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Excitonic properties and resonance widths in biased (Ga,In)As-GaAs double quantum wells

Pierre Bigenwald and Bernard Gil
Groupe d'Etude des Semiconducteurs, Université de Montpellier II, Case Courrier 074, 34095 Montpellier CEDEX 5, France
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We calculate the exciton properties and resonance widths of the charge carriers trapped in (Ga,In)As-GaAs double quantum wells embedded in *p-i-n* diodes. The calculation is made for both the (001)- and (111)*B*-oriented structures and evidences the dramatic influence of the built-in piezoelectric field that occurs when the growth axis is (111). We also perform excitation calculations, which show that, when the band lineups are deformed due to the excitonic interaction between particles, the overlap integral of the wave functions increases and so the carriers' escape time out of the quantum-well diodes increases with respect to the values calculated for the "empty" crystal.

I. INTRODUCTION

The optical properties of low-dimensional systems based on III-V and II-VI semiconductors are, in general, anisotropic. This topic is deserving theoretical and experimental investigation. Reasons invoked earlier for this include, for instance, the possibility of obtaining lasers which can display lower threshold current when grown along the (111) direction instead of (001).[1] Most semiconductors are lattice mismatched, except when peculiar alloying is performed. We can take advantage of the strain that results from this lattice mismatch, such as for gallium-rich (Ga,In)As layers coherently grown on GaAs substrates, to design laser diodes with optimized characteristics compared to the conventional $Ga_{1-x}Al_xAs$-GaAs ones.[2] The growth of strained-layer compounds away from the (001) direction results in anisotropic displacements of the barycenters of anions and cations and causes built-in piezoelectric fields.[3]

Phase-space-filling effects are very efficient in heterostructures having such piezoelectric fields. This opens the opportunity to build devices that display strong nonlinear optic behaviors in the regime of moderate photoexcitation densities. We addressed this effect quantitatively in previous papers devoted to (Ga,In)As-GaAs single and double quantum wells.[4,5] Besides this, one also has to consider the design and conception of advanced devices. They can be multiple-quantum-well modulators based on the quantum confined Stark effect, which are of interest for optical switching, optical interconnects, signal processing, and optical computing.[6–8] This is the reason why we have studied in detail the properties of (Ga,In)As-GaAs double quantum wells grown along both the (001) and (111) directions. Since the ultimate goal of the physics is the technological application, the structures are embedded in *p-i-n* diode architectures.

We have first calculated the resonance widths of the pseudoconfined electronic states. These resonance widths may be large, because we are dealing with a double-quantum-well system embedded in *p-i-n* diodes for which there is one semitransparent barrier at one side of the asymmetric structure that induces a significant tunneling of the charge carriers out of these rather shallow wells.[9]

Second, the exciton binding energies and excitonic oscillator strengths were computed in the context of the envelope-function approach, where the electric-field problem is included at the initial stage of the calculation.[10] These calculations of the excitonic interaction were made using a two-parameter trial function, as previously presented in Ref. 11. The excitonic interaction remains significant in strong-field regimes even when the overlap of electron and hole wave functions has collapsed. We also calculated the exciton interaction for high-index bands and showed that, in the case of (001) growth, the e_1hh_2 exciton may be stronger than e_1hh_1, which is not true for the (111) growth direction.

Finally, we show that, for some selected cases, the overlap of the envelope functions can be enhanced if we perform a calculation where the band lineups are modified by the presence of the excitonic dipole.

The paper is organized in the following way. In the next section, we briefly recall some of the physical and mathematical issues encountered when treating the problem of a quantum well under an electric field. We calculate the resonance widths for $Ga_{1-x}In_xAs$-GaAs double-quantum-well diodes (DQWD's) and compare them with the values for single quantum wells. In the third section of this paper, we present the full results concerning the exciton properties as a function of the internal barrier width and of the growth orientation. We also address the excitonic calculations before giving some concluding remarks in the last section.

