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Sign inversion of the Stark shift in single non-abrupt $GaAs/Al_xGa_{1-x}As$ quantum wells

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Abstract. Stark shifts are calculated for the ground and first excited electron energy levels in single $GaAs/Al_{0.35}Ga_{0.65}As$ quantum wells with non-abrupt interfaces, which is a more appropriate picture for actual samples. The quantum-confined Stark shift for the ground-state electron energy level decreases as the non-abrupt interfaces become larger, but it is always negative. In striking contrast with this behaviour, the existence of non-abrupt interfaces can change the sign of the quantum-confined Stark shift for the first excited energy level in comparison to that calculated considering abrupt interfaces. These effects are shown to be stronger in the case of symmetric rather than asymmetric non-abrupt interfaces.

Although $GaAs/Al_xGa_{1-x}As$ quantum well structures are very promising for technological applications like in lasers and optical switching, the actual crystal growth processes are incapable of providing abrupt and/or roughness-free interfaces [1–3]. Consequently, non-abrupt interface potentials and carrier effective-mass profiles, with or without superimposed roughness, as well as the existence of islands, have to be taken into account if better agreement between experimental and theoretical results is sought. More recently, it was shown by experiments that the interfaces in AlAs/GaAs(001) systems are not symmetrical, i.e. the widths of normal (AlAs deposited on GaAs) and inverted (GaAs deposited on AlAs) interfaces are not the same, the latter being thinner than the former [4].

When an electric field is applied to a semiconductor quantum well, the electron ground-state energy decreases and the electron wave function is shifted against the field direction [5,6]. This double influence of an electric field on the states of a semiconductor quantum well is called the quantum-confined Stark effect. The observation of Stark shifts in quantum well intersubband transitions by Harwit and Harris Jr [7] presented a clear departure from calculated values based on the abrupt interface picture. The calculated intersubband absorption energy peaks were shown to be ∼15 meV smaller than the measured values. In order to achieve a better agreement, the well widths in the theoretical calculations of Harwit and Harris Jr [7] were assumed to be 107 Å, smaller than the value 120 Å estimated for their $GaAs/Al_{0.5}Ga_{0.5}As$ quantum well samples. Recently, Li *et al* [8] found that the existence of graded interfaces can change the 1S exciton Stark shift in semiconductor quantum wells [8]. By considering a AlGaAs/GaAs quantum well with an interdiffusion length of 40 Å, Li *et al* [8] showed, for a 100 Å single interdiffused GaAs quantum well modelled by a confinement profile based on an

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error function, that the 1S exciton Stark shift energy is twice as large as that produced by an 86 Å equivalent square well for an applied electric field of 50 kV cm^{-1} [8].

The main purpose of this work is to investigate the influence of non-abrupt interfaces on the quantum-confined Stark shift (QCSS) for the ground and the first excited electron energy states in single GaAs/Al_xGa_{1-x}As quantum wells. To the knowledge of the authors, this is the first time that Stark shift calculations have been performed considering the existence of non-abrupt interfaces in GaAs/Al_xGa_{1-x}As quantum wells, a picture which seems more appropriate for actual quantum wells since the interface widths of the best grown samples are not sharp (abrupt). The existence of non-abrupt interfaces is considered here as a consequence of the heterostructure growth process that produces an interfacial aluminium molar fraction variation. The model of a single non-abrupt GaAs/Al_xGa_{1-x}As quantum well is based on the interface description of Freire, Auto, and Farias [9]. Numerical results are obtained using the multistep approach of Ando and Itoh [10]. We also perform a detailed study on the dependence of the QCSS for the ground and the first excited electron energy states on the well width, the electric field intensity, and the interfacial asymmetry. The present theoretical calculations show that the existence of non-abrupt interfaces reduces the QCSS for the ground-state electron energy level, and can be responsible for a sign change in the QCSS for the first excited electron energy level. Finally, it is found that the interfacial asymmetry decreases the interface-related contributions to the QCSS in GaAs/Al_xGa_{1-x}As quantum wells.

