Operator ordering of a position-dependent effective-mass Hamiltonian in lattice-matched semiconductor superlattices and quantum wells

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ABSTRACT

The position-dependent effective mass Hamiltonian $H = -(\hbar/2)[m(z)]^{\alpha}\nabla[m(z)]^{\beta}\nabla[m(z)]^{\alpha}+V(z)$ with $2\alpha + \beta = -1$ is applied to the problem of periodic heterostructure with abrupt interfaces and discontinuous mass distribution. In order to determine the most suitable operator ordering, numerical results for interband and intersubband transition energies are compared with experimental data for various GaAs/$Al_xGa_{1-x}As$ superlattices and quantum wells. The ordering-related energy shift as a function of structural parameters (well thickness, barrier thickness and height) is investigated. We find that variation of kinetic energy operator ordering can cause transition energy shift exceeding 40 meV. The model with $\alpha = 0$ and $\beta = -1$ consistently produces the best fit to experimental results.

Keywords: Operator ordering, quantum wells, superlattices

1. INTRODUCTION

The effective-mass theory (EMT) is an important and widely used tool in semiconductor physics. Originally developed for analysis of homogenous crystals [Slater 1949], [James 1949], EMT when applied to heterostructures with position-dependent effective mass becomes asymptotically exact when perturbations to the electron and hole states are sufficiently smooth. The entire approach would have to be modified in order to accommodate the case of structures with abrupt heterointerfaces [Burt 1992], [Burt 1994]. In spite of that, EMT continues to be widely used as a major computational tool to determine dynamic and static electrical, electronic, and optical properties of many abrupt-interface heterostructures, including superlattices and quantum wells. It has been recognized for a long time [Morrow 1987a], [Einevoll 1990a], [Einevoll 1990b] that application of EMT to abrupt interfaces suffers from ambiguity in kinetic energy operator (KEO) ordering, caused by non-vanishing commutator of the momentum operator $-i\hbar\nabla$ and the position-dependent effective mass $m(z)$. This leads to non-uniqueness of KEO, which in its most general form can be written as [von Roos 1983]

$$K = -\frac{\hbar^2}{4}\left[m^{\alpha}\nabla m^{\beta}\nabla m^{\gamma} + m^{\gamma}\nabla m^{\beta}m^{\alpha}\right] , \tag{1}$$

with $\alpha + \beta + \gamma = -1$. By construction, the KEO (1) is Hermitian. For abrupt interfaces with discontinuous step-like distribution of effective masses, α must be equal to γ, as otherwise the wave function would have to vanish at heterointerfaces, which would have been clearly unphysical [Morrow 1984]. Also, unless $\alpha = \gamma$, the ground-state energy diverges in the limit of abrupt interfaces [Thomsen 1989]. This reduces Eq. (1) to

$$K = -\frac{\hbar^2}{2}[m(z)]^{\alpha}\nabla[m(z)]^{\beta}\nabla[m(z)]^{\alpha} , \tag{2}$$

with $2\alpha + \beta = -1$. Possible values of the β parameter span the range from 0 to -1. Corresponding to this one-

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parameter family of operators, the matching conditions for the envelope wave function $F(z)$ and its derivative $dF(z)/dz$ are also parameterized, with continuity of $[m(z)]^{\alpha}F(z)$ and $[m(z)]^{\beta}d(([m(z)]^{\alpha}F(z))/dz$ at the interfaces [Einevoll 1990a].

Considerable effort has been expended in order to establish a preferred value of β, with the expectation that this value should be universal and should not depend on particular materials forming the abrupt interfaces. Various authors arrived, however, at diametrically opposed conclusions, with either $\beta = 0$ or $\beta = -1$ judged to be the most plausible choice. It should be noted that the requirement of continuity of the probability current across an abrupt interface cannot be used as criterion for selecting the value of β, as it leads to a boundary condition which is independent of ordering [Einevoll 1990a].

The $\beta = 0$ conclusion was reached by reformulating the connection rule problem by first extrapolating the envelope function on either side of a heterointerface as if the semiconductor were homogenous [Zhu 1983], and by comparing the results of Kronig-Penney's calculation of electron transmission through a set of δ-function scatterers with the transmission coefficient obtained using the EMT [Morrow 1987a].

The opposite conclusion, with $\beta = -1$, was reached by using the Kohn-Luttinger representation and canonical transformation [von Roos 1985], by studying matching conditions for the wave function across an abrupt heterojunction in three dimensions [Morrow 1987b], by considering inherent limitations for consistency in an extremely narrow δ-well [Thomsen 1989], by comparing exact microscopic band structure calculations with the corresponding EMT results for superlattices, quantum well and localized potentials [Einevoll 1990a], by applying instantaneous Galilean invariance [Lévy–Leblond 1995], and by considering the abrupt limit of analytical solution for a continuous and smoothly varying potential and mass step [Dekar 1999]. It should also be noted that $\beta = -1$, corresponding to the Ben Daniel-Duke Hamiltonian [BenDaniel 1966], is the ordering obtained for slowly-varying inhomogeneous semiconductors by using a basis set that exactly diagonalizes the Hamiltonian in the homogeneous limit [Young 1989].

