

tional Si-N feature at 875 cm^{-1} , and leading to a reinterpretation of the data for the low- T_s $a\text{-Si}:(\text{H},\text{N})$ film. The Si-N absorption band in the $a\text{-Si}:(\text{H},\text{N})$ films at 790 cm^{-1} is accompanied by absorption due to H-N groups at 3350 and 1150 cm^{-1} ; in a parallel manner absorption at 705 cm^{-1} in $a\text{-Si}:(\text{D},\text{N})$ films is accompanied by D-N vibrations at 2445 and 980 cm^{-1} . For the low- T_s $a\text{-Si}:(\text{H},\text{N})$ films (1) the triad of vibrations discussed above, 3350 , 1150 , and 790 cm^{-1} , is assigned to a local bonding arrangement involving a terminal H-N group attached to the Si-host network (the 790-cm^{-1} feature is one of two Si-N vibrations at this site), and (2) a vibration at 875 cm^{-1} (in part obscured by the polysilane scissors-wagging doublet) is assigned to regions in which the incorporation of N atoms results in a local bonding geometry that is like that of $a\text{-Si}_3\text{N}_4$. Thus the low- T_s films are viewed as being inhomogeneous with one phase having a polysilane component, and the second being an $a\text{-Si}:\text{N}$ alloy in which the

N atoms are clustered in regions with a stoichiometry corresponding to Si_3N_4 . Prior to the studies of the deuterated films, we had assumed the second phase to contain isolated, rather than clustered N-atom sites. For further details, please refer to a paper by G. Lucovsky, S. S. Chao, J. Yang, J. Tyler, and W. Czubatyj to appear in [J. Vac. Sci. Technol. A (in press)].

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Variational calculations on a quantum well in an electric field

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We present variational calculations of the eigenstates in an isolated-quantum-well structure subjected to an external electric field. At weak fields a quadratic Stark shift is found whose magnitude depends strongly on the finite well depth. In addition, the electric field induces a spatial shift of the particle wave function along or opposite to the field direction, depending on the sign of the particle mass. This field-induced spatial separation of conduction and valence electrons in GaAs quantum wells decreases the overlap between their associated wave functions, leading to a reduction of interband recombination.

I. INTRODUCTION

The photoluminescence of multiple-quantum-well (MQW) structures, mostly $\text{GaAs-Ga}_{1-x}\text{Al}_x\text{As}$, has received considerable attention. So far the emphasis has been on their structural properties, e.g., the one-dimensional quantization along the quantum well^{1,2} (QW) and the quasi-two-dimensional behavior of the exciton³⁻⁵ and impurity levels.⁶⁻⁹ Recently we have shown¹⁰ that the application of an external electric field in the range 10–50 kV/cm, perpendicular to the layers defining the quantum wells, sensitively decreases or even completely quenches the luminescence of $\text{GaAs-Ga}_{1-x}\text{Al}_x\text{As}$ MQW's. We associated this phenomenon with the field-induced spatial separation between electrons and holes confined in the GaAs QW's.

We report in this paper variational calculations of the effect of an external electric field on the eigenstates of an isolated QW. The results are relevant to any optical processes, such as emission and absorption, in QW's. In this regard, they could be useful in the understanding of quantum-well lasers. Although it is not possible to make a full comparison between this theoretical model and the complex MQW structure of the experiments reported,¹⁰ the calculations are consistent with the observations, especially when a finite QW is considered, in leading to field-induced reductions of both the energy and the intensity of the luminescence. In Sec. II we consider the effect of an electric field on an infinite QW. In Sec. III we show that this effect is greatly enhanced for a QW with a finite height and then apply it to the case of a single GaAs QW. The main results of the paper are summarized in Sec. IV.

II. INFINITE QUANTUM WELL

The calculation of the eigenstates of an infinite QW subject to a constant electric field is, in principle, an exactly solvable problem whose solutions are linear combinations of two independent Airy functions. These solutions are, however, fairly complicated and it is sometimes desirable to use approximated, but more manageable, schemes. Lukes and Ringwood¹¹ have used an approach based on the Green's function to calculate, to the second order, the corrections to the energy eigenvalues induced by the electric field. On the other hand, Fernandez and Castro¹²

have employed recently a hypervirial-perturbational treatment that gives the energy corrections up to the fourth order in an analytical way. Here we present a variational approach which not only gives a reasonable accuracy but, in addition, provides the field-dependent wave functions.