II. AN OVERVIEW OF THE QUANTUM-WELL PROBLEM UNDER AN ELECTRIC FIELD

We treat here the case where the electric field is intrinsic and is related to the *p-i-n* polarization of the diode within which the symmetric double-quantum-well system is embedded. For the (111)-grown samples, the built-in piezoelectric field superimposes colinearly on the *p-i-n* field. The *p-i-n* field equals 1.5×10^4 V/cm in the DQWD region and corresponds to 1 μm of undoped GaAs sandwiched between thick n^+- and p^+-type GaAs layers. The indium composition we have chosen is 8% as in our previous papers.[4,5] In this situation, the *p-i-n* field is

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about one order of magnitude smaller than the built-in piezoelectric field. As previously shown, interesting properties for devices are expected if the *p-i-n* and piezoelectric fields have opposite signs.[6–8] This has important implications at the growth stage and requires the growth of samples on (111)*B* *n*-type GaAs substrates. As the (Ga,In)As layers are in biaxial compression, the electric-field polarization vector points from the cation (*A*) to the anion (*B*) face, thus lowering the conduction-band edge with respect to its position in the substrate.

The electric field noticeably modifies the band lineups, depending on the growth orientation.[4–6] It also significantly alters the optical properties of the microstructures.[6–8] The effective-mass Hamiltonian includes a potential term (qFz) and the inversion symmetry is broken with respect to the flatband problem. The envelope functions of the carriers are modified and interband transitions *a priori* forbidden between electron (e_i) and heavy-hole (hh_j) states with i and j of different parity; e_1-hh_2 and e_2-hh_1 transitions become allowed since the overlap integrals $\langle \Psi_i | \Psi_j \rangle$ no longer equal zero.[9] Simultaneously, due to the spatial separation of the carriers, the intensity of the fundamental transition decreases. For multiple structures like multiple quantum wells and superlattices, a decoupling of the spatially extended states and a localization may be produced. This has been predicted earlier by various authors but only recently observed.[12]

The metastable states belonging to the quantum wells are coupled to the continuum of energy states in the large triangular well, when the vacuum-semiconductor interfaces behave like infinite barriers.[13] This coupling is ignored hereafter. However, it is necessary to consider that the trapping of the carriers is limited in time. After the time τ, the particles have a significant probability of tunneling out of the structure. In particular conditions, it is possible to neglect this reduction of the lifetime in the quantum well.[13] Figure 1 of Ref. 5 shows that fundamental electron and hole levels are well confined for the (111)-grown sample while the tunneling effect out of the well is not negligible for (001) growth. We will show later that for our cases $\tau(001) \ll \tau(111)B$.

Bastard[13] proposed a criterion to distinguish "rather" confined states from actually resonant ones for a biased single quantum well of GaAs-$Ga_{1-x}Al_xAs$ grown along (001). This criterion consists in comparing, for the ground state, the confinement energy at zero bias $(V_b - E_1)$ with the field-induced barrier shrinkage $(eF\mathbf{k}_b^{-1})$: if $eF\mathbf{k}_b^{-1} \ll (V_b - E_1)$, the escape of the particle out of the well can be neglected. This is not the case for a GaAs-$Ga_{0.9}In_{0.1}As$-GaAs single well 50 Å wide in a *p-i-n* field of 0.15 mV/Å: we calculate $eF\mathbf{k}_b^{-1} = 6.5$ meV, $(V_b - E_1) = 30.62$ meV, and $eF\mathbf{k}_b^{-1} = 2.9$ meV, $(V_b - E_1) = 26.33$ meV, for the electron and heavy hole, respectively.

In contrast to the case studied in Ref. 13, the electric field should not be treated within the context of perturbation theory because the potential are not deep enough: the confinement is comparable to the barrier height shrinkage. Each eigenenergy is modified: it is a complex quantity containing the solution of the time-dependent Schrödinger equation and is written as $E = E_0 - i\hbar/2\tau$, where E_0 is the energy of the state broadening, which is neglected, and τ the lifetime of the trapped particle.

