Interface and high energy band effects in quantum dots: beyond 8-band $\mathbf{k}\cdot\mathbf{p}$ Hamiltonian

Stanko Tomić[a)] and Nenad Vukmirović[b)]
[a)] *Joule Physics Laboratory, School of Computing, Science and Engineering, University of Salford, UK*
[b)] *Scientific Computing Laboratory, Institute of Physics Belgrade, University of Belgrade, Serbia*

***Abstract*—The role of interfaces and higher bands on the electronic structure of embedded semiconductor quantum dots (QDs) was investigated. The term in the multiband k·p Hamiltonian that captures the effect of interfaces was derived starting from the microscopic theory. It was shown, analytically and numerically, that, with such a term included, the right symmetry of the QD system can be captured. The inclusion of additional higher bands beyond the ones from the standard eight-band model also leads to the reduction of symmetry from an artificially high one to the true atomistic symmetry of the system. These results prove that the multiband k·p Hamiltonians are fully capable of describing the correct symmetry of a QD.**

I. Introduction

Typical self-assembled QDs have millions of atoms and therefore the methods based on *ab initio* density functional theory, the workhorse of modern electronic structure calculations, are far beyond the reach of present day computational capabilities. All the methods developed so far are therefore to a lesser or larger extent semi-empirical. [1], [2], [3] In the multiband envelope function methods (including the $\mathbf{k}\cdot\mathbf{p}$ method) the system Hamiltonian is represented through a matrix of operators that act upon a vector of envelope functions of several bands. The multiband $\mathbf{k}\cdot\mathbf{p}$ Hamiltonians [4], [5], [6], [7], [8] are capable of reproducing the bulk bandstructure more accurately than the standard 8-band Hamiltonian. The effect of interface band mixing[9], [10], [11], [12] has also so far been analyzed only for a single interface or a quantum well structure. The goal of this work is to explore the effects of higher bands and interfaces on the electronic structure of QDs. The most widely used form of the multiband Hamiltonians for the description of III-V material nanostructures with zincblende crystal structure is the 8-band $\mathbf{k}\cdot\mathbf{p}$ Hamiltonian. This Hamiltonian indeed yields a higher symmetry of the nanostructure than the true atomistic one. There is however a widespread belief [13], [14] that the mentioned deficiency is a general feature of multiband $\mathbf{k}\cdot\mathbf{p}$ Hamiltonians and that these cannot be used if one needs to capture the true symmetry of the nanostructure. If this were true, this would be indeed a serious issue. For example, models with high symmetry lead to degeneracies of eigenstates that would be otherwise split and as a consequence lead to inaccuracies in the prediction of polarization dependence of interaction with electromagnetic radiation or the distribution of the excitonic state population and their dynamics.

We find no reason that the mentioned shortcomings of the 8-band $\mathbf{k}\cdot\mathbf{p}$ Hamiltonian would manifest also in multiband $\mathbf{k}\cdot\mathbf{p}$ Hamiltonians in general. In this work, we therefore analyze the $\mathbf{k}\cdot\mathbf{p}$ Hamiltonians beyond the standard 8-band one, which either include the effect of higher energy bands or include the effect of interface induced band mixing. For the latter, we derive the analytical form of the appropriate Hamiltonian for all interfaces present in the model QD structure and use it in the calculation to understand its role both qualitatively and quantitatively. We show that beyond 8-band Hamiltonians are capable of capturing the correct atomistic symmetry of QDs and quantify the amount of symmetry breaking induced by each of them.

II. Theoretical method

As a model system, we consider a QD in the shape of a square-based pyramid, with base to height ratio $b/h = 2$. The coordinate system is chosen so that vertices of the pyramid are at the points $(b/2, -b/2, 0)$, $(b/2, b/2, 0)$, $(-b/2, b/2, 0)$, $(-b/2, -b/2, 0)$ and $(0, 0, h)$.

The $\mathbf{k}\cdot\mathbf{p}$ Hamiltonians discussed in this paper have a general form:

$$H_{\mathbf{k}\cdot\mathbf{p}} = H_{\mathbf{k}} + H_{\mathrm{so}} + H_{\epsilon} + H_{\mathrm{pz}} + H_{\mathrm{if}} \tag{1}$$

where $H_{\mathbf{k}}$ is the kinetic part, H_{so} is the part arising from spin-orbit interaction, H_{ϵ} is the strain part, H_{pz} is the piezoelectric part of the Hamiltonian that depends on strain, and H_{if} describes the interface induced band-mixing contribution to the Hamiltonian $H_{\mathbf{k}\cdot\mathbf{p}}$. To identify the origins and to quantify the amount of symmetry breaking, we have analyzed our model system using several different Hamiltonians that include only certain terms from Eq. (1), each with a different level of sophistication:

(a) The 8-band $\mathbf{k}\cdot\mathbf{p}$ Hamiltonian consisting of the kinetic part only.
(b) The 8-band $\mathbf{k}\cdot\mathbf{p}$ Hamiltonian consisting of the kinetic part with the spin-orbit interaction taken into account.
(c) The 8-band $\mathbf{k}\cdot\mathbf{p}$ Hamiltonian consisting of the kinetic part with the interface band-mixing effects taken into account.
(d) The standard 8-band $\mathbf{k}\cdot\mathbf{p}$ Hamiltonian consisting of the kinetic part with the spin-orbit interaction and strain, as well as the strain-induced piezoelectric potential.
(e) The 8-band $\mathbf{k}\cdot\mathbf{p}$ Hamiltonian consisting of the kinetic part with the spin-orbit interaction and strain, as well as the strain-induced piezoelectric potential and the interface Hamiltonian.
(f) The 14-band $\mathbf{k}\cdot\mathbf{p}$ Hamiltonian consisting of the kinetic part only.

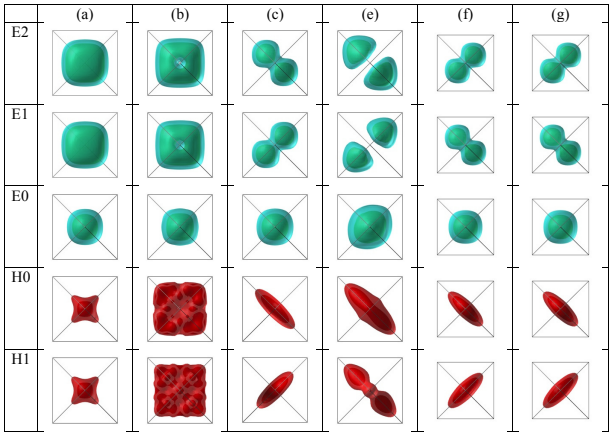

Fig. 1: (Color online) The wavefunctions squared for top two hole states and bottom three electron states for a square-based pyramidal InAs/GaAs QD with base width $b = 100$ Å, and base to height ratio $b/h = 2$ calculated using different models. The letters (a)-(c), (e)-(g) specify the model defined in the main text. The isosurfaces are plotted at 25% (transparent) and 75% of the maximal charge density.

(g) The 16-band $\mathbf{k} \cdot \mathbf{p}$ Hamiltonian consisting of the kinetic part only.
(h) The 14-band $\mathbf{k} \cdot \mathbf{p}$ Hamiltonian consisting of the kinetic part with the spin orbit-interaction and strain.
(i) The 16-band $\mathbf{k} \cdot \mathbf{p}$ Hamiltonian consisting of the kinetic part with the spin orbit-interaction and strain.

In the derivation of the interface-induced band mixing terms in the 8-band **k·p** Hamiltonian applied to our model QD system, we follow the approach of Foreman outlined in Ref. [12]. The envelope function Hamiltonian at a point $\mathbf{R}$ in space (in the absence of strain and spin-orbit interaction) is equal to: [12]

$$H_{mn}(\mathbf{R}) = \frac{\hbar^2 k^2}{2m_0}\delta_{mn} + \frac{\hbar}{m_0}\mathbf{k}\cdot\mathbf{p}_{mn} + [u_m|H_{\mathrm{a}}|u_n]_{\mathbf{R}}. \quad (2)$$

In this equation, u_m and u_n are the periodic Bloch functions of band m and n of a bulk reference crystal, $H_{\mathrm{a}} = p^2/2m_0 + V(\mathbf{r})$ is the microscopic Hamiltonian that we model using the local empirical pseudopotentials, the square brackets denote the averaging over a unit cell centered on $\mathbf{R}$, $\mathbf{k} = -i\nabla$ and $\mathbf{p}_{mn} = [u_m|\mathbf{p}|u_n]$, where $\mathbf{p}$ is the momentum operator.

The kppw code,[15], [16] appropriately extended to treat the multiband Hamiltonians and the effect of interfaces, was used for all electronic structure calculations presented.