After the sample growth process or annealing at moderate temperatures has been proceeding for a short time, the interdiffusion of Al and Ga across the interfaces does not significantly change the fractional aluminium composition in the GaAs region (this assumption is not valid for the GaAs/Al_xGa_{1-x}As interfacial region). The depth V_x of the quantum well is determined exclusively from the constant aluminium molar fraction x of the Al_xGa_{1-x}As alloy, in contrast to the case in the work of Li *et al* [8]. According to the interface model of Freire, Auto, and Farias [9], it can be assumed that the aluminium molar fraction changes linearly in the interfaces [11]. The potential and electron effective-mass expressions that describe a single asymmetric GaAs/Al_xGa_{1-x}As non-abrupt quantum well subjected to an electric field E_F applied in the growth direction z are given by the expressions of table 1.

Table 1. Interface potential and electron effective-mass expressions for a single non-abrupt GaAs/Al_xGa_{1-x}As quantum well subject to an electric field E_F.

Growth direction z	Mass $m(z)/m_0$	Potential $V(z)/C$
$z \leqslant -l/2$	$\mu_1 + \mu_2 x$	$V_x - eE_F z$
$-l/2 \leqslant z \leqslant a - l/2$	$\mu_{0,a} - \mu_{1,a} z$	$V_{0,a} + (V_{1,a} - eE_F)z + V_{2,a} z^2$
$a - l/2 \leqslant z \leqslant -b + l/2$	μ_1	$-eE_F z$
$-b + l/2 \leqslant z \leqslant l/2$	$\mu_{0,b} + \mu_{1,b} z$	$V_{0,b} + (-V_{1,b} - eE_F)z + V_{2,b} z^2$
$z \geqslant l/2$	$\mu_1 + \mu_2 x$	$V_x - eE_F z$

The parameters in table 1 are defined according to the following relations:

$$\mu_{0,s} = \mu_1 - \mu_{1,s}\frac{(l - 2s)}{2} \tag{1.1}$$

$$\mu_{1,s} = \mu_2\left(\frac{x}{s}\right) \tag{1.2}$$

$$V_x = \varepsilon_1 x + \varepsilon_2 x^2 \tag{2.1}$$

$$V_{0,s} = \left(\frac{l-2s}{2s}\right)^2 \varepsilon_2 x^2 - \left(\frac{l-2s}{2s}\right)\varepsilon_1 x \tag{2.2}$$

$$V_{1,s} = \left(\frac{l-2s}{s^2}\right)\varepsilon_2 x^2 - \left(\frac{\varepsilon_1}{s}\right)x \tag{2.3}$$

$$V_{2,s} = \frac{\varepsilon_2}{s^2}x^2 \tag{2.4}$$

where C is the electron band offset, e is the electric charge, and m_0 is the free-space mass of the electron; l is the width of the abrupt quantum well, and $s = \{a, b\}$ is the ensemble of interfacial widths a, b (see figure 1); $\varepsilon_1, \varepsilon_2$ (μ_1, μ_2) are experimental parameters related to the dependence of the $Al_xGa_{1-x}As$ energy gap in the Γ direction (electron effective mass) on x [12], and E_F is the applied electric field intensity. The centre of the single non-abrupt $GaAs/Al_xGa_{1-x}As$ quantum well is at $z = 0$, where the potential is always zero.

A graphical representation of the potential of the single non-abrupt $GaAs/Al_xGa_{1-x}As$ quantum well and the electron effective mass is depicted in figure 1. The external non-abrupt quantum well borders coincide with those of an abrupt quantum well, in agreement with the