Several approaches have been proposed to evaluate the escape time of the particle. The simplest method is based on the Heisenberg uncertainty principle. If the width of the resonance is ΔE, one can get τ from $\Delta E \tau \sim \hbar$. It is also possible to use the semiclassical approach of Ref. 13. Let $T(E_1)$ be the period of the classical oscillations of the bound particle and $D(E_1)$ the transmission coefficient through the most transparent barrier at the energy E_1. Then $1/\tau = D(E_1)/T(E_1)$. Going back to the preceding case, we calculate for the electrons $D = 1.9 \times 10^{-3}$ and $T^{-1} = 1.88 \times 10^{13}$ Hz. This gives $\tau_e = 100$ ps and $\hbar/2\tau_e = 0.033$ meV. For heavy holes the results are $D = 1.2 \times 10^{-5}$ and $T^{-1} = 7.61 \times 10^{12}$ Hz, so $\tau_{hh} = 11$ ns and $\hbar/2\tau_{hh} = 6 \times 10^{-5}$ meV.

Another method called the phase-shift analysis method has been proposed by Austin and Jaras to determine τ.[10] We will adopt their formalism here. For energies in the vicinity of the resonance E_0, a phase shift Φ is determined and $\Gamma = 1/\tau$ is derived from the Breit-Wigner formula

$$\tan[\Phi(E)] = \frac{\Gamma}{2(E_0 - E)} .$$

In the same article, they also proposed to calculate Γ from the derivative $\Gamma = 2/(d\Phi/dE)_{max}$. Applied to our example, this gives the following. If grown along (001), for electrons we have $\tau_e = 63$ ps and $\hbar/2\tau_e = 0.053$ meV, and $\tau_{hh} = 1.1$ ns and $\hbar/2\tau_{hh} = 2.9 \times 10^{-4}$ meV for heavy holes. If grown along (111)*B*, we have for electrons $\tau_e = 23$ ns and $\hbar/2\tau_e = 2.8 \times 10^{-5}$ meV, and $\tau_{hh} > 80$ ns and $\hbar/2\tau_{hh} < 1 \times 10^{-5}$ meV for heavy holes.

III. VARIATIONAL PROCESS FOR (Ga,In)As-GaAs DOUBLE QUANTUM WELLS

Through all the calculations, we kept the value used in the previous section for the *p-i-n* field. Calculations were performed for $Ga_{0.92}In_{0.08}As$ DQWD's with a constant well width and a variable thickness of the internal barrier layer. For every case, the calculation was performed for both the (001)- and its twin (111)*B*-oriented design. Figures 1 [(001) orientation] and 2 [(111)*B* orientation] show the evolution of the energies of the electron and valence states with the thickness of the internal barrier. The energy levels are marked with respect to the energies of the lineups at positions *A*, *B*, *C*, or *D*. Insets represent in both cases the band lineups and the *A*, *B*, *C*, and *D* positions. The dashed lines represent the evolution of *A*, *B*, *C*, and *D* positions, while the full lines represent the evolution of the electronic and hole levels. We remark from Fig. 2 that, for the (111) orientation, the fundamental states are well localized and the width of the barrier does not significantly influence the valence states. The situation is very different for the (001) orientation. The localization of the particles is rather weak; this is particularly true for the second electron which requires an internal barrier larger than ~50 Å to be localized in the right-hand well. Due to the type-II configuration in flatband,[14]

