding energy. The electron and hole pairs form excitons due to the Coulombic interaction. In the presence of an electric field, the electrons and holes are spatially separated so that electron hole interaction is reduced which causes E_b to decrease. However, the change of E_b which is controlled by the overlap of the electron and hole wave functions is an order of magnitude smaller than the change produced in the values of E_e and E_h. To calculate the exciton binding energy by variational method, we evaluate the expectation value of Hamiltonian [Eq. (1)][12]:

$$E = \frac{\iiint \Psi^* H \Psi dz_e\, dz_h\, \rho\, d\rho}{\iiint \Psi^* \Psi dz_e\, dz_h\, \rho\, d\rho} \quad (5)$$

and minimize E as a function of trial parameter λ. The binding energy E_b is obtained by subtracting E_{min} from total ground state energy of electron and hole.

B. Tunneling rate of electron and hole

In presence of a transverse electric field, the electron and hole states of the quantum well are only quasibound states and have a nonzero probability of tunneling out of the well. These tunneling rates are important because (a) the electron and holes that tunnel out cannot contribute to the exciton collapse and hence to photoluminescence and (b) the tunneling, which effectively removes the space charge from the quantum well and consequently "resets" the well in the optical modulation of a pulse stream of an optical signal, can eventually control the speed of optical modulation.

We have calculated the electron and hole subband levels and the associated energies and wave functions which allow us to calculate the tunneling probabilities. The tunneling rates can be evaluated either by WKB method (for the Monte Carlo approach which is variational in nature) or by the wave function in the numerical solution of the Schrödinger equation.[18]

C. Exciton volume

It is important to determine the spatial extent of the exciton in the quantum well. This information is important in understanding the broadening of the exciton line due to structured fluctuations in the quantum well. It is also important in calculating the light intensity at which excitons start overlapping with each other and causing very strong optical nonlinearities.

The total exciton volume is given by

$$\Theta = \iiint \overline{\omega}\rho \, d\rho \, d\phi \, dz |\Psi|^2 \rho^2 |z| |\cos\phi \sin\phi| . \qquad (6)$$

Here, $\overline{\omega} = 1$.

It is also useful to consider the fraction of the exciton volume in the barrier region versus the volume inside the well. The exciton in the barrier sees the material quality of the barrier which often is an alloy and causes broadening of the excitonic transition linewidth. With this motivation we have calculated the exciton volume in the barrier region and inside the well. In the case of the graded wells the inside volume is calculated by using ω; $\omega(z) = [v(B) - v(z)]/v(B)$, where $v(z)$, $v(B)$ are the electron or hole potentials at the point z and deep in the barrier in the absence of electric field.

III. RESULTS AND DISCUSSION

In this section we will present the results of our calculations and compare the results for the various shaped quantum wells. The exciton binding energy results for the 100 Å (denoted by 1) and 70 Å (denoted by 2) square wells are shown in Fig. 1. In this calculation we used total band-gap discontinuity of 360 meV. To study the effect of band-edge discontinuity distribution, the results are presented for two extreme cases of 60:40 (case a) and 85:15 (case b). We note that considerable controversy has been present on the exact value of the discontinuity, the earlier work of Dingle[19] suggested a 85:15 value while recent work[20,21] suggests a much larger valance-band ratio. The variation of the exciton binding energies for the heavy hole (solid line) and light hole (dotted lines) with the applied transverse electrical field are displayed.

The results show that, in general, the light-hole exciton has somewhat higher binding energy than heavy-hole case in larger wells and its binding energy decrease more rapidly with electric fields. This occurs due to a greater separation of the electron and light-hole wave functions as the field is increased. For the 70-Å well in the 85%:15% case, the heavy-hole exciton binding energy is larger than that of the light hole. This result agrees with other worker's results who solved the zero field problem. We also find that hole state is unbounded (i.e., there is no quasibounded state) when elecric field intensity approaches 40 kV/cm in the 85%:15% case. These results for the 85%:15% cases are different from the high-field results of Bastard *et al.*,[22] which were based on the variational method. The reason for the 85%:15% cases having difficulty to form a bound state is that the hole confinement is very poor at high fields due to the relatively low barriers. In Fig. 2 we show the results for the exciton binding energies for asymmetric quantum wells. These kinds of wells