Let us consider a particle, with charge e and effective mass m^* , in an infinite quantum well of width L in the presence of an electric field F along the direction of the well, z . (In semiconductor heterostructures, this direction is perpendicular to the material layers.) The origins of distance and of electrostatic potential are chosen at the center of the well. The Hamiltonian of the problem is therefore

$$H = H_0 + |e| Fz, \quad (1)$$

where H_0 is the zero-field quantum-well Hamiltonian. The spectrum of H_0 is discrete and given by

$$E_n^{(0)} = \frac{\hbar^2 \pi^2}{2m^* L^2} n^2 \quad n = 1, 2, \dots \quad (2)$$

For weak fields, such that

$$|e| FL \ll \frac{\hbar^2 \pi^2}{2m^* L^2}, \quad (3)$$

a second-order perturbation calculation gives for the energy shift of the ground state,¹²

$$\Delta E_1^{(2)} = -C_{\text{pert}} \frac{m^* e^2 F^2 L^4}{\hbar^2}, \quad (4)$$

with

$$C_{\text{pert}} = \frac{2^9}{\pi^6} \sum_{p=1}^{\infty} \frac{p^2}{(4p^2-1)^5} = \frac{1}{24\pi^2} \left[\frac{15}{\pi^2} - 1 \right]. \quad (5)$$

For thick enough wells and/or large enough electric fields, this perturbation treatment is invalid because Eq. (3) is not satisfied. The particle is pushed against (for $m^* > 0$) or along (for $m^* < 0$) the direction of the field (we have considered e negative) and, at high fields, the charge distribution is concentrated near the well barrier. This physical situation can be well described if we use a variational wave function given by

$$\psi(z) = N(\beta) \cos \left[\frac{\pi z}{L} \right] \exp \left[-\beta \left(\frac{z}{L} + \frac{1}{2} \right) \right], \quad \frac{|z|}{L} < \frac{1}{2}, \quad (6)$$

where β is the variational parameter and $N(\beta)$ a normalization constant. Note that $\psi(z)$ becomes exact in the limit $F \rightarrow 0$, when $\beta \rightarrow 0$, and, at high fields, approaches the Fang-Howard wave function, which has been used successfully to describe the bound-electron levels in a metal-oxide-semiconductor (MOS) structure.¹³ Using Eqs. (1) and (6) we obtain for the energy, whose value is to be minimized,

$$E(\beta) = E_1^{(0)} \left[1 + \frac{\beta^2}{4\pi^2} + \phi \left(\frac{1}{\beta} + \frac{2\beta}{4\pi^2 + \beta^2} - \frac{1}{2} \coth \frac{\beta}{2} \right) \right], \quad (7)$$

where $E_1^{(0)}$ is the ground-state energy at zero field and ϕ is the dimensionless electrostatic energy,

$$\phi \equiv \frac{|e|FL}{E_1^{(0)}}. \quad (8)$$

Figure 1 shows the binding energy for the particle, obtained by minimizing $E(\beta)$ with respect to β , as a function of ϕ . Because the QW is defined only by its length L , the curve in Fig. 1 is universal. In the limits of low or high fields the minimization can be done analytically.

In the low-field limit, $\phi \ll 1$, $\beta_{\min} = \phi(\pi^2/6 - 1)$, and

$$\Delta E_1 \equiv E(\beta_{\min}) - E_1^{(0)} = -C_{\text{var}} \frac{m^* e^2 F^2 L^4}{\hbar^2}, \quad (9)$$

with

$$C_{\text{var}} = \frac{1}{8} \left[\frac{1}{3} - \frac{2}{\pi^2} \right]^2. \quad (10)$$

A quadratic Stark shift is thus obtained whose value is in very good agreement with that from the second-order per-