Other, intermediate values of β have also been suggested. For example, considerations of a nonrelativistic limit of the Dirac Hamiltonian with position-dependent mass taken into account via the Foldy-Wouthuysen approximation, led to adoption of $\alpha = 0$, $\beta = -½$, and $\gamma = -½$ [Cavalcante 1997], violating the $\alpha = \gamma$ rule of Morrow and Brownstein. Cavalcante *et al.* found that for $GaAs/Al_{0.3}Ga_{0.7}As$ heterojunctions with interface regions at least two lattice constants thick, there is virtually no difference between various KEO models. They write: "..present results suggest that no experiment can be performed nowadays on semiconductor samples to determine the KEO form in systems with spatially varying effective mass since the actual interface widths preclude this possibility." We would argue that contemporary growth technologies are so advanced that it is possible to grow heterostructure with extremely well-defined interfaces. Considering electron transmission, Cavalcante *et al.* note that the transmission is sensitive to KEO only when an abrupt interface is considered. However, we believe it is essential to include discontinuity of the effective mass distribution when treating an abrupt interface (unless the heterostructure is severely degraded):

$$m(z) = m_1 + \Delta m_{12}\Theta(z) \quad , \tag{3}$$

where $\Theta(z)$ is the Heaviside step-like function, m_1 is the effective mass in one of the two materials, and Δm_{12} is the difference of effective masses for those materials.

Given that in the framework of EMT different initial assumptions may lead to different KEO laws, it is questionable whether a general solution exists. Nevertheless, it is always possible to compare the theoretical predictions for β with the experimental results in order to determine the correct form of parameterized KEO. In contrast to a large body of literature containing theoretical considerations of KEO ordering, there have been only a few attempts to settle the issue by using experimental data. Galbraith and Duggan compared the calculated optical transition energies with low-temperature photoluminescence excitation spectra of $GaAs/Al_{0.35}Ga_{0.65}As$ quantum wells [Galbraith 1988]. Their results for two samples (well thickness of 5 and 7 nm) indicated $\beta = -1 \pm 0.1$ and $\beta = -1.1 \pm 0.5$. Unfortunately, their set of parameters was not completely published so we could not repeat their results. In another work, Fu and Chao reported that experimentally observable interband transition energies were not sensitive to the choice of β [Fu 1989]. In this paper, we demonstrate that, contrary to Fu and Chao's assertion, the interband

transition energies do vary substantially with β. In addition, we show that within the framework of EMT for abrupt interfaces and discontinuous mass distribution, the intersubband transition energies can also vary substantially with β. Comparison with available data confirms that the choice of β = -1 provides the best fit to experimental results.

This paper is organized as follows. In Section 2 we formulate our theoretical model for superlattices and quantum wells based on the transfer matrix approach. The model is then used in Section 3 to calculate the miniband energies for $GaAs/Al_xGa_{1-x}As$ superlattices. In Section 4, we study the ordering-related effects and determine conditions for maximum sensitivity of transition energies to the choice of β for various AlGaAs/GaAs superlattice parameters (well length, barrier height, and barrier thickness). Finally, Section 5 contains a comparison of theoretical results with available experimental data, which is then used to deduce the best value for the ordering parameter β. Concluding remarks are given in Section 6.

2. THEORETICAL MODEL

Consider a periodic superlattice that consists of lattice-matched semiconductors 1 and 2 (*e.g.*, the $GaAs/Al_xGa_{1-x}As$ system, illustrated in Fig. 1.). The thicknesses of alternating well and barrier layers are d_1 and d_2, respectively, with the superlattice period denoted as $d = d_1 + d_2$. When d_2 is large enough (≈15-20 nm), the quantum wells are practically decoupled and the superlattice problem becomes equivalent to a single quantum well problem. The interfaces are assumed to be parallel to the *x-y* plane, while the superlattice axis is oriented along the *z* direction. The Schrödinger equation for an electron in the conduction band is given by

$$\{K + E_c(z) - E\}F_c(z) = 0 \quad , \tag{4}$$

with the KEO K given by Eq. (2), $E_c(z)$ standing for the bulk conduction band edge profile, $F_c(z)$ denoting the envelope wave function, and E representing the energy eigenvalue. Analogous equations can be written for the light- and heavy holes in the valence band. In the simplest case of rectangular wells and barriers (see Fig. 1), Eq. (4) takes the form

$$\left[-\frac{\hbar^2}{2m_{cj}}\left(\frac{d^2}{dz^2}\right) + E_{cj} - E\right]F_{cj}(z) = 0 \qquad \begin{cases} 0 < z < d_1 & j = 1 \\ d_1 < z < d_1 + d_2 & j = 2 \end{cases} \tag{5}$$

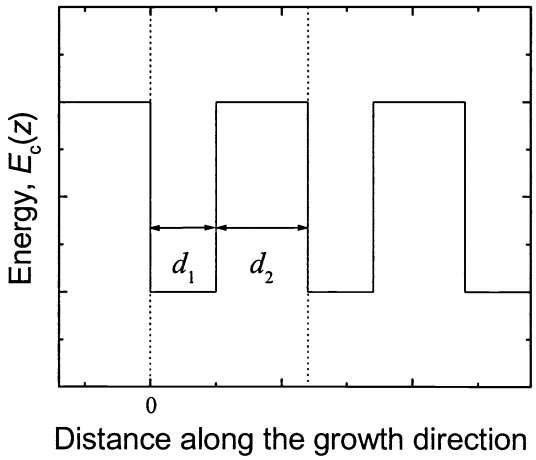

Fig. 1. Schematic representation of bulk conduction band edge for periodic superlattice structure. Alternating layers of two lattice-matched materials have thickness d_1 for the well and d_2 for the barrier, respectively.