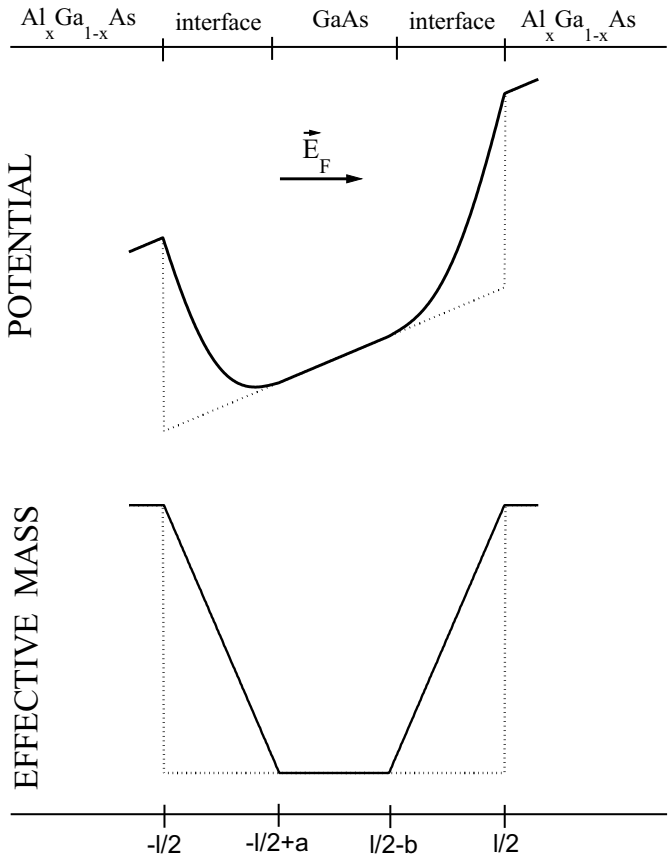

Figure 1. The electron potential (top) and effective-mass (bottom) representation of an abrupt (dotted) and a non-abrupt (solid) $GaAs/Al_xGa_{1-x}As$ quantum well subject to an electric field E_F. l is the width of the abrupt well, and a (b) the width of its left-hand (right-hand) interface.

interface positioning used by Proctor *et al* [13]. It is worth mentioning that the majority of previous works on interface effects in semiconductor quantum wells consider that it is the middle of the interfacial region in a non-abrupt quantum wells that determines the interface positioning of the abrupt quantum well—see references [14–17], for example. Measurements performed on quantum well samples (that are actually non-abrupt) are then compared to the properties calculated within an abrupt quantum well picture (as defined previously, for example).

The usual definition of an abrupt quantum well has to be considered in fact as a definition of an *abrupt equivalent quantum well* (AEQW), i.e., an abrupt well whose energy levels are as close as possible to those of its associated non-abrupt quantum well—see Delalande and Bastard [19]. It is argued here that it should not be used for a direct description of actual quantum wells in semiconductor samples, as it usually is [1]. Since an actual GaAs/Al_xGa_{1-x}As quantum well must begin when the aluminium molar fraction $\mathcal{X}(z)$ starts to be smaller than the aluminium molar content x of the Al_xGa_{1-x}As alloy, the definition used in this paper and by Proctor *et al* [13] seems to be more appropriate than those usually considered (see references [14–17], for example). In this case, the width of the abrupt quantum well as defined in this work and by Proctor *et al* [13] is closely related to the experimentally estimated width of the quantum well in a semiconductor sample. Consequently, one can argue that with this definition of abrupt quantum well borders, the abrupt quantum well widths as defined in this paper have to be closer to the widths of quantum well semiconductor samples (as determined from experiments) than to the widths of the definition of the *abrupt equivalent quantum well* (AEQW), i.e., the halfway borders definition [14–17]. It must be stressed that the usual abrupt interface positioning is responsible for a theoretical underestimation of effects related to the existence of interfaces in semiconductor heterostructures. The abrupt quantum well as defined here can contribute to explaining why Harwit and Harris Jr [7] have considered a shorter well width to improve the agreement between their measurements and their theoretical calculations based on the abrupt interface picture. Taking into account the existence of non-abrupt interfaces in the samples used by Harwit and Harris Jr [7] in their experiments, the AEQW border positioning results in a shorter well width.

As a consequence of the model of a single non-abrupt GaAs/Al_xGa_{1-x}As quantum well considered in this work (see figure 1), the interfacial electron effective mass is position dependent. To describe this dependence, the BenDaniel and Duke [18] kinetic energy operator, $-(\hbar^2/2)(\mathrm{d}/\mathrm{d}z)[m(z)]^{-1}(\mathrm{d}/\mathrm{d}z)$, seems to be the best choice and it is also the one most used in the literature. The energy levels of the single non-abrupt GaAs/Al_xGa_{1-x}As quantum wells subjected to an electric field are calculated by solving the Schrödinger-like equations through the transfer-matrix scheme as proposed by Ando and Itoh [10]. Single abrupt and both interfacial symmetric ($a = b$) and asymmetric ($a \neq b$) non-abrupt GaAs/$Al_{0.35}Ga_{0.65}$As quantum wells are considered with well widths in the range 80 Å $< l <$ 140 Å, an electron band offset $C = 0.6$, and interface widths up to four GaAs lattice parameters (LP), where 1 LP = 5.653 Å. The $Al_{0.35}Ga_{0.65}$As experimental parameters for the interface model are $\mu_1 = 0.067$, $\mu_2 = 0.083$, $\varepsilon_1 = 1.155$, and $\varepsilon_2 = 0.37$ at 300 K [12].