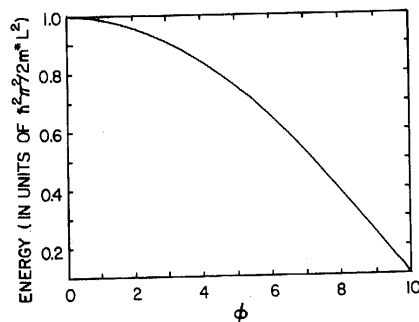


FIG. 1. Dimensionless binding energy, obtained from the minimization of Eq. (7) with respect to β , of a charged particle in an infinite QW vs the dimensionless parameter ϕ . This quantity is proportional to an electric field applied along the direction of the well.

turbation result, $C_{\text{var}}/C_{\text{pert}} = 0.97$. A detailed comparison between the variational and perturbation calculations is shown in Fig. 2, where we have used a QW width of 30 Å and an effective mass of 0.067 times the free-electron mass m_0 .

In the high-field limit, $\phi \gg 1$, β_{\min} becomes proportional to $F^{1/3}$, and

$$\Delta E_1 = -\frac{|e|FL}{2} + \left(\frac{3}{2} \right)^{5/3} \left[\frac{e^2 F^2 \hbar^2}{m^*} \right]^{1/3}. \quad (11)$$

A semiclassical (sc) calculation for the ground state of a particle bound by an infinite barrier between $z = -L/2$ and a linearly varying potential $|e|Fz$ gives

$$E_{\text{sc}} = -\frac{|e|FL}{2} + \left[\frac{\pi^2}{12} \right]^{1/3} \left[\frac{3}{2} \right]^{5/3} \left[\frac{e^2 F^2 \hbar^2}{m^*} \right]^{1/3}, \quad (12)$$

which is accurate to better than 1%,¹⁴ and indicates that in this limit our variational result is accurate within 8%.

III. FINITE QUANTUM WELL

In semiconductor heterostructures (e.g., GaAs-Ga_{1-x}Al_xAs) the band discontinuities that are the origin of quantum wells for valence and conduction electrons are at most of the order of a few hundred meV and the infinite-well approximation is not good, except for very thick wells. For a thin QW (e.g., $L \leq 50$ Å for conduction electrons in GaAs QW's) the energy spectrum at $F=0$ consists of a single bound level and a continuum, instead of the increasingly spaced levels given by Eq. (2). In the presence of an electric field there is no true bound state for a finite QW of depth V_0 , since the potential energy is negative and large in absolute value, for large, negative values of z . If, however, the field is not excessively large, the states will have a long lifetime and can therefore be considered as quasibound. To set the criterion, let L/q_0 be the characteristic decay length of the unperturbed ($F=0$)

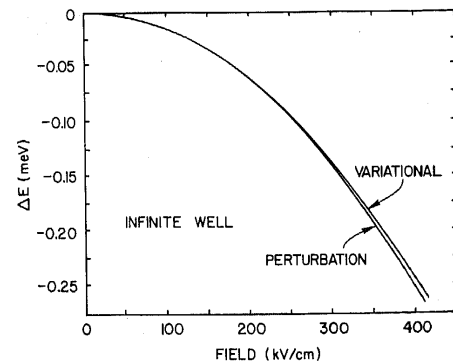


FIG. 2. Comparison of the energy shifts obtained from a variational and a second-order perturbation calculation, for a conduction electron in an infinite GaAs QW, 30 Å thick, and subject to an electric field.

ground-state wave function inside the finite barrier,

$$q_0^2 = \frac{2m^*L^2}{\hbar^2} (V_0 - E_1), \quad (13)$$

where q_0 is the characteristic dimensionless wave vector and E_1 represents the zero-field ground-state energy. We can talk about quasibound states if

$$\frac{|e|FL}{q_0} \ll \frac{\hbar^2 q_0^2}{2m^*L^2}. \quad (14)$$

(We have assumed, for simplicity, that the particle effective mass in the barrier is the same as in the well.) When condition (14) is satisfied, the wave function decays fast inside the barrier so that its amplitude becomes negligible where the barrier height has been significantly lowered by the electric field. In the following we will consider two field regimes, weak and strong, for values of F and q_0 (which depends on L) such that Eq. (14) holds.