Although β no longer appears explicitly in Eq. (5), it is still present in the boundary conditions connecting the solutions F_{c1}, F_{c2} at the interfaces. The miniband structure of superlattices with arbitrary well and barrier profiles can

be conveniently found using the well-known transfer-matrix approach [Merzbacher 1997]. Each layer d_j (j = 1 or 2) is divided into N_j intervals of sufficiently small length Δ_j, and Eq. (5) is approximated by a difference equation leading to

$$\begin{bmatrix} F_j(d_1+\delta_{j2}d_2) \\ F'_j(d_1+\delta_{j2}d_2) \end{bmatrix} = \begin{bmatrix} 1 & \Delta_j \\ \dfrac{2m_j(E_{cj}-E)\Delta_j}{\hbar^2} & 1 \end{bmatrix}^{N_j} \times \begin{bmatrix} F_j[(j-1)d_1] \\ F'_j[(j-1)d_1] \end{bmatrix} = \tilde{P}_j^{N_j} \begin{bmatrix} F_j[(j-1)d_1] \\ F'_j[(j-1)d_1] \end{bmatrix} , \tag{6}$$

where δ_{j2} stands for the Kronecker symbol. The boundary conditions connecting the solutions at either side of an interface yield the following relations

$$\begin{bmatrix} F_2(d_1) \\ F'_2(d_1) \end{bmatrix} = \begin{bmatrix} (m_1/m_2)^{\alpha} & 0 \\ 0 & (m_1/m_2)^{\alpha+\beta} \end{bmatrix} \begin{bmatrix} F_1(d_1) \\ F'_1(d_1) \end{bmatrix} , \tag{7}$$

$$\begin{bmatrix} F_1(0) \\ F'_1(0) \end{bmatrix} = \begin{bmatrix} (m_2/m_1)^{\alpha} & 0 \\ 0 & (m_2/m_1)^{\alpha+\beta} \end{bmatrix} \begin{bmatrix} F_2(0) \\ F'_2(0) \end{bmatrix} . \tag{8}$$

Numerical solution of Eqs. (6)-(8) can be significantly simplified by diagonalizing the matrix $\tilde{P}_j$ and noting that its N_j-th power can be expressed in terms of the diagonal eigenvalue matrix $\tilde{\Lambda}_j$ (see [Strang 1988]):

$$\tilde{P}_j^{N_j} = \tilde{S}_j \tilde{\Lambda}_j^{N_j} \tilde{S}_j^{-1} , \tag{9}$$

where $\tilde{S}_j$ is a matrix formed by the eigenvectors of $\tilde{P}_j$, and the matrix $\tilde{\Lambda}_j$ is given by

$$\tilde{\Lambda}_j = \begin{bmatrix} \lambda_{j+} & 0 \\ 0 & \lambda_{j-} \end{bmatrix} . \tag{10}$$

λ_{j+} and λ_{j-} are the eigenvalues of $\tilde{P}_j$ given by

$$\lambda_{j\pm} = 1 \pm \frac{\Delta_j [2m_j(E_{cj}-E)]^{1/2}}{\hbar} . \tag{11}$$

Combining Eqs. (6)-(9), we obtain the transfer matrix $\tilde{T}(E)$ which translates the envelope function by one superlattice period comprising two interfaces:

$$\begin{bmatrix} F_2(d) \\ F'_2(d) \end{bmatrix} = \tilde{T}(E) \begin{bmatrix} F_2(0) \\ F'_2(0) \end{bmatrix} , \tag{12}$$

$$\tilde{T}(E) = \tilde{S}_2 \tilde{\Lambda}_2^{N_2} \tilde{S}_2^{-1} \begin{bmatrix} 1 & 0 \\ 0 & (m_1/m_2)^{\beta} \end{bmatrix} \times \tilde{S}_1 \tilde{\Lambda}_1^{N_1} \tilde{S}_1^{-1} \begin{bmatrix} 1 & 0 \\ 0 & (m_2/m_1)^{\beta} \end{bmatrix} . \tag{13}$$

Taking the origin z = 0 at the interface as indicated in Fig. 1, the eigenvalues of $\tilde{T}(E)$ can be calculated analytically as

$$\xi_{1,2} = \frac{1}{4}\left\{\left[\nu_1\nu_2 + \frac{1}{2}\mu_1\mu_2\left(MR + \frac{1}{MR}\right)\right] \pm \left[\left(\nu_1\nu_2 + \frac{\mu_1\mu_2}{2}\left(MR + \frac{1}{MR}\right)\right)^2 - \left(\nu_1^2\nu_2^2 + \mu_1^2\mu_2^2 - \mu_1^2\nu_2^2 - \mu_2^2\nu_1^2\right)\right]^{1/2}\right\} \quad (14)$$

where

$$\mu_j = \lambda_{j+}^{N_j} - \lambda_{j-}^{N_j} \ , \quad (15)$$

$$\nu_j = \lambda_{j+}^{N_j} + \lambda_{j-}^{N_j} \ , \quad (16)$$

$$M = \left(\frac{m_1}{m_2}\right)^{\beta} \ , \quad (17)$$

$$R = \left[\frac{m_1(E_{c1} - E)}{m_2(E_{c2} - E)}\right]^{1/2} \ . \quad (18)$$