The QCSS dependences for the ground state and the first excited electron energy level on the interface widths are presented in figure 2. In this figure, the abrupt quantum well width is 100 Å and the interfaces are symmetric ($a = b$). Its left-hand scale refers to the ground-state energy level curves, the solid ones for quantum wells as defined in this work, and the dashed ones for the AEQW as defined by Delalande and Bastard [19]. The right-hand scale in figure 2 refers to the first excited energy level curves, the dotted ones for quantum wells as defined in this work, and the dotted–dashed ones for the AEQW [19]. The QCSS for the ith energy states are given by $\Delta_i = E_i(E_F) - E_i(E_F = 0\ \mathrm{kV\ cm^{-1}})$. It is shown that an increase of the

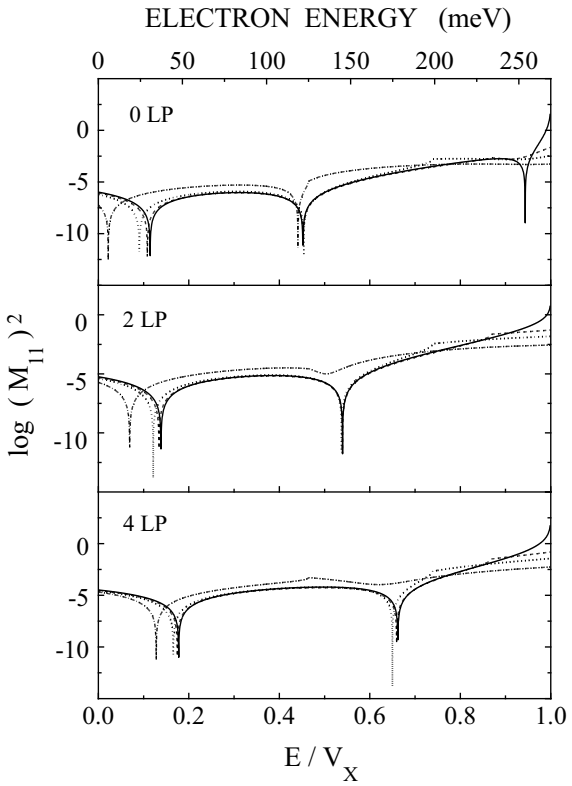

Figure 2. The electron energy levels in the ground (solid) and first excited (dotted) states of abrupt ($a = b = 0$ LP) and non-abrupt ($a = b = 2, 4$ LP) GaAs/$Al_{0.35}Ga_{0.65}$As single quantum wells subjected to an electric field in the range 0 kV cm^{-1} $< E_F <$ 100 kV cm^{-1}. Also included are the electron energy levels of the ground (dashed) and first excited (dotted–dashed) states of an abrupt equivalent GaAs/$Al_{0.35}Ga_{0.65}$As single quantum well whose width is $l - a$. The left-hand scale refers to the ground-state energy level curves (solid and dashed), and the right-hand scale refers to the first excited energy level curves (dotted and dotted–dashed).

interface widths reduces the QCSS for the ground-state electron energy level (Δ_0), as can be seen by comparing the bowing of the solid curves in figure 2, but it is always negative. This is due to the quantum well shortening related to the existence of non-abrupt interfaces. On the other hand, the QCSS first excited electron energy level (Δ_1) in the case of abrupt interfaces is very small and positive (see the lowest dotted curve in figure 2). The existence of interfaces in this case changes both the intensity and the sign of the QCSS Δ_1, as shown by the two highest dotted curves in figure 2. The sign change occurs for an interface width larger than or of the order of two GaAs LP, which is common for actual GaAs/Al_xGa_{1-x}As samples [20, 21]. In contrast to the negative QCSS Δ_0, the QCSS Δ_1 increases when the interface becomes larger (at least up to a 100 kV cm^{-1} electric field intensity as shown in figure 2), as can be seen by comparing Stark shifts calculated for 100 Å wells with 2 LP and 4 LP interfaces.