A. Weak field

In the presence of an electric field, the even ground state of a thin finite QW is admixed with odd extended states. A variational wave function that takes into account this admixture is

$$\psi(z) = N(\beta) (1 + \beta z/L) \psi_0(z), \quad (15)$$

where $\psi_0(z)$ is the unperturbed ground-state wave function, β is a variational parameter, and $N(\beta)$ a normalization constant. The energy to be minimized can be written as

$$E(\beta) = E_1 + \frac{L^2}{L^2 + \beta^2 \langle z^2 \rangle_0} \left[\frac{2e\beta F \langle z^2 \rangle_0}{L} + \frac{\hbar^2 \beta^2}{2m^*L^2} \right], \quad (16)$$

where

$$\langle z^2 \rangle_0 = \langle \psi_0 | z^2 | \psi_0 \rangle. \quad (17)$$

For weak fields, such that $\beta^2 \langle z^2 \rangle_0 \ll L^2$, it is

$$\beta_{\min} = -\frac{2|e|Fm^*L}{\hbar^2} \langle z^2 \rangle_0, \quad (18)$$

and then the field-induced energy shift can be expressed as

$$\Delta E_1 = -\frac{\Omega^2 m^* e^2 F^2 L^4}{8 \hbar^2} \quad (19)$$

with

$$\Omega(k_0, q_0) = A \left[\frac{1}{3} + \frac{\sin k_0}{k_0} + \frac{2 \cos k_0}{k_0^2} - \frac{2 \sin k_0}{k_0^3} + \frac{2}{q_0} \left[1 + \frac{2}{q_0} + \frac{2}{q_0^2} \right] \cos^2 \left[\frac{k_0}{2} \right] \right], \quad (20)$$

$$A = \left[1 + \frac{\sin k_0}{k_0} + \frac{2}{q_0} \cos^2 \left[\frac{k_0}{2} \right] \right]^{-1}, \quad (21)$$

where k_0 is the dimensionless wave vector which characterizes the unperturbed ground state in the well,

$$k_0^2 = \frac{2m^*L^2}{\hbar^2} E_1. \quad (22)$$

For an infinite barrier height $q_0 \rightarrow \infty$, $k_0 \rightarrow \pi$, and $\Omega \rightarrow \Omega_\infty$:

$$\Omega_\infty = \frac{1}{3} - \frac{2}{\pi^2} \quad (23)$$

so that Eq. (19) reproduces Eq. (9) in this limit. Plotted in Fig. 3 is the L dependence of Ω/Ω_∞ for two values of m^* and V_0 , which shows an increase of the finite-well polarizability with decreasing L .

As Fig. 3 shows, the effect of a finite barrier height is to enhance drastically the field-induced energy shift, especially for thin wells. For instance, for $L=30$ Å, $m^*=0.067m_0$, and $V_0=0.4$ eV the shift is 38 times larger than that calculated in the infinite-well approximation; for $m^*=0.45m_0$ and $V_0=0.07$ eV it is 28 times larger. These numbers for m^* and V_0 describe, respectively, the electron and heavy-hole states in GaAs-Ga_{0.62}Al_{0.38}As QW's.

B. Strong field

At large fields, but for which Eq. (14) still holds, deviations from the quadratic field dependence, Eq. (19), are to be expected. To describe the case in which F can take any value [subject to Eq. (14)], we can generalize Eqs. (6) and (15) in terms of the variational parameter β in the following way:

$$\psi(z) = \begin{cases} A \exp \left[(q_0 - \beta) \left(\frac{z}{L} + \frac{1}{2} \right) \right], & z \leq -L/2 \\ B \sin \left[k_0 \frac{z}{L} + \delta \right] \exp \left[-\beta \frac{z}{L} \right], & |z| \leq L/2 \\ C \exp \left[-(q_0 + \beta) \left(\frac{z}{L} - \frac{1}{2} \right) \right], & z \geq L/2 \end{cases} \quad (24)$$