Due to periodicity of the superlattice potential, the eigenvalues of $\tilde{T}(E)$ must satisfy the Bloch theorem with period d, *i.e.* they must have the form of exp ($\pm ikd$):

$$\xi_{1,2} = \exp(\pm ikd) = \cos(kd) \pm i\sin(kd) \ . \quad (19)$$

Eq. (19) in conjunction with Eq. (14) provides us with a dispersion relation between E and k. We note that if the reference zero potential is chosen to coincide with the bottom of the well, *i.e.* $E_{c1} = 0$, then μ_1 is a purely imaginary number of the form

$$\mu_1 = 2i\left(1 + \frac{2\Delta_1^2 m_1 E}{\hbar^2}\right)^{N_1/2} \sin(N_1\theta) \ , \quad (20)$$

where

$$\theta = \tan^{-1}\left(\sqrt{\frac{2\Delta_1^2 m_1 E}{\hbar^2}}\right) \ . \quad (21)$$

In addition, for E real and smaller than E_{c2}, (*i.e.* for confined states), R given in Eq. (18) is purely imaginary, while ν_1, ν_2, and μ_2 are all real. This implies that Eq. (19) can be further simplified (see [Burns 1985]) to

$$\frac{1}{4}\left[\nu_1\nu_2 + \frac{1}{2}\mu_1\mu_2\left(MR + \frac{1}{MR}\right)\right] = \cos(kd) \quad (22)$$

and

$$\frac{1}{4}\left\{\left[\nu_1\nu_2 + \frac{\mu_1\mu_2}{2}\left(MR + \frac{1}{MR}\right)\right]^2 - \left(\nu_1^2\nu_2^2 + \mu_1^2\mu_2^2 - \mu_1^2\nu_2^2 - \mu_2^2\nu_1^2\right)\right\}^{1/2} = \sin(kd) \ . \quad (23)$$

3. MINIBAND ENERGY CALCULATION

In this section, we will examine sensitivity of GaAs/$Al_xGa_{1-x}As$ superlattice band structure to the value of β parameter. The choice of GaAs/$Al_xGa_{1-x}As$ is dictated by the fact that this material system is the most extensively studied among all III-V semiconductors. Material parameters are known to a very good precision [Vurtgaftman 2001]. Furthermore, the lattice mismatch between GaAs and $Al_xGa_{1-x}As$ is very small, hence the correction of the miniband energy due to strain effects in negligible. The $Al_xGa_{1-x}As$ effective masses m_e, m_{hh}, m_{lh}, the bandgap energy E_g, and the band offset ratio ΔE_c:ΔE_v used in our calculations are listed in the Table 1 (see [Adachi 1985], [Bosio 1988], [Vurtgaftman 2001]). For comparison with the low-temperature experiments, we use the Varshni formula to estimate bandgap variation with temperature. The parameters appearing in the Varshni formula are taken from [Kangarlu 1988]. We use the approximation of a temperature-independent bowing parameter for $Al_xGa_{1-x}As$ (see [Vurtgaftman 2001]). In order to predict correctly the transition energy at low temperatures, it is imperative to take into account the exciton binding energies. Some of the experiments considered in our work provide this information alongside with their photoluminescence results. For the other cases, we consult [Gurioli 1993]. Generally, the exciton binding energy for the cases considered in this paper is in the range of 10-15 meV.

Table 1: Main $Al_xGa_{1-x}As$ material parameters

m_e/m_0	m_{hh}/m_0	m_{lh}/m_0	E_g [eV]	ΔE_c:ΔE_v
$0.0667 + 0.083x$	$0.34 + 0.412x$	$0.094 + 0.0667x$	$1.427 + 1.247x$	65:35

Fig. 2 depicts the dispersion curves for the conduction-band electrons in a GaAs/$Al_{0.25}Ga_{0.75}As$ superlattice. Dashed (β = 0) and solid (β = -1) curves indicate the ordering-related miniband energy shift. The thicknesses of GaAs wells and $Al_{0.25}Ga_{0.75}As$ barriers are d_1= 15 nm and d_2= 2.5 nm, respectively. Each layer is divided into $N_1 = N_2 = 30{,}000$ segments.