Delalande and Bastard [19] have suggested that the energy levels of graded non-abrupt quantum wells are very close to those of an AEQW. To investigate this suggestion, the energy levels of the AEQW of width $l - a$ were calculated—see the dashed curves for the ground-state energy level and the dotted–dashed curves for the first excited energy level in figure 3. One

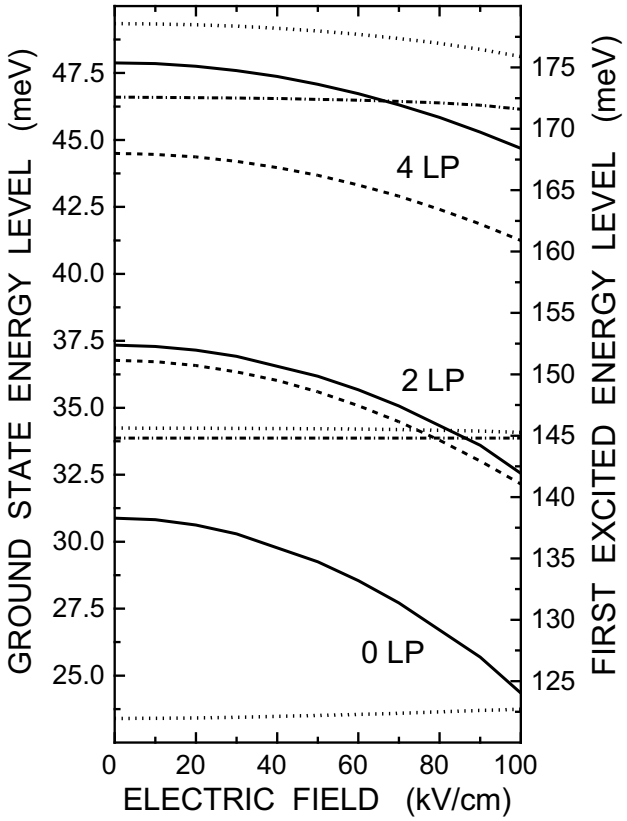

Figure 3. The Stark shift Δ_0 of the electron ground state in GaAs/$Al_{0.35}Ga_{0.65}$As single quantum wells whose widths are (a) 80 Å, (b) 100 Å, (c) 120 Å, and (d) 140 Å. The widths of the interfaces are 0 LP (dashed); 2 LP (solid); and 4 (dotted–dashed).

can observe that the energy levels of an AEQW are always smaller than those of non-abrupt quantum wells (the dashed curves are always below the solid curves). When $a \sim 2$ LP, the difference between the ground (first excited) energy level of the non-abrupt quantum well as defined in this work and the AEQW is of the order of 0.6 meV (0.8 meV). The difference is almost independent of the applied electric field intensity, and it is as important as that related to small random alloy fluctuations of the aluminium content in the alloy $Al_xGa_{1-x}As$. When $a = 4$ LP, the difference between the ground-state (first excited) energy level of the non-abrupt and the AEQW is of the order of 3.5 meV (5.0 meV). As can be seen in figure 2, the suggestion of Delalande and Bastard [19] is very good when the interface widths are smaller than 2 LP.

Non-abrupt interface effects on the QCSS Δ_0 can be observed easily in figure 3, where one can see that the variation of the QCSS Δ_0 is always smaller when the existence of interfaces is taken into account. It is shown that the interface effects turn out to be more important when the electric field intensity and the width of the quantum well increase. While the existence of a 2 LP interface in a 80 Å quantum well is responsible for a QCSS Δ_0 of -0.6 meV (-2.5 meV) when the electric field intensity $E_F = 50$ kV cm^{-1} ($E_F = 100$ kV cm^{-1}), the same interface in a 140 Å quantum well is responsible for a QCSS Δ_0 of -3.4 meV (-12.6 meV) when the electric field intensity $E_F = 50$ kV cm^{-1} ($E_F = 100$ kV cm^{-1}). When the interface width

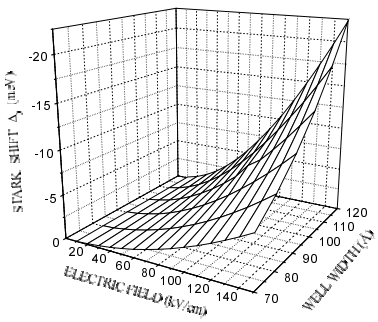

Figure 4. The Stark shift Δ_1 of the electron first excited state in abrupt and non-abrupt $GaAs/Al_{0.35}Ga_{0.65}As$ single quantum wells whose widths are (a) 80 Å, (b) 100 Å, (c) 120 Å, and (d) 140 Å. The widths of the interfaces are 0 LP (dashed); 2 LP (solid); and 4 (dotted–dashed).