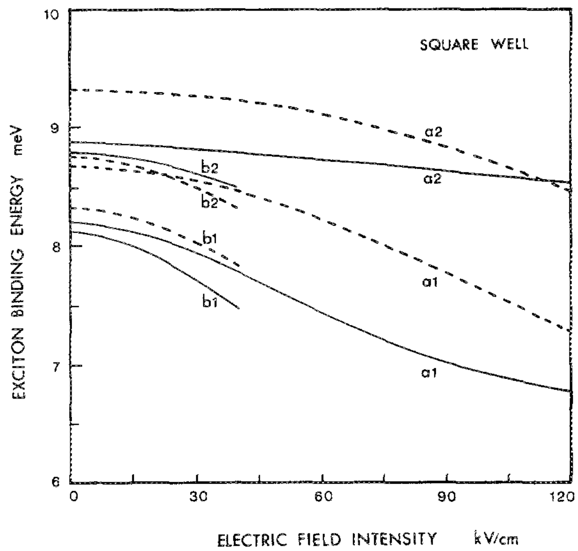

FIG. 1. Exciton binding energy vs electric field in square wells. Solid line is for heavy hole and dotted line for light hole. "1" stands for the well width of 100 Å, and "2" for 70 Å. "a" stands for 60%:40% distribution of band discontinuity over conduction band and balance band, respectively, and "b" for 85%:15%.

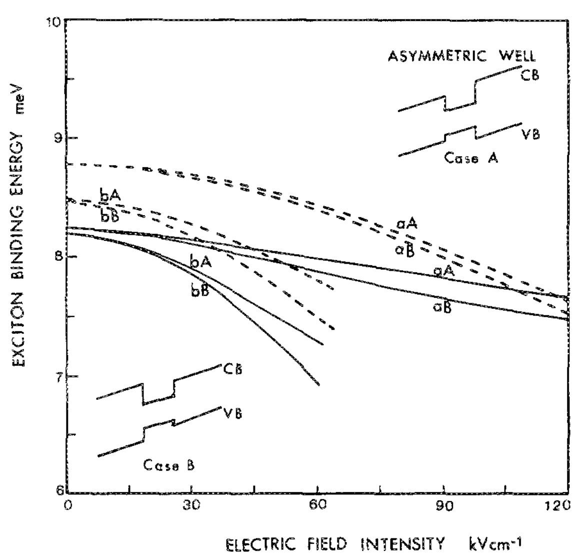

FIG. 2. Exciton binding energy vs electric field in asymmetric wells. Case A has 1.5 times higher barrier in low potential side than that of the opposite side. Case B has 1.5 times higher barrier in high potential side. Solid line is for heavy hole, and dotted line for light hole. "a" stands for 60%:40% distribution of band discontinuity over conduction band and balance band, respectively, and "b" for 85%:15%.

have potential applications in devices such as quantum-well modulators because as will be shown later, the electron- and hole-tunneling rates can be independently tailored. We show results for two cases of the asymmetric wells—case A where the field is in the direction of the lower barrier to higher barrier and case B where the situation is reversed. We have assigned the total band-gap discontinuity of the lower barrier side to be 360 meV, and that of the higher barrier to be 1.5 times that of the lower barrier. To compare with the square-well case, the well width is chosen to be 100 Å. The results show that the binding energy is greater and its decrease with electric field is less than that of the square wells because of the higher barrier. The binding of the case A decreases slower with electric field than that of the case B. This again is due to the stronger hole confinement. Exciton survives until 60 kV/cm for both of A and B cases even in the 85%:15% case. In Fig.3 we show results for the graded well case. We have chosen the same band-gap discontinuity as in Fig.1, but graded the potential linearly such that the narrow part of the well is 80 Å and the wide part is 120 Å giving an average of 100 Å. We find from a comparison of Fig.1 and Fig.3 that the graded well with the average width of 100 Å has almost identical exciton binding energy as the 100 Å square well. This is especially true for the electric fields although even at high fields the difference are less than 5%.