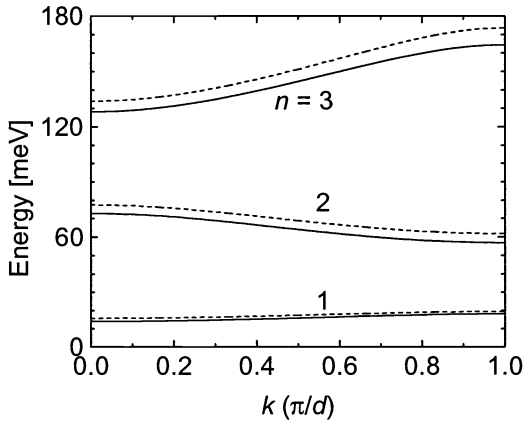

Fig. 2. Three lowest minibands CB_1, CB_2, and CB_3 for conduction-band electrons in a GaAs/$Al_{0.25}Ga_{0.75}As$ superlattice with d_1= 15 nm and d_2= 2.5 nm. The dashed curves correspond to β = 0, and the solid curves to β = -1.

As indicated in Fig. 2, the shift in the miniband energies for the cases of β=-1 and β=0 becomes larger as the miniband index increases. Thus, the higher minibands are more sensitive to the value of β than the lower minibands. For the case of Fig. 2, the calculated shift in energy at $k = 0$ is 1.66 meV for CB_1, 4.75 meV for CB_2, and 5.74 meV for CB_3.

As shown in Fig. 3, for intermediate values of β within the range of $-1 < \beta < 0$, the miniband energy levels fall between the two extreme cases of β = -1 and β = 0. The case illustrated in Fig. 3 corresponds to the miniband CB_1 of the same GaAs/$Al_{0.25}Ga_{0.75}As$ superlattice as in Fig. 2. It is worthwhile to note that the shift of CB_1 to higher energies with increasing value of β is linear. This linear dependency simplifies the analysis of operator ordering effects, as it is sufficient to just consider the two extreme cases of β = -1 and β = 0.

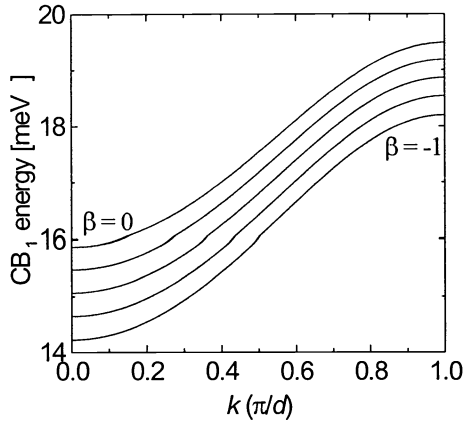

Fig. 3. Shift in the CB_1 dispersion curves associated with various choices of the KEO parameter β for a $GaAs/Al_{0.25}Ga_{0.75}As$ superlattice with d_1= 15 nm, d_2= 2.5 nm. The intermediate results between the extreme cases of $\beta = 0$ and $\beta = -1$ correspond to $\beta = -0.25$, $\beta = -0.50$, and $\beta = 0.75$, respectively.

4. SENSITIVITY OF CONDUCTION BAND ENERGY LEVELS TO OPERATOR ORDERING

In this section, we investigate the effects of operator ordering on the lowest miniband energies at $k = 0$ for $GaAs/Al_xGa_{1-x}As$ superlattices, with the purpose of determining conditions under which the effect of the choice of β is maximal. We will then use these results in Section 5 to compare our theoretical results with experimental data for structures where the effect of operator ordering is expected to be significant.

4.1. Effects of Well Thickness Variation

Since the KEO ordering is a quantum-mechanical problem related mostly to the heterointerfaces and their adjacent areas, intuitively, we can expect that placing interfaces close enough should make the results more sensitive to the choice of β. We check this hypothesis by considering several quantum-well and superlattice cases with varied well thicknesses.

As a convenient measure of the sensitivity to KEO ordering, we define a parameter ζ_{CBi} which describes the ordering-related miniband energy difference for miniband i at $k = 0$:

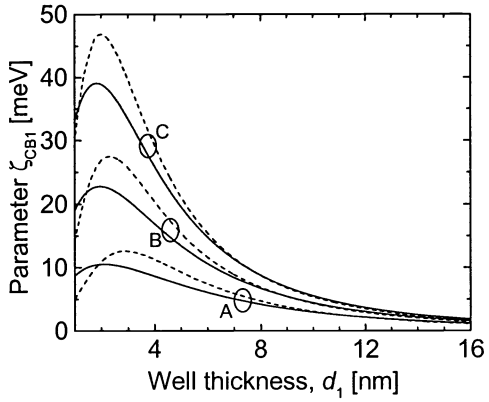

Fig. 4. Sensitivity of the fundamental transition energy shift due to KEO ordering, represented by the parameter ζ_{CB1} defined in Eq. (24), to the quantum well thickness variation in $GaAs/Al_xGa_{1-x}As$ superlattices. The cases A, B, and C correspond to 20, 30, and 40 percent of aluminum in the barrier material. The dashed curves correspond to the barrier thickness d_2= 12.5 nm, while solid curves represent the results for d_2= 2.5 nm.

$$\zeta_{CBi} = CB_i\big|_{\beta=0} - CB_i\big|_{\beta=-1} \quad . \tag{24}$$

Here, the first term on the right-hand side stands for the CB_i energy calculated assuming $\beta = 0$, while the second term is the CB_i energy calculated with $\beta = -1$.