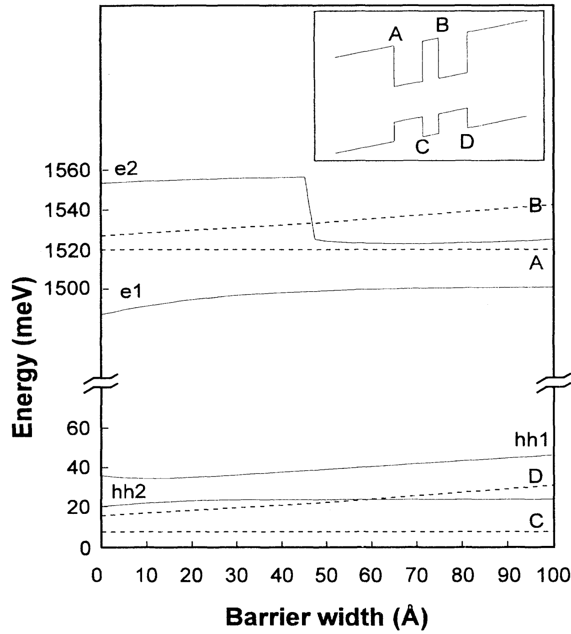

FIG. 1. Eigenenergies of the fundamental electron and heavy-hole states for a biased double quantum well (50 Å/L/50 Å) GaAs-$Ga_{0.92}In_{0.08}As$-GaAs versus the barrier width L for the (001) growth axis.

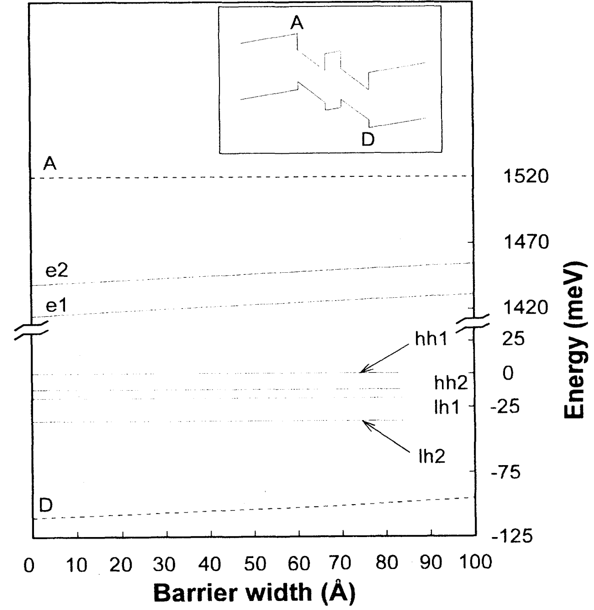

FIG. 2. Eigenenergies of the fundamental conduction and valence states for a biased double quantum well (50 Å/L/50 Å) GaAs-$Ga_{0.92}In_{0.08}As$-GaAs versus the barrier width L for the (111)B growth axis.

the light-hole (lh) states cannot be computed for the (001) orientation because the electric field separates the electron and light-hole wave functions too strongly.

Figure 3 represents the theoretical broadenings that we calculate for the e_1, hh_1, and hh_2 states using Austin and Jaros's[10] criterion in these double quantum wells. The spatial separation of the electron state e_1 (left-hand well) and heavy-hole state hh_1 (right-hand well) increases with increase of the thickness of the GaAs barrier. Due to the existence of the p-i-n field, the lifetime of the rather localized carriers out of the e_1 and hh_1 states decreases while it increases for hh_2. This gives increasing e_1 and hh_1 and decreasing hh_2 resonance widths. These significant broadenings are high: up to 0.6 meV for hh_2 and 0.4 meV for e_1, indicating that the lifetime of electrons and holes trapped in such DQWD's will be greatly shortened with respect to the single-quantum-well case. Besides this, several authors have studied similar structures under strong electric fields. In these conditions, their experimental results show that the excitonic structures persist.[15–17]

We now try to appreciate how the Coulomb interaction between the particles can enhance the localization of the wave functions. For the (111)B orientation, we expect little change since the potential wells are deeper and the heavy-hole mass greater. The exciton eigenfunction Ψ is written as the product of two terms: $\chi_{eh}(z_e,z_h)$ depends on z_e and z_h and $\chi(\rho,z)$ is a trial function with two parameters (α,λ).[11] Then the global function $\chi_{eh}(z_e,z_h)$ satisfies the relation