III. Results and Discussion

In contrast to a popular belief that multiband envelope function **k·p** Hamiltonians cannot capture the right symmetry of QDs, we showed here the opposite. The symmetry group of 8-band **k·p** Hamiltonian applied to a pyramidal square-based QD is the C_{4v} group. We showed that the inclusion of interface band mixing effects leads to the reduction of symmetry from an artificial C_{4v}, to the correct C_{2v} one. Our analytical derivations, demonstrate that interface effects lead to the reduction of symmetry from C_{4v} to C_{2v} since commutator $[D(R_\varphi), H_{\mathrm{if}}] = 0$ only if $\varphi = n\pi$, where D is the group representation generators. The main manifestation of interface effects are the energy level splittings between (e_1, e_2), (h_0, h_1), and (h_4, h_5) states of the order of 1–3 meV in InAs/GaAs material system. The splittings decrease as the dot size and consequently the volume to surface ratio increase. The inclusion of the additional bands beyond the standard 8 bands also leads to symmetry reduction to C_{2v}, with splittings which are however weaker than the ones due to interfaces. We have found analytically that the 14 band **k·p** Hamiltonian commutes with the operators of the C_{2v} group representation, i.e., $[D(R_\varphi), H_{14\mathbf{k}\cdot\mathbf{p}}] = 0$ only for $\varphi = n\pi$. We have found indeed that the 14-band **k·p** Hamitonian is the lowest order multiband Hamiltonian whose kinetic part has the correct C_{2v} atomistic symmetry. This symmetry reduction originates from the coupling between the top of the valence (Γ_{5v}) and the second conduction (Γ_{5c}) band. The observed splittings are comparable to the ones that originate from spin-orbit coupling (these do not reduce the symmetry) and are much smaller than the ones from piezoelectric effect in strained systems. Our work provides a very important conceptual message – with appropriate treatment of relevant effects, the multiband envelope function Hamiltonians are fully capable of capturing the right atomistic symmetry of QD structures.

References

[1] L.-W. Wang and A. Zunger, "Linear combination of bulk bands method for large-scale electronic structure calculations on strained nanostructures," *Phys. Rev. B*, vol. 59, pp. 15 806–15 818, 1999.
[2] S. Lee, J. Kim, L. Jönsson, J. W. Wilkins, G. W. Bryant, and G. Klimeck, "Many-body levels of optically excited and multiply charged InAs nanocrystals modeled by semiempirical tight binding," *Phys. Rev. B*, vol. 66, p. 235307, 2002.
[3] O. Stier, M. Grundmann, and D. Bimberg, "Electronic and optical properties of strained quantum dots modeled by 8-band $k \cdot p$ theory," *Phys. Rev. B*, vol. 59, pp. 5688–5701, 1999.
[4] P. Pfeffer and W. Zawadzki, "Five-level k·p model for the conduction and valence bands of GaAs and InP," *Phys. Rev. B*, vol. 53, pp. 12 813–12 828, 1996.
[5] M. Cardona, N. E. Christensen, and G. Fasol, "Relativistic band structure and spin-orbit splitting of zinc-blende-type semiconductors," *Phys. Rev. B*, vol. 38, pp. 1806–1827, 1988.
[6] M. Cardona and F. H. Pollak, "Energy-band structure of germanium and silicon: The k·p method," *Phys. Rev.*, vol. 142, pp. 530–543, 1966.
[7] D. Rideau, M. Feraille, L. Ciampolini, M. Minondo, C. Tavernier, H. Jaouen, and A. Ghetti, "Strained Si, Ge, and $Si_{1-x}Ge_x$ alloys modeled with a first-principles-optimized full-zone $k \cdot p$ method," *Phys. Rev. B*, vol. 74, p. 195208, 2006.
[8] S. Richard, F. Aniel, and G. Fishman, "Energy-band structure of Ge, Si, and GaAs: A thirty-band k·p method," *Phys. Rev. B*, vol. 70, p. 235204, 2004.
[9] U. Rössler and J. Kainz, "Microscopic interface asymmetry and spin-splitting of electron subbands in semiconductor quantum structures," *Solid State Communications*, vol. 121, pp. 313 – 316, 2002.
[10] J.-B. Xia, "Γ-X mixing effect in GaAs/AlAs superlattices and hetero-junctions," *Phys. Rev. B*, vol. 41, no. 5, pp. 3117–3122, Feb 1990.
[11] P. C. Klipstein, "Operator ordering and interface-band mixing in the kane-like hamiltonian of lattice-matched semiconductor superlattices with abrupt interfaces," *Phys. Rev. B*, vol. 81, p. 235314, 2010.
[12] B. A. Foreman, "Analytical envelope-function theory of interface band mixing," *Phys. Rev. Lett.*, vol. 81, no. 2, pp. 425–428, 1998.
[13] A. Zunger, "On the farsightedness (hyperopia) of the standard k·p model," *phys. stat. solidi (a)*, vol. 190, pp. 467–475, 2002.
[14] G. Bester and A. Zunger, "Cylindrically shaped zinc-blende semiconductor quantum dots do not have cylindrical symmetry: Atomistic symmetry, atomic relaxation, and piezoelectric effects," *Phys. Rev. B*, vol. 71, 2005.
[15] S. Tomić, A. G. Sunderland, and I. J. Bush, "Parallel multi-band $k \cdot p$ code for electronic structure of zinc blend semiconductor quantum dots," *J. Mater. Chem.*, vol. 16, pp. 1963–1972, 2006.
[16] S. Tomić and N. Vukmirović, "Symmetry reduction in multiband Hamiltonians for semiconductor quantum dots: The role of interfaces and higher energy bands," submitted to *Phys. Rev. B* .