Fig. 4 shows dependence of the shift ζ_{CB1} in the lowest miniband energy CB_1 on the well thickness d_1. Three different Al concentrations for $GaAs/Al_xGa_{1-x}As$ superlattices are considered, namely $x = 0.2$, $x = 0.3$, and $x = 0.4$. The barrier thicknesses are chosen to be $d_2 = 2.5$ nm (solid curves) and $d_2 = 12.5$ nm (dashed curves). The latter case corresponds to poor coupling between the wells, thus approaching a single-quantum-well situation. The energy shift parameter ζ_{CB1} has its peak value of more than 45 meV for a thin-well superlattice with $d_1 = 2$ nm (case C, dashed curve). It should be relatively easy to detect such a large energy difference experimentally. The smallest peak value of ζ_{CB1} is just above 10 meV (case A, solid curve). Given all possible experimental and numerical errors, such a small energy difference would be very difficult to verify. Most experimental results reported in literature are for $x = 0.3$ (case B). In this case, for thin decoupled quantum wells we can expect up to 20 meV energy shift due to different choices of the ordering parameter β.

4.2. Effects of Barrier Thickness Variation

The results of Section 4.1 indicate that the shift in the CB_1 energy at the Γ point for cases of $\beta = -1$ and $\beta = 0$ is expected to be higher for decoupled quantum wells. Fig. 5 shows the results of varying the barrier thickness for six different sets of $GaAs/Al_xGa_{1-x}As$ superlattices with well thicknesses of 2.5 and 12.5 nm and aluminum content of $x = 0.2$, $x = 0.3$, and $x = 0.4$. Our calculations show that the influence of barrier thickness on the parameter ζ_{CB1} is significant only for very thin barriers. All other calculations show negligible influence of the choice of β, which can be easily understood by recognizing that at sufficiently thick barriers the superlattice becomes equivalent to a collection of decoupled quantum wells.

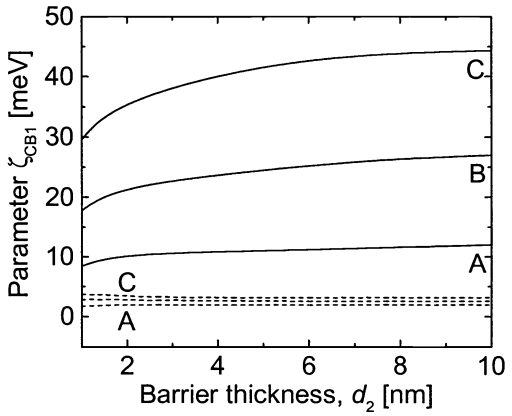

Fig. 5. Sensitivity of the lowest miniband energy shift due to KEO ordering, represented by the parameter ζ_{CB1} defined in Eq. (24), to the barrier thickness variation in $GaAs/Al_xGa_{1-x}As$ superlattices. The cases A, B, and C correspond to aluminum content of $x = 0.2$, $x = 0.3$, and x=0.4 in the barriers. The dashed curves correspond to the well thickness $d_1 = 12.5$ nm, while solid curves represent the results for $d_1 = 2.5$ nm.

4.3. Effects of Barrier Height Variation

Next, we consider the effect of barrier height on the parameter ζ_{CB1}. The barrier height is increased by changing the aluminum concentration up to $x = 0.4$. As shown in Fig. 6, we find that as the barrier height increases, the shift in the CB_1 energy for cases of $\beta = 0$ and $\beta = -1$ also increases. Our calculations show a shift in the $k = 0$ energy of 15.04 meV and 33.54 meV for cases 1 and 2, respectively. Together with the variation of quantum well thickness, the variation of aluminum content leads to the strongest ordering-related miniband energy shift.

In principle, the aluminum content could be changed up to $x = 1$, with pure AlAs in the barriers. Although it would result in a significant increase of the parameter ζ_{CB1}, the accuracy of our model for a superlattice with an indirect-bandgap material in the barrier would be questionable. For this reason, the maximum concentration of aluminum we consider in this paper is $x = 0.5$.

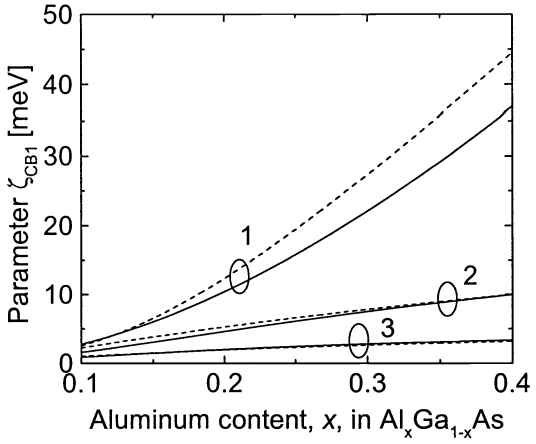

Fig. 6. Sensitivity of the lowest miniband energy shift due to KEO ordering, represented by the parameter ζ_{CB1} defined in Eq. (24), to aluminum content variation in GaAs/$Al_xGa_{1-x}As$ superlattices. The cases 1, 2, and 3 correspond to three different choices of well thickness d_1, namely 2.5, 7.5, and 12.5 nm, respectively. The dashed curves correspond to barrier thickness d_2= 12.5 nm, the solid curves represent the results for d_2= 2.5 nm.