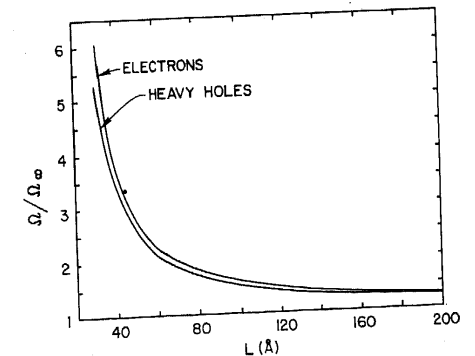


FIG. 3. Plot to the ratio Ω/Ω_∞ , for electrons and heavy holes, vs thickness of a finite GaAs QW, in an electric field. The term $(\Omega/\Omega_\infty)^2$ represents the enhancement factor of the energy shift, at low fields, due to the finite value of the barrier height. The parameters used in the calculation were, for electrons, $m^*=0.067m_0$, $V_0=0.4$ eV, and for holes, $m^*=0.45m_0$, $V_0=0.07$ eV.

where A , B , and C are normalization constants to be determined using the continuity condition of $\psi(z)$ and $m^{-1}(z)d\psi/dz$. If we neglect the change of the effective mass $m(z)$ at the interface, the boundary conditions imply that k_0 and q_0 retain their zero-electric-field values, related to each other by

$$k_0^2 - q_0^2 = 2q_0 k_0 \cot k_0, \quad (25)$$

and that

$$\delta = \frac{k_0}{2} + \tan^{-1} \frac{k_0}{q_0}. \quad (26)$$

It should be noted in Eq. (24) that, in order to maintain an evanescent wave function in the barrier, β must be smaller than q_0 times the sign of the particle effective mass. Because β_{\min} is a function of the field, this condition is reminiscent of Eq. (14).

The analytical expression for the expectation value of the Hamiltonian, $E(\beta)$, is too complicated to be shown here. We present in Fig. 4 the calculated energy shift as a function of the electric field for conduction electrons in a GaAs QW, 30 Å thick. In this case the shift is quadratic in F up to ~ 100 kV/cm, so that the weak-field formalism can be applied. On the other hand, for valence electrons, the shift is quadratic only up to ~ 10 kV/cm. The magnitude of the electric field in recent experiments¹⁰ in GaAs QW's was in the range 10–50 kV/cm. According to our calculations, for $F=50$ kV/cm the energy shift for conduction and valence electrons, in a 30-Å-thick QW, would be -0.2 and -0.8 meV, respectively. For $L=100$ Å the corresponding shifts would be -1.2 and -6.4 meV.

In the above example, because the valence-band QW barrier is quite small and the effective mass is heavy, the valence shift is much larger than the conduction shift. Accordingly, the valence envelope wave function ψ_v is more strongly affected by the field than the conduction wave function ψ_c . This is illustrated in Fig. 5, which shows the calculated conduction and valence wave functions for an electric field of 100 kV/cm for (a) $L=30$ Å and (b) $L=100$ Å. The polarization effect is more pro-

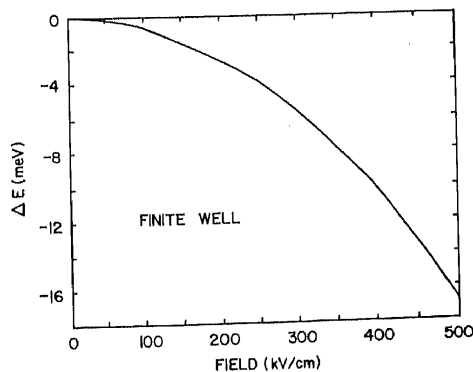


FIG. 4. Energy shift vs electric field of the eigenvalue of a conduction electron in a finite GaAs QW, 30 Å thick.