is 4 LP in the 80 Å quantum well, there is a QCSS Δ_0 of -0.4 meV (-1.8 meV) when the electric field intensity $E_F = 50$ kV cm^{-1} ($E_F = 100$ kV cm^{-1}), while the same interface in a 140 Å quantum well is responsible for a QCSS Δ_0 of -2.2 meV (-9.0 meV) when the electric field intensity $E_F = 50$ kV cm^{-1} ($E_F = 100$ kV cm^{-1}).

The role of non-abrupt interfaces on the QCSS for the first excited electron energy levels (Δ_1) can be observed in figure 4. In this case, one can see that the Stark shifts for the first excited electron energy levels in the non-abrupt wells are bigger than those of the abrupt ones for any electric field intensity and well width. This is in striking contrast with the interface dependence of the QCSS for the electron ground-state energy levels. The QCSS Δ_1 is always positive (negative) for abrupt and non-abrupt wells with widths $\gtrsim$140 Å ($\lesssim \sim$80 Å). When the well width is 100 Å wide, the QCSS Δ_1 is positive only when the interfaces are abrupt, and is negative when the interfaces are non-abrupt (see figure 4(b)). While the existence of a 2 LP interface in a 80 Å quantum well is responsible for a QCSS Δ_1 of -1.2 meV (-4.1 meV) when the electric field intensity $E_F = 50$ kV cm^{-1} ($E_F = 100$ kV cm^{-1}), the same interface in a 140 Å quantum well is responsible for a QCSS Δ_1 of 0.67 meV (1.8 meV) when the electric field intensity $E_F = 50$ kV cm^{-1} ($E_F = 150$ kV cm^{-1}). When the interface width is 4 LP in the 80 Å quantum well, there is QCSS Δ_1 of -3.4 meV (-5.2 meV) when the electric field intensity $E_F = 50$ kV cm^{-1} ($E_F = 150$ kV cm^{-1}), while the same interface in a 140 Å quantum well is responsible for a QCSS Δ_1 of 0.3 meV (0.5 meV) when the electric field intensity $E_F = 50$ kV cm^{-1} ($E_F = 150$ kV cm^{-1}).

The results presented in figure 4 show that the QCSS for the first electron excited state is sensitive to the well and interface (in the case of the non-abrupt quantum well) widths. As pointed out by Matsuura and Kamizato [22] for abrupt quantum wells, this behaviour is related to the existence of a large amplitude of the wave function for the first excited state in the $z < 0$ direction, even for large electric fields. It is this part of the wave function that is associated to the positive QCSS for the quantum well excited states, as was shown by Matsuura and Kamizato [22]. However, they considered only abrupt wells. In this work, it was observed that the sign of the QCSS Δ_1 is very sensitive to the interface width, which means that the oscillation of the QCSS for the first excited state depends on the interface width.

Since interfaces with $Al_xGa_{1-x}As$ deposited on GaAs are wider than interfaces with GaAs deposited on $Al_xGa_{1-x}As$ [4], we have also studied in this work the influence of asymmetric interfaces on the QCSS Δ_0 and Δ_1 in 100 Å $GaAs/Al_{0.35}Ga_{0.65}As$ quantum wells subjected to 100 kV cm^{-1}. The QCSS Δ_0 and Δ_1 for the abrupt case ($a = b = 0$) are plotted as

dashed curves in figure 5 for the sake of comparison. The increasing of the ratio b/a means the accentuating of the interfacial asymmetry characteristics of the wells, which produces an important augmentation of the Stark shift. The influence of the interfacial asymmetry is more important for higher energy level states. When $a = 1$ LP, a 50% degree of interfacial asymmetry ($b/a = 2$) produces an increase of the QCSS Δ_0 (Δ_1) of the order of 1.3 meV (1.8 meV). When $a = 2$ LP, the same degree of interfacial asymmetry produces an increase of the QCSS Δ_0 (Δ_1) of the order of 2.7 meV (3.5 meV).