5. EFFECT OF OPERATOR ORDERING ON TRANSITION ENERGY SHIFT

To analyze the ordering-related interband or intersubband transition energy shift, we calculate subband energy levels at $k = 0$ in conduction and valence bands, and compare predicted transition energies with experimental data.

In analogy to the definition (24) of the parameter ζ_{CBi}, we define a parameter $\zeta_{\sigma i}$ which describes the ordering-related interband transition energy difference:

$$\zeta_{\sigma i} = \left(CB_i - \sigma H_i\right)\Big|_{\beta=0} - \left(CB_i - \sigma H_i\right)\Big|_{\beta=-1} \quad , \tag{25}$$

where the symbol σ = H, L specifies whether the transition involves heavy- or light-holes.

In order to minimize the uncertainty of numerical results and inaccuracy of experimental measurements, we used the results of Section 4 in which superlattice/quantum well structures with energy levels most sensitive to operator ordering were identified. To maximize the expected $\zeta_{\sigma i}$, we considered experiments where the aluminum content in the barriers was high (30-50%), the wells were thin (4-10 nm), and the barriers were thick (up to 50 nm). Using the two extreme cases of KEO ordering ($\beta = 0$ and $\beta = -1$), we compare deviation of the experimental results from the theoretical values and find relative and absolute errors (see Table 2). Twenty different experimental cases were considered. Most of them (10 cases) are CB_1–HH_1 interband transitions. We also considered CB_1–LH_1 (4 cases), CB_2–HH_2 (2 cases), and one case for each CB_2–LH_2 and CB_3–HH_3 interband transition. In addition, we have also considered two cases of intersubband transition CB_2–CB_1. As indicated in Table 2, six transition energies were measured at room temperature, while the remaining data were measured at 2, 4, 8, and 77 K. In our calculations, the Varshni formula was used to account for bandgap variation with temperature [Vurtgaftman 2001]. The exciton binding energy was neglected for room-temperature experiments.

Fig. 7 summarizes all results for the CB_1–HH_1 and CB_1–LH_1 transitions in the most concise form. Two extreme cases of operator ordering ($\beta = 0$ and $\beta = -1$) are compared. To clarify data analysis, we arrange all calculated results in order of increasing values of the parameter $\zeta_{\sigma 1}$. For the purpose of generality, all theoretical results are normalized to zero-line (dashed curve). We plot experimental data with respect to this line. For every case, we vary

the well thickness within the range of plus-minus one monolayer. This is done is to check implications of Li and Kuhn's argument that the exact position of an abrupt interface is not well defined [Li 1994]. The resulting triangular shaded areas are shown in Fig. 7. They indicate an increase in uncertainty of the calculated transition energy caused by the ill-defined interface position correlated with an increase in the value of $\zeta_{\sigma 1}$. The experimental results are expected to fall inside these shaded regions. The most important result of Fig. 7 is a definite preference for the $\beta = -1$ ordering.

Table 2. Effects of operator ordering on comparison between theoretical and experimental transition energies in $Al_xGa_{1-x}As$ superlattices and quantum wells

Transition type	Temperature [K]	Al content	Thickness [nm]		Transition energy [eV]			Ref.*	Energy deviation** [meV]		Relative error [%]	
		(x)	Well	Barrier	$\beta = 0$	$\beta = -1$	Expt.		$\beta = 0$	$\beta = -1$	$\beta = 0$	$\beta = -1$
Interband transitions												
CB_1-HH_1	4	0.465	10	20	1.549	1.542	1.538	1	-11	-4	0.72	0.26
CB_1-HH_1	4	0.465	10	20	1.564	1.555	1.547	1	-17	-8	1.10	0.52
CB_1-HH_1	4	0.3	8	50	1.571	1.563	1.564	2	-7	1	0.45	0.09
CB_1-HH_1	300	0.5	8.5	8	1.488	1.477	1.480	3	-8	3	0.55	0.17
CB_1-LH_1	300	0.5	8.5	8	1.513	1.499	1.490	3	-23	-9	1.53	0.60
CB_1-HH_1	4	0.5	7	30	1.597	1.580	1.584	4	-12	4	0.78	0.25
CB_1-HH_1	4	0.3	6	50	1.603	1.588	1.591	2	-12	2	0.76	0.14
CB_1-HH_1	8	0.35	5.5	30	1.621	1.602	1.611	5	-10	9	0.62	0.57
CB_1-LH_1	4	0.5	7	30	1.629	1.608	1.606	4	-23	2	1.42	0.11
CB_1-LH_1	8	0.35	5.5	30	1.658	1.634	1.635	5	-23	1	1.38	0.06
CB_1-HH_1	300	0.31	4.4	5.2	1.563	1.540	1.541	6	-22	1	1.41	0.08
CB_1-HH_1	4	0.5	5	30	1.654	1.622	1.629	4	-25	6	1.56	0.39
CB_2-LH_2	300	0.5	8.5	8	1.767	1.726	1.700	3	-67	-26	3.93	1.54
CB_1-HH_1	4	0.3	4	50	1.665	1.640	1.647	2	-18	8	1.11	0.46
CB_1-LH_1	4	0.5	5	30	1.703	1.663	1.660	4	-43	3	2.58	0.21
CB_2-HH_2	300	0.5	8.5	8	1.728	1.691	1.630	3	-98	-61	6.01	3.75
CB_2-HH_2	8	0.35	5.5	30	1.886	1.863	1.837	5	-49	-27	2.71	1.45
CB_3-LH_3	300	0.5	8.5	8	2.002	1.970	1.820	3	-182	-150	10.0	8.24
Intersubband transitions												
CB_2-CB_1	77	0.3	7.5	10	0.148	0.140	0.127	7	-20	-12	16.0	9.54
CB_2-CB_1	2	0.3	26.4	19.8	0.020	0.019	0.019	8	1	0	6.88	0.88