$$\begin{aligned}
\Bigg\{ & \left[\frac{\hbar^2}{4}\right]\left[\alpha^1\left[\frac{1}{m_e}+\frac{1}{m_h}\right]+\frac{1}{2\mu}-\frac{\alpha|z|}{\lambda\mu}\right]e^{-2\alpha|z|/\lambda}-\frac{e^2}{4\pi\varepsilon_0\varepsilon r}\int_0^\infty\frac{1}{\sqrt{\rho^2+z^2}}e^{-2f(\rho,z)}\rho\,d\rho \\
& -\frac{\hbar^2\alpha^2}{2\lambda^4}\int_0^\infty z^2\left[\frac{f(\rho,z)+1}{f(\rho,z)^3}\right]\left[\alpha^2\left[\frac{1}{m_e}+\frac{1}{m_h}\right]-\frac{1}{\mu}\right]e^{-2f(\rho,z)}\rho\,d\rho \\
& +\alpha^2\left[\frac{\hbar^2}{2}\right]\left[\frac{1}{m_e}\frac{\partial}{\partial z_e}-\frac{1}{m_h}\frac{\partial}{\partial z_h}\right]z e^{-2\alpha|z|/\lambda}+N^2\Bigg[-\frac{\hbar^2}{2m_e}\frac{\partial^2}{\partial z_e^2}+V_e(z_e)-\frac{\hbar^2}{2m_h}\frac{\partial^2}{\partial z_h^2} \\
& +V_h(z_h)-E_e-E_h-E_b\Bigg]\Bigg\}\chi_{eh}(z_e,z_h)=0 .
\end{aligned}$$

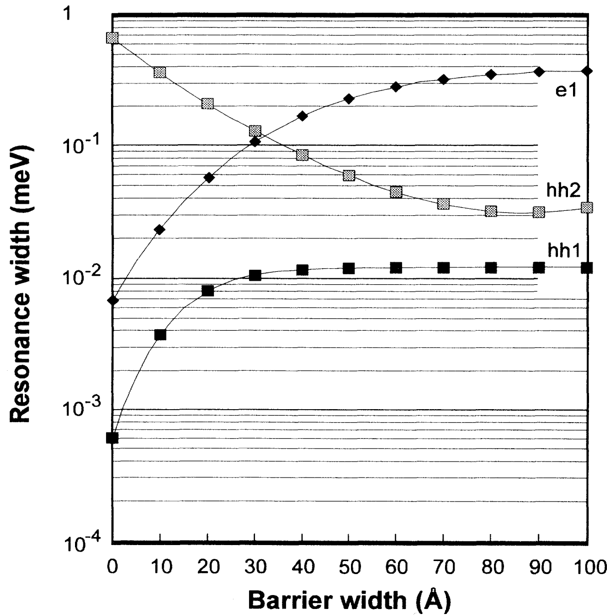

FIG. 3. Theoretical broadening of the e_1, hh_1, and hh_2 states of a biased double quantum well (50 Å/L/50 Å) GaAs-$Ga_{0.92}In_{0.08}As$ (001) versus the barrier width. The *p-i-n* field equals 0.15 mV/Å.

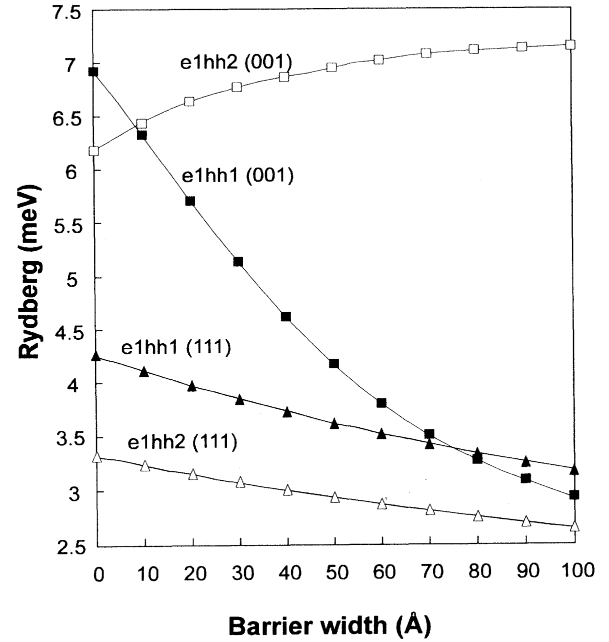

FIG. 4. Rydberg energy of the e_1-hh_1 and e_1-hh_2 transitions for a biased double quantum well (50 Å/L/50 Å) GaAs-$Ga_{0.92}In_{0.08}As$ versus the barrier width for the (001) and (111)B orientations. The *p-i-n* field equals 0.15 mV/Å.