From the above results it appears that the exciton binding energy is not very sensitive to the well shape. However, since the exciton binding energy is only one of the important excitonic parameters, it is important to see if this conclusion holds for other excitonic properties. We have also calculated the results for the exciton emission energies as a function of electric field as given by Eq. (4). Note that the electron- and hole-subband energies decrease much more rapidly with electric field than the exciton binding energy. We find that for the 100 Å well for the 60:40 case, the emission energy changes by ~ 30 meV for the heavy-hole exciton and ~ 20 meV for the light-hole exciton case. When the electric field changes by 120 kV/cm, the change for the 70-Å well are comparatively very small. This is because there is a much stronger overlap between the electron and hole for the narrower quantum well. The results for the asymmetric and graded quantum well are very similar and display little dependence on the well shape.

In Figs. 4–6 we present results for the tunneling rate of electron and hole for the various quantum wells. In Fig.4 we show the results for the square-well case. Several features are apparent from these results. The light-hole tunneling rate is orders of magnitude higher than the heavy-hole case. Since the exciton collapse times are $\sim 10^{-9}$ s, we find that light-hole excitonic photoluminescence will be quenched by 50% at ~ 10 kV/cm for 85:15 discontinuity and ~ 50 kV/cm for 60:40 discontinuity. For the heavy-hole excitonic transitions, the quenching fields are approximately 30 and 90 kV/cm, respectively. The result for the asymmetric wells are shown in Fig.5. The various symbols are explained in the figure caption. As expected, the tunneling rates for the electron in A-type barrier and holes in B-type barriers are the same as in Fig. 5. However, the electron tunneling rate has been decreased by orders of magnitude in case B, while the hole tunneling rate has been decreased by orders of magnitude in case A. As shown earlier, the changes in exciton binding and emission energies for these different well shapes are negligible. Thus, the electron and hole tunneling rates are independently adjusted by asymmetric wells. In Fig.6 we show the tunneling rates for the graded well. We see that

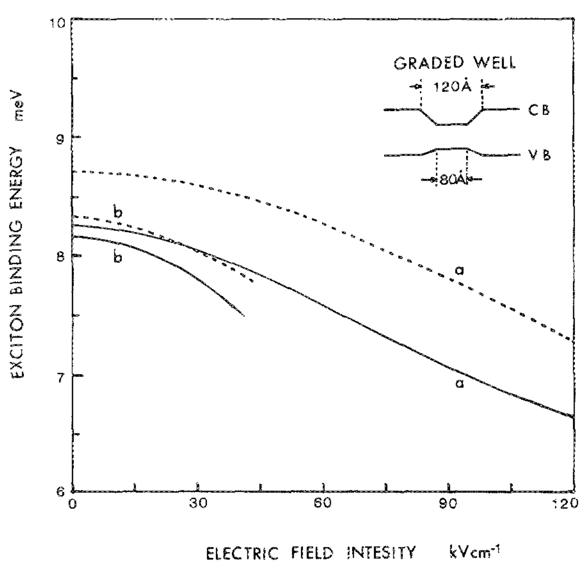

FIG. 3. Exciton binding energy vs electric field in graded well. Solid line is for heavy hole, and dotted line for light hole. "a" stands for 60%:40% distribution of band discontinuity over conduction band and balance band, respectively, and "b" for 85%:15%.