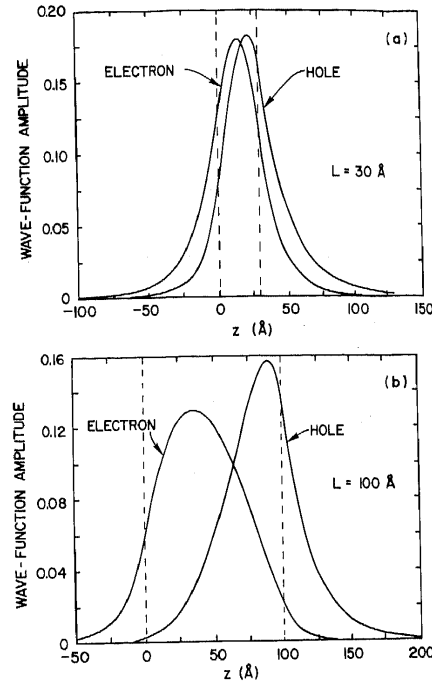


FIG. 5. Wave-function amplitudes vs distance for electrons and heavy holes in a GaAs QW of thickness (a) 30 Å and (b) 100 Å, subject to an electric field of 100 kV/cm. In the figure, the origin of distance is taken at the edge of the well so that the dashed lines represent the well boundaries.

nounced in the latter case, where $\psi_v(z)$ peaks near the well edge. This spatial separation of carriers strongly affects the overlap integral M_{cv} , defined as

$$M_{cv} \equiv \int_{-\infty}^{\infty} dz \psi_c(z) \psi_v(z). \quad (27)$$

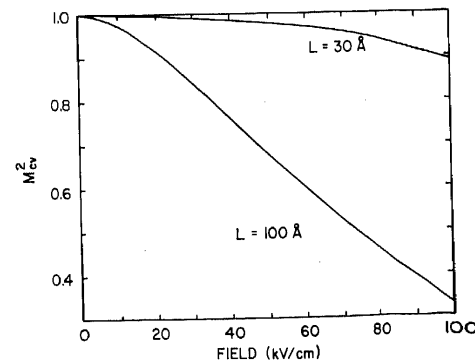


FIG. 6. Square of the overlap integral between electrons and heavy holes as a function of electric field for a QW of 30 or 100 Å.

The recombination rate between conduction and valence electrons is proportional to M_{cv}^2 , so that this quantity, plotted in Fig. 6 versus electric field, is most significant in photoluminescence experiments. For example, for a field of 100 kV/cm, M_{cv}^2 decreases by $\sim 10\%$ for a QW with $L=30$ Å and by $\sim 67\%$ when $L=100$ Å.

These results predict qualitatively the trends observed recently¹⁰ but give rise to too small an effect for the electric fields under consideration. Experimentally, fields in the range 10–50 kV/cm were sufficient to quench completely the luminescence originating in GaAs QW's. For a field in this range, 50 kV/cm for example, the reduction in photoluminescence intensity is calculated to be only $\sim 3\%$ for $L=30$ Å. These calculations have been based on barrier heights following the 85–15% rule for the conduction- and valence-band discontinuities.¹⁵ A deviation of this empirical rule could make the valence-band well shallower, resulting in a larger polarization, which in turn would further reduce the recombination rate. Furthermore, the field-induced leakage of the wave functions into the $\text{Ga}_{1-x}\text{Al}_x\text{As}$ barriers, where nonradiative recombination centers abound, should accelerate the luminescence quenching process. We believe, however, that the major reason for the discrepancy between the theoretical and experimental results lies in the fact that the experiments were done using not one but several QW's. In view of the narrow wells (30 Å) and barriers (100 Å) employed, the coupling among wells is considerable. The resulting energy bandwidth from the coupling is estimated to be ~ 1 meV, which implies an escape time from one well to another of $\sim 10^{-12}$ sec. The escape of electrons to an adjacent well in one direction and that of holes in the oppo-

site direction, a situation analogous to the field effect in an ordinary junction, is expected to give rise to a significant enhancement in the polarization of the carrier distribution and, consequently, to a drastic reduction in luminescence.

IV. CONCLUSION

We have investigated the electric-field-induced polarization effects of conduction and valence envelope functions in isolated QW's by means of perturbation and variational methods. The finite height of the confining barriers has been shown to enhance considerably these effects with respect to those in infinitely deep QW's of the same thickness. The results show that large polarizations are expected for GaAs QW's with a thickness $L \geq 100$ Å. In particular, the induced spatial separation of conduction and valence electrons should decrease the interband absorption or the radiative emission rate. A comparison of the calculations with the recent observation of electric-field-induced quenching of luminescence in GaAs MQW's shows a quantitative discrepancy, probably due to the considerable coupling among wells of the structures used in the experiments.

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