In our notation,

$$\rho=\sqrt{(x_e-x_h)^2+(y_e-y_h)^2}\ ,$$

$z=z_e-z_h$, $\quad f(\rho,z)=\sqrt{\rho^2+\alpha^2 z^2}$, $\quad$ and $\quad \chi(\rho,z)=\exp[-f(\rho,z)/\lambda]$. N^2 is the normalization factor

$$N^2=\left[\frac{\lambda^2}{4}\right]\int_{-\infty}^{+\infty}\chi_e^2 dz_e\int_{-\infty}^{+\infty}\chi_h^2 dz_h\left[1+\frac{2\alpha|z|}{\lambda}\right]\times e^{-2\alpha|z|/\lambda}\ .$$

z_e (z_h), V_e (V_h), and E_e (E_h) are the position along (Oz), potential, and eigenenergy of the electron (heavy hole). E_b is the Rydberg of the transition.

This model is inspired by Sumi's model[18] and the previous equation is obtained after some cumbersome algebraic manipulations. As discussed in Ref. 18 for the bulk semiconductor and illustrated in Eqs. (2.10) of that paper, integration of $|\chi_{eh}|^2(z_e,z_h)$ over $z_e(z_h)$ gives the hole (electron) density of probability.

We first calculate the exciton binding energy using a two-parameter trial function in the standard variational approach.[11] In our DQWD's, the electric field localizes the envelope functions of the same "parity" in opposite wells. Combined with the increase of the internal barriers' width, it can have a significant influence on the excitonic parameters. Figure 4 illustrates how big the influence of the internal barrier is at the scale of the exciton binding energies for both (001)- and (111)B-oriented DQWD's. We find that the influence of the orientation of the DQWD is important: for the (001) orientation, the e_1-hh_2 transition may be favored with respect to e_1-hh_1 by changing the internal barrier thickness and therefore the real-space localization of the wave functions. This also appears on Fig. 5, where the excitonic oscillator strengths are plotted. This reversal of the strength of these two excitons is not found for the (111)B orientation. Thus, for the latter orientation, the two lowest hole states are localized in the same well and are spatially separated

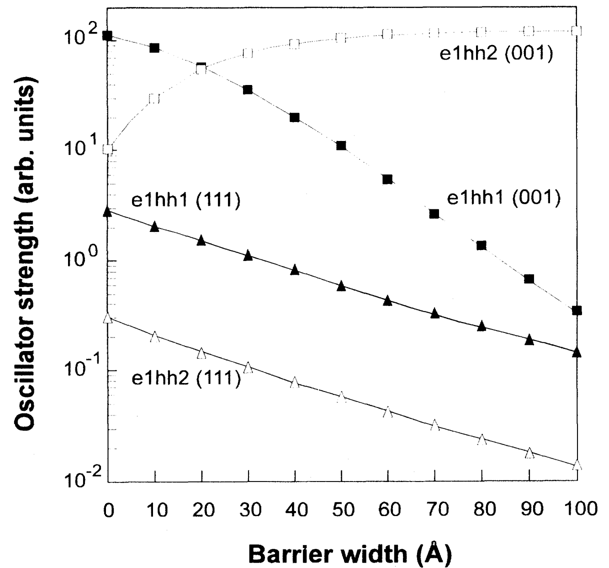

FIG. 5. Oscillator strength of the e_1-hh_1 and e_1-hh_2 transitions for a biased double quantum well (50 Å/L/50 Å) GaAs-$Ga_{0.92}In_{0.08}As$ versus the barrier width for the (001) and (111)B growth axes. The *p-i-n* field equals 0.15 mV/Å.