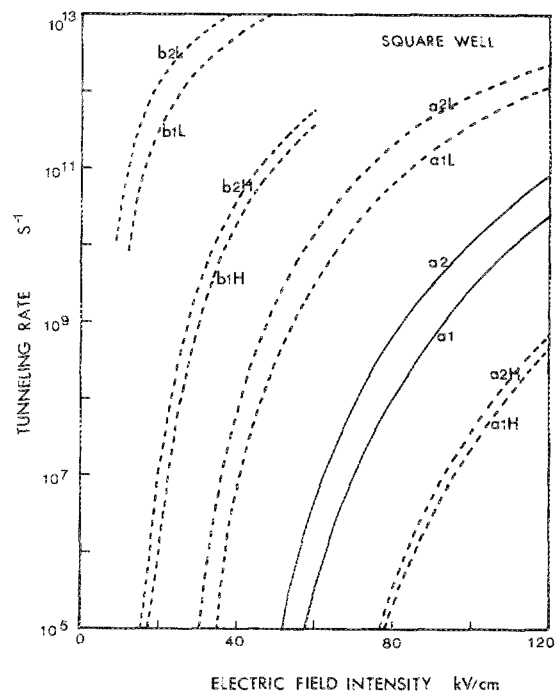

FIG. 4. Electron and hole tunneling rates vs transverse electric field in square wells. Solid line is for electron and dotted line for hole. H means heavy-hole case, L light-hole case. "1" stands for the well width of 100 Å, and "2" for 70 Å. "a" stands for 60%:40% distribution of band discontinuity over conduction band and balance band, respectively, and "b" for 85%:15%.

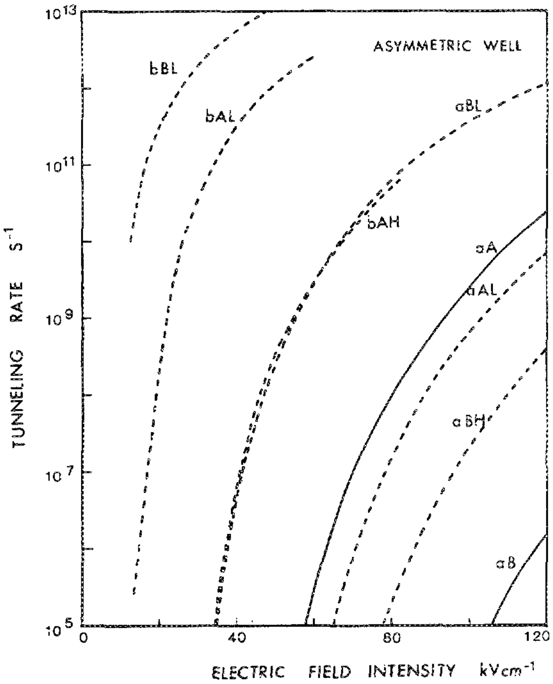

FIG. 5. Electron and hole tunneling rates vs transverse electric field in asymmetric wells. Case A has 1.5 times higher barrier in low potential side than that of the opposite side. Case B has 1.5 times higher barrier in high potential side. Solid line is for electron and dotted line for hole. H means heavy-hole case, L light-hole case. "a" stands for 60%:40% distribution of band discontinuity over conduction band and balance band, respectively, and "b" for 85%:15%.

both the electron and the hole rates have dramatically increased and substantial photoluminescence quenching is to be expected even at very low electric fields.

Finally, in Figs. 7–9 we present information on the exciton shape in the different wells. The excitonic volume inside and outside the wells is shown as a function of electric field. In all cases, as expected, the exciton volume increases as the electric field increases. In the 85:15 discontinuity case, the exciton volume increases dramatically due to the hole state leaking out of the well. For the 70-Å quantum well, the exciton volume changes for the 60:40 discontinuity are negligible, but for the 100 Å case, the volume increases by ~50% as the field increases from 0 to 100 kV/cm. In Fig. 8, we show the results for the exciton volume for the asymmetric wells for the 60:40 discontinuity. For the B-type well, the exciton volume increases rapidly with applied field, while for the A-type well the change is much slower. The outside volume of the type-A well showed little decrease in low electric field. This decrease is caused by asymmetry, which can confine exciton in smaller space by help of electric field. In Fig.9 we show the results for the graded well. In this case there is a considerably higher volume of the exciton in the AlGaAs region. For example, there is 30% higher light-hole exciton volume in the barrier compared to the square-well case. This value for the heavy-hole exciton is ~70%. From these results it is clear that the exciton volume is quite sensitive to the electric field and can to some extent be controlled by the shape of the well.

