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GaAs quantum structures: Comparison between direct pseudopotential and single-band truncated-crystal calculations

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A single-band approach for semiconductor clusters which accounts for the nonparabolicity of the energy bands was recently used by Rama Krishna and Friesner @M.V. Rama Krishna and R.A. Friesner, Phys. Rev. Lett. **67**, 629 ~1991!#. We compare the results of this method ~denoted here as single-band truncated-crystal, or SBTC, approximation! with a direct pseudopotential band-structure calculation for free-standing hydrogen-passivated GaAs quantum films, wires, and dots. The direct pseudopotential calculation, which includes coupling between all bands, shows that isolated GaAs quantum films, wires, and dots have an indirect band gap for thicknesses below 16, 28, and at least 30 Å ~8, 14, and at least 15 ML!, respectively; beyond these critical dimensions the transition becomes direct. A comparison of the SBTC approximation with the direct pseudopotential calculation shows that ~i! the confinement energy of the valence-band maximum is overestimated by the SBTC method, because the zero-confinement character of this state is neglected;

tion, mainly because of the assumption of infinite potential barriers at the boundaries; ~iii! the confinement energy of the *X*-derived conduction state ~indirect gap! is severely *underestimated* by SBTC; ~iv! while the SBTC approximation predicts quantum deconfinement for the direct gap of thin GaAs quantum wires, such effect is not present in the direct pseudopotential calculation, and is therefore an artifact of the SBTC approximation.

II. METHODS

A. Direct diagonalization

In the direct diagonalization ~DD! approach we solve for the single-particle Schrödinger equation ~in atomic units!:

$$\left(-1/2\nabla^2+V_{\mathrm{QS}}(\mathbf{r})\right)\psi_i(\mathbf{r})=\varepsilon_i\psi_i(\mathbf{r}), \tag{1}$$

where $V_{\mathrm{QS}}(\mathbf{r})$ is the total pseudopotential of the quantum structure. V_{QS} is described here by the sum of screened atomic pseudopotentials centered at the atomic positions $\mathbf{R}_n$:

$$V_{\mathrm{QS}}(\mathbf{r})=\sum_n v_n(\mathbf{r}-\mathbf{R}_n). \tag{2}$$

For Ga and As atoms we use the nonrelativistic atomic pseudopotentials developed by Mäder and Zunger.[11] Using a

B. Single-band truncated-crystal approximation

The same Ga and As atomic pseudopotentials used in the direct minimization procedure are applied to the calculation of the bulk band structure, in order to extract the quantum

standing GaAs quantum film oriented in the ~1$\bar{1}$0! plane. The set of **k*** points satisfying the particle-in-a-box quantization rule is given by

$$\mathbf{k}_l^* = p/(\sqrt{2}L)\ \sim l, -l, 0!, \qquad \sim 5!$$

where L is the film thickness and l is a nonzero integer. According to the SBTC method, the film eigenvalues e_i^{film} are approximated by the bulk band energies calculated at these **k*** points.

The band-edge energies at the zone center of the two-dimensional Brillouin zone, calculated with the SBTC approximation and with the direct pseudopotential approach, are compared in Fig. 3~a! as a function of the film thickness L ~note that in the ^110& directions 1 GaAs ML . 2.0 Å!. In the ''exact'' pseudopotential calculation the VBM of the quantum film is a zero-confinement state,[8,14] whose energy is almost independent of the film thickness L. The conduction-band minimum originates from the X_{1c} valley for $L<8$ ML's, while it becomes a Γ_{1c} derived state for $L>8$ ML's; the $X \rightarrow \Gamma$ crossover is evident in Fig. 3~a! from the change of slope of the energy vs thickness curve. The SBTC approximation gives a good overall description of the near-edge states of GaAs quantum films, but fails to describe the zero-confinement nature of the VBM, and slightly overestimates the CBM confinement energy. Figure 3~b! shows the band gap as a function of L. The SBTC band gap is overestimated typically by ; 0.2 eV for $L<15$ ML's.

The X-derived and Γ-derived conduction-band energies are shown in more detail in Fig. 4 as a function of the film thickness L. We see that the SBTC error in energy is larger for the Γ-derived state than for the X-derived state. The $X \rightarrow \Gamma$ crossover is clearly visible in this figure; it is predicted to occur at L ; 8 ML's by the direct diagonalization method (L ; 13 ML's for a GaAs/AlAs quantum well[15]!, and at L ; 10 ML's by the SBTC approximation ~see also Table I!. Note that for 2D quantum films ~both in the ''exact'' pseudopotential results and in the SBTC approximation! there is no quantum deconfinement in either the Γ-derived state or the X-derived state: the energy of these states increases monotonically as the film thickness decreases.

B. Quantum wires

We now consider GaAs quantum wires with square cross section; the surface planes are oriented in the ~110! and ~1$\bar{1}$0! directions, and the wires are periodic in the @001# direction. The **k*** points satisfying the particle-in-a-box quantization rules are

amplitude is plotted in the ~$1\overline{1}0$! $\times$ ~110! plane for L =

the SBTC method picks up only *one* ~or a few, if degenerate! $\mathbf{k}^*$ point~s! in the Brillouin zone as the CBM wave vector~s!; in general, this approximation is insufficient to satisfy zero boundary conditions in quantum wires and dots. This error can be partially corrected by introducing an ''extended'' single-band truncated-crystal ~E-SBTC! approximation, where each conduction-band valley is treated independently from the others. This method was first discussed by Zhang, Yeh and Zunger[8] in the case of Si quantum films. Denoting by $\mathbf{k}_0$ the wave vector of a particular valley[16] ~e.g., Γ_{1c}, X^x_{1c}, X^y_{1c}, X^z_{1c}, etc.!, in the E-SBTC approximation we ~i! apply the particle-in-a-box quantization rules to the $(\mathbf{k}-\mathbf{k}_0)$ wave vector; ~ii! calculate the quantized wave vectors $\mathbf{k}^*$ corresponding to the lowest allowed particle-in-a-box indexes ~e.g., $\mathbf{k}^*=\mathbf{k}_0+p(\pm 1/L_1,\pm 1/L_2,\pm 1/L_3)$ in the case of a rectangular quantum box#; ~iii! average over the bulk band energies calculated at these $\mathbf{k}^*$ points ~which are, in general, nondegenerate!. The results of this approach, including the X_{1c}

$$\mathbf{k}^* = \mathbf{k}_0 \pm \hbar/\sqrt{2}\,(1,-1,0), \tag{8}$$

where $\hbar$ is the smallest nonvanishing solution of the implicit equation

$$\tan\left(\hbar \frac{L}{2}\right) = \frac{(2m_0^* V_0 - \hbar^2)^{1/2}}{\hbar}. \tag{9}$$

Here m