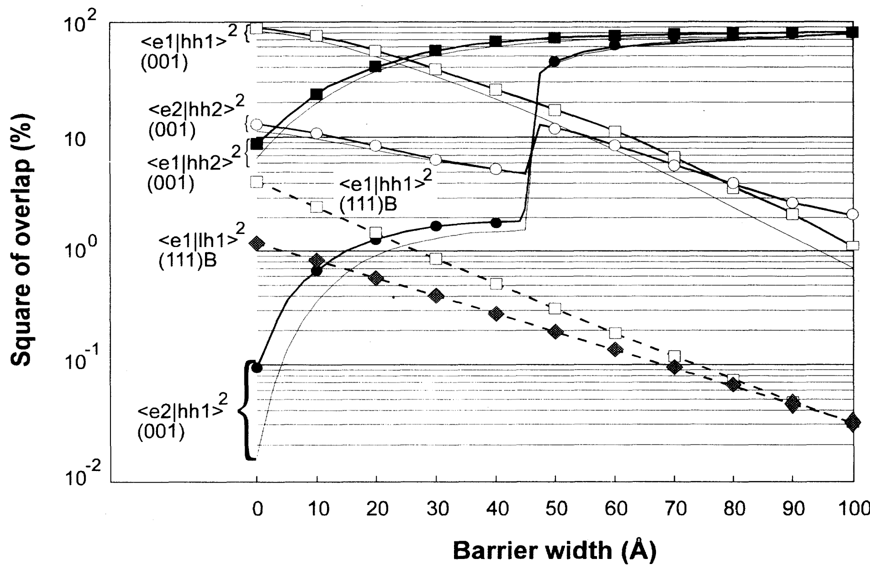

FIG. 6. Square of the overlap for the fundamental transitions of a biased double quantum well (50 Å/L/50 Å) GaAs-$Ga_{0.92}In_{0.08}As$ (001) versus the barrier width. The thin line refers to the classical envelope function model and the bold line to the more complete calculation.

from both the first and second electron states. We remark that the Rydberg is a slowly varying function of the spatial separation of the carriers while the oscillator strength is more sensitive to this separation.

We have limited our study to the first conduction-electron state; however, a second state exists in DQWD's. In (001) DQWD's, e_2 remains coupled to e_1 and a barrier larger than 47 Å is required in order to weakly confine it (see Fig. 1). For the (111)B orientation, this e_2 state remains localized in the second well (see Fig. 2) and the overlap with the hh_1 and hh_2 states remains smaller than the overlap with e_1. In the last part of this study, we compare results obtained using the two methods: the simple ($|\langle \chi_e | \chi_h \rangle|^2$) or the complete [$\int \chi_{eh}^2(\xi,\xi) d\xi$] calculations. On Fig. 6, the square of the overlap integrals is plotted as a function of the internal barrier thickness. We see that, for (001) growth, the influence of the electrostatic interaction is far from being negligible and strongly reacts to compensate the dipole created by the electric field. This causes important modification of this overlap. Besides this, the effect on the Rydberg is smaller and has not been given here. For the (111)B case, the correction is extremely small, due to the strength of the built-in piezoelectric field. By analogy with the case of the e_1-lh exciton we studied elsewhere,[9] we can distinguish two regimes: (i) a type-I regime when the excitonic parameters are strong and when the interband overlap is larger than 50%, and (ii) a type-II regime for the opposite situation. This is the case for the fundamental interband transitions in (111)B DQWD's.

IV. CONCLUSIONS

We have calculated the excitonic properties of (Ga,In)As-GaAs double-quantum-well diodes in p-i-n structures. We have shown that the growth orientation has a dramatic influence on them, due to the possible existence of a built-in piezoelectric field in the strained layers. This influence appears (i) at the scale of the valence confinement masses of the holes, (ii) at the scale of the escape time of the carrier out of the DQWD's, due to the existence of the p-i-n field, which may or not compete with the built-in piezoelectric field, and (iii) at the scale of the strength of optical transitions, which are better estimated if a complete calculation is done including the effect of the exciton dipole on the conduction- and valence-band lineups.

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