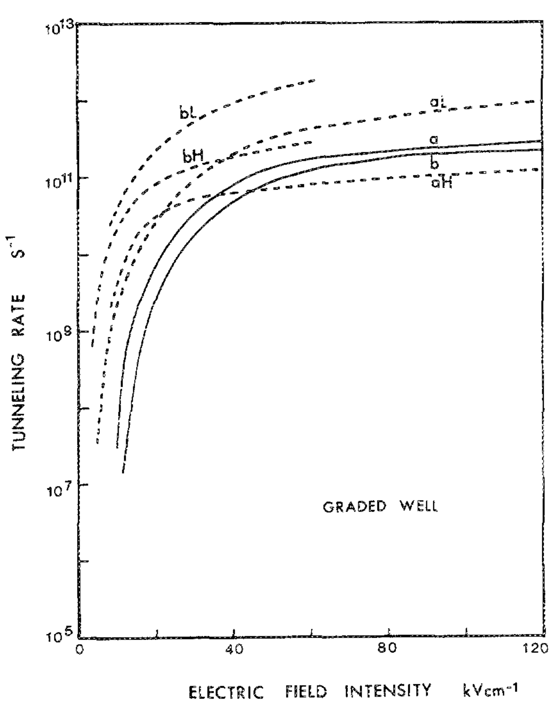

FIG. 6. Electron and hole tunneling rates vs transverse electric field in graded well. Solid line is for electron and dotted line for hole. H means heavy-hole case: L light-hole case. "a" stands for 60%:40% distribution of band discontinuity over conduction band and balance band, respectively, and "b" for 85%:15%.

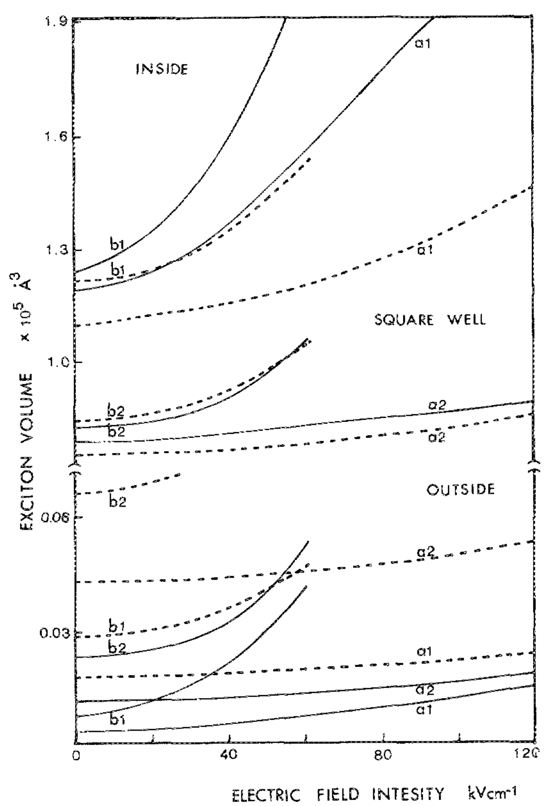

FIG. 7. Exciton volumes inside and outside of the square wells. Solid line is for heavy hole, and dotted line for light hole. "1" stands for the well width of 100 Å, and "2" for 70 Å. "a" stands for 60%:40% distribution of band discontinuity over conduction band and balance band, respectively, and "b" for 85%:15%.