*Reference numbers used in Table 2 are: 1 = [Ky 1992], 2 = [Martinez-Pastor 1993], 3 = [Kirchoefer 1982], 4 = [Gurioli 1993], 5 = [Galbraith 1988], 6 = [Raccah 1987], 7 = [Jogai 1992], 8 = [Bajema 1987]

**Energy deviation is defined as the difference between experimental and theoretical transition energy values

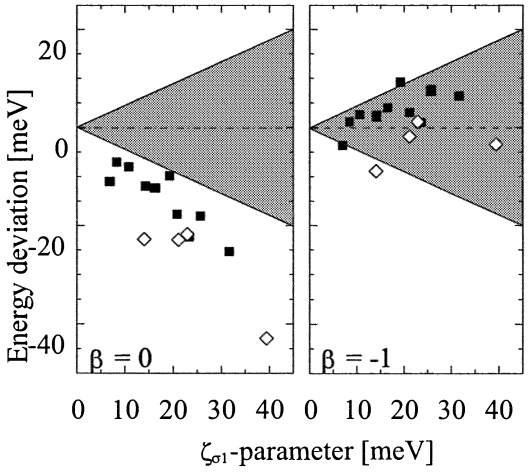

Fig. 7. Deviation of experimental results for the CB_1–HH_1 (full squares) and CB_1–LH_1 (open rhombs) transitions listed in Table 2 from their theoretical predictions (dashed line) corresponding to the two extreme choices of operator ordering. The triangular shaded areas indicate the range of uncertainty of theoretical transition energy caused by variation of quantum well thickness by plus-minus one GaAs monolayer.

6. DISCUSSION AND CONCLUSIONS

The main objective of this work was to find the correct KEO ordering for abrupt-interface heterostructures. The theoretical model was developed within the framework of the effective-mass theory. One-dimensional solutions of Schrödinger's equation for a periodic system with position-dependent mass have been considered. Using the Hamiltonian $H = -\frac{1}{2}[m(z)]^{\alpha}\nabla[m(z)]^{\beta}\nabla[m(z)]^{\alpha}+V(z)$ with $2\alpha + \beta = -1$, we have studied the effect of the parameter β on the transition energies for GaAs/$Al_xGa_{1-x}As$ superlattices and quantum wells. We found that variation of this parameter resulted in a significant shift of subband energy levels. Our results clearly indicate that $\alpha = 0$ and $\beta = -1$ is the optimal choice of ordering parameters to provide the best fit of experimental data.

We have analyzed twenty different GaAs/$Al_xGa_{1-x}As$ superlattice and quantum well systems using the transfer matrix technique. The results show that an increase in the miniband-edge energy within the conduction band is approximately linear within the entire range of $-1 \le \beta \le 0$. We investigated the effects of superlattice parameters, such as miniband index, thicknesses of both constituent materials, and barrier height (composition) effects on the ordering-related shifts of miniband-edge energy. The two extreme cases of $\beta = -1$ and $\beta = 0$ give larger energy shifts for higher miniband index, smaller well thickness, larger barrier thickness, and higher aluminum concentration in the barriers.

In order to maximize the accuracy of our test for the best choice of β, we have selected twenty sets of experimental data for structures with well thickness $4 \le d_1 \le 10$ nm, barrier thickness $5 \le d_2 \le 50$ nm, and aluminum concentration $0.3 \le x \le 0.5$. We define the parameter $\zeta_{\sigma i}$ in Eq. (25) as a measure of ordering-related transition energy shift. For low temperature results, excitonic correction to transition energy was included in the calculations. Theoretical results indicate that the values of parameter $\zeta_{\sigma i}$ for some experimental cases can be as high as 40 meV (Fig. 7). In principle, it is possible to increase the energy difference even further, for example, by considering ultrathin quantum wells with very high aluminum concentration ($x > 0.5$) in the barriers.

Uncertainty in the exact interface position was also taken into account. Deviations from the average well thickness by plus-minus one monolayer were considered. The effect of this variation is proportional to the ordering-related transition energy shift $\zeta_{\sigma i}$ and can amount to more than 40 meV in magnitude (Fig. 7). Nevertheless, $\beta = -1$ gives much better results even when this uncertainty is considered. The results obtained with $\beta = 0$ all fall outside the range of one monolayer uncertainty

For all twenty different experimental cases, the choice of $\beta = -1$ consistently gives a much better fit than $\beta = 0$. We therefore conclude that the correct choice for the kinetic energy operator ordering for abrupt heterointerfaces, consistent with experimental data, is $\beta = -1$.

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