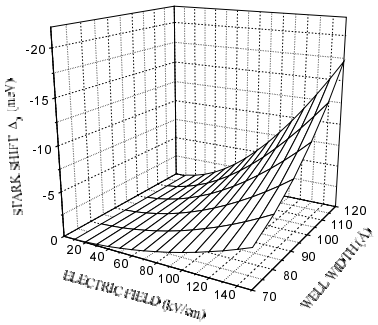

Figure 5. The dependences of the electron Stark shifts Δ_0 and Δ_1 on the degree of interfacial asymmetry. The 100 Å wide GaAs/$Al_{0.35}Ga_{0.65}$As single quantum wells are subjected to a 100 kV cm^{-1} electric field. The widths of the quantum well interfaces a are: 0 LP (dashed); 1 LP (solid); and 2 LP (dotted–dashed).

It is worth stressing that, although the possibility of a positive Stark shift for the first excited state was previously predicted and explained by Matsuura and Kamizato [22], they have not investigated the conditions for its existence related to the well width, electric field intensity, and thickness of the non-abrupt interfaces. The results presented in figures 2–5 seem to be the most complete investigation performed on this subject. Although a qualitative understanding of the results presented is easily achieved by relating the existence of interfaces to an effective shortening of the quantum well, the model used in this work allows a quantitative study.

Since recent experiments have shown that GaAs/Al_xGa_{1-x}As interface widths are at least of the order of 2 LP [20,21]; the results obtained here suggest that to achieve a better agreement between the theoretical and experimental results as regards Stark effects, i.e., to achieve an agreement better than 3 meV (2 meV if interfacial asymmetry is taken into account) between the theoretically calculated and experimentally measured Stark shifts, it is necessary to have not only an estimate of the actual interface widths of the quantum well semiconductor samples, but also to use a non-abrupt model for the interfaces in the theoretical calculations. Recently, Xie *et al* [23] have calculated the ground-state energy and the binding energy of an exciton in a GaAs/Al_xGa_{1-x}As quantum well under an electric field. They have found that the energy corrections related to different phonon modes are smaller than 3 meV [23]. Since the shifts of the electron energy levels calculated in this work considering interface effects are of the order of 3 meV, interface-related corrections to Stark shifts in GaAs/Al_xGa_{1-x}As quantum wells are more important than or at least as important as phonon-related corrections.

Excitons in quantum wells do exist even when high electric fields are applied [6]. The confined exciton energy in GaAs/Al_xGa_{1-x}As quantum wells is given by $E_{ex} = E_g + E_e + E_h + E_{e-h}$, where E_g is the GaAs gap energy, E_e (E_h) is the electron (hole) energy level in the quantum well, and E_{e-h} is the energy associated with the electron–hole interaction. The Stark shifts of the carrier energies produce the exciton energy E_{ex}, which can turn out to be

bigger than +5 meV, which seems high enough to be detected by photoluminescence and/or absorption measurements. Accordingly, photoluminescence and/or absorption measurements can be useful for detecting sign inversion and/or non-abrupt interface effects on the Stark shifts in single non-abrupt $GaAs/Al_xGa_{1-x}As$ quantum wells. A strong indication of this possibility is contained in the work of Yang *et al* [24], where electric field effects on the photoluminescence in modulation-doped pseudomorphic AlGaAs/InGaAs/GaAs single quantum wells were investigated. However, they restricted their experiments to voltages smaller than 1 V. A detailed study of electric field effects on the exciton energy in non-abrupt $GaAs/Al_xGa_{1-x}As$ quantum wells is beyond the scope of this work, but will be a subject of a forthcoming paper by the authors.

In conclusion, we have presented a study of the Stark effect in single abrupt and non-abrupt $GaAs/Al_{0.35}Ga_{0.65}As$ quantum wells. It was shown that the existence of non-abrupt interfaces considerably reduces the Stark shift of the electron ground-state energy level, without changing the sign of the shift. In contrast, both the values and the sign of the Stark shift of the first excited electron energy level change when interfaces are taken into account. It was also demonstrated that the abrupt equivalent-quantum-well description can give an estimate of the energy levels of actual $GaAs/Al_xGa_{1-x}As$ single-quantum-well samples only when their interfaces are certain to be thinner than 2 LP. Although obtained for the specific case of $GaAs/Al_{0.35}Ga_{0.65}As$ single quantum wells, the results presented is this work should also be valid for other types of semiconductor quantum wells, and are worth confirming experimentally.

Acknowledgments

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