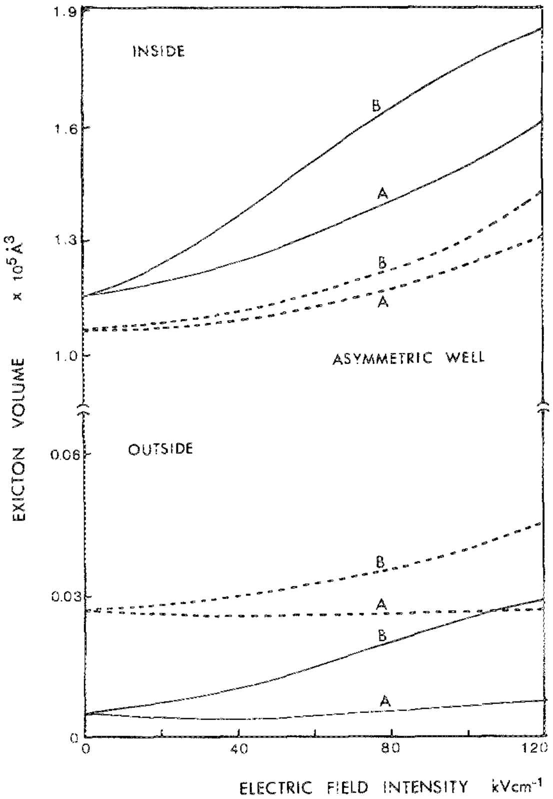

FIG. 8. Exciton volumes inside and outside of the asymmetric wells. Solid line is for heavy hole, and dotted line for light hole. Case A has 1.5 times higher barrier in low potential side than that of the opposite side. Case B has 1.5 times higher barrier in high potential side. For 60%:40% case.

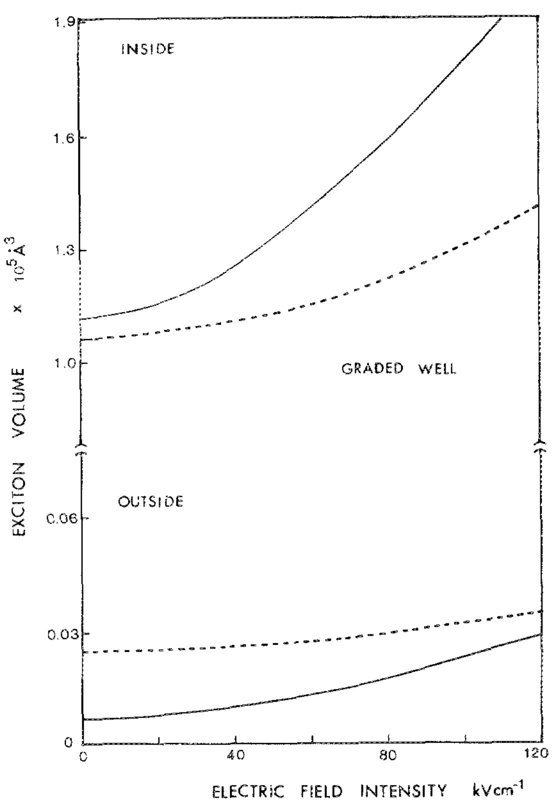

FIG. 9. Exciton volumes inside and outside of the graded well. Solid line is for heavy hole; and dotted line for light hole. For 60%:40% case.

IV. CONCLUSION

In this paper we have presented a numerical approach to address the problem of quasibound states in arbitrarily shaped quantum wells. For the case of the square well, the results from these calculations agree with the results of other workers. However, the versatility of this approach allows us to study excitonic properties in other wells. Advances in heterostructure fabrication techniques can now allow growth of graded and asymmetric quantum wells, although experimentally not much has been done in studying the excitonic properties of such structures. We find from the calculations that the shape of the quantum well does not play an important role in controlling the exciton binding and emission energies. For example one can replace a graded well by an average square well and get a reasonably accurate value for these parameters. However, the tunneling rates of the electron and hole in presence of electric field are extremely sensitive to the shape of the wells, as well as to the band-edge discontinuities assumed (i.e., 85:15 or 60:40). Since in optical modulators, it may be important to keep the electron and hole tunneling rates as close to each other as possible to avoid buildup of space charge, it may be useful to use asymmetric barriers. We have also examined in detail the exciton volume in different shaped quantum wells and its dependence on the applied field. The exciton volume can be tailored by both the shape of the well and the applied field. The exciton spatial extent plays an important role in the oscillator strength of the excitonic transitions, the exciton linewidth as well as optical nonlinearities so that the ability to tailor this important quantity could be of considerable use.

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