Application of a Total-Angular-Momentum Basis to Quantum-Dot Band Structure

Kerry J. Vahala and Peter C. Sercel

Department of Applied Physics, Mail Stop 128-95, California Institute of Technology, Pasadena, California 91125

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Multiband envelope theory is reformulated in terms of a basis of total envelope and zone-center Bloch angular momentum. The $\mathbf{k} \cdot \mathbf{p}$ operator is scalar with respect to the total-angular-momentum operator and hence is block diagonal in its eigenbasis. The projections of the Hamiltonian into the $F = \frac{1}{2}$ and the $F = \frac{3}{2}$ subspaces of total angular momentum are derived and quantum-dot band structure is calculated in GaAs(AlGaAs) and InAs(GaSb) systems as an example application of the formalism.

Multiband envelope theory has emerged as a powerful extension of the $\mathbf{k} \cdot \mathbf{p}$ formalism for calculation of band structure in semiconductor heterostructures.[1] The underlying motivation of this formalism is based on the bulk $\mathbf{k} \cdot \mathbf{p}$ Hamiltonian. In short, this Hamiltonian can be viewed as originating from an envelope wave equation. For the special case of flat energy bands (i.e., bulk semiconductor material) the envelopes are plane-wave functions that maintain the lattice symmetry. In general, however, the energy bands will have spatial dependences as, for example, results in a superlattice or quantum-well structure. For these cases, solution of the envelope wave equation will yield new envelope functions that do not, in general, preserve the translational or the rotational symmetry of the underlying lattice. Interestingly, however, the lattice translational symmetry does influence the preferred coordinate system, Cartesian coordinates, used in formulating the envelope wave equation.

Until recently, this observation has had little consequence in heterostructure band-structure calculations, since most of the semiconductor heterostructures studied are inherently one dimensional, thus lending themselves to a solution in a Cartesian coordinate system. Several classes of heterostructures with two- and three-dimensional spatial dependences are of increasing interest, however. Quantum wires and quantum dots are examples. These structures can be viewed as two- and three-dimensional extensions of the conventional quantum well. Their band structure and the nature of their electronic state space is of importance in potential device applications.[2,3] Separable, finite, two- and three-dimensional potentials representing realistic quantum dots and quantum wires cannot be constructed in Cartesian coordinates, however. Therefore, to compute band structure in hypothetical structures, either an infinite potential in Cartesian coordinates can be applied or the envelope wave equation must be solved in cylindrical or spherical coordinates with a corresponding separable finite potential. The former approach is unphysical for small structures and the latter approach is tedious, involving a solution of coupled partial differential equations.

In this Letter we reformulate multiband envelope theory using a basis which takes advantage of the symmetry imposed by the heterostructure and which simultaneously simplifies the nature of the $\mathbf{k} \cdot \mathbf{p}$ interaction operator. The reformulation is based on states of total zone-center Bloch ($\mathbf{j}$) and envelope angular momentum ($\mathbf{l}$), i.e., $\mathbf{F} = \mathbf{j} + \mathbf{l}$. We note that a total-angular-momentum basis was first used in a number of theoretical investigations of the Coulomb impurity problem.[4-6] Although the basis used here is similar, our motivation is new and our results are more general. Specifically, our use of these states is based on our observation, described further below, that the $\mathbf{k} \cdot \mathbf{p}$ interaction operator is block diagonal in the basis of total angular momentum. As a direct result, our formalism provides a simple description of band structure in centrosymmetric heterostructure potentials, but is general enough to rigorously account for effects such as band coupling between conduction and valence states in narrow-band-gap systems. The eight-band $\Gamma_6, \Gamma_8, \Gamma_7$ system (conduction, heavy-hole, light-hole, and split-off bands) will be discussed as an example of the formalism and band structure in spherical GaAs(AlGaAs) and InAs(GaSb) quantum dots will be worked out.

Beyond the convenience and advantages of a separable potential in three dimensions, it is not clear that a total-angular-momentum basis will necessarily simplify the solution of centrosymmetric heterostructure band structure. The key simplification results from consideration of the $\mathbf{k} \cdot \mathbf{p}$ interaction. In the envelope approximation this operator is the vector inner product between envelope momentum (i.e., $\hbar \mathbf{k}$) and a momentum operator which effectively operates only on the crystal periodic part of the Bloch wave function. It is an essential feature of the envelope approximation that these momenta are conjugate to independent position coordinates (one in the periodic Bloch space and the other in the slowly varying envelope space). As such, the momenta are independent observables in the approximation, as are the angular momenta associated with the respective spaces. The inner product $\mathbf{k} \cdot \mathbf{p}$ is hence invariant under rotations generated by the total-angular-momentum operator of

the combined spaces; i.e., the $\mathbf{k} \cdot \mathbf{p}$ operator is scalar with respect to total angular momentum and block diagonal in its eigenstate representation. As discussed below, this block-diagonal simplification does not, in general, hold for the remainder of the Hamiltonian because the crystal symmetry is lower than that of the full rotation group. We will show, however, that certain important projections of the full Hamiltonian are block diagonal in the total-angular-momentum representation, decomposing as follows:

$$\mathbf{H} = \sum_{F,F_z,\pi} \mathbf{H}^{\pi}_{F,F_z} , \tag{1}$$

where π is subspace parity. To illustrate this point we will consider the eight-band $\Gamma_6, \Gamma_7, \Gamma_8$ system and compute projections of $\mathbf{H}$ into the $F = \frac{1}{2}$ and $F = \frac{3}{2}$ subspaces. In what follows, we will denote the total-angular-momentum quantum numbers as F and F_z, the zone-center Bloch total-angular-momentum quantum numbers as j and j_z, the zone-center Bloch orbital-angular-momentum quantum number as l_B, and the envelope-angular-momentum quantum numbers as l and l_z.

When the conventional multiband envelope formalism is applied in the $\Gamma_6, \Gamma_7, \Gamma_8$ system, the Hamiltonian is constructed using the basis set $\{|j,j_z,l_B\rangle |\mathbf{k}\rangle\}$. These states are a product ket consisting of a zone-center Bloch ket $|j,j_z,l_B\rangle$ and an envelope ket $|\mathbf{k}\rangle$ representing a plane-wave state with wave vector $\mathbf{k}$. In the present formalism, two modifications are made to the basis. First, the Cartesian plane-wave kets $|\mathbf{k}\rangle$ are replaced with spherical plane waves in anticipation of the centrosymmetric heterostructure potential. These spherical waves, $|k,l, l_z\rangle$ have the following projection into spherical coordinates:

$$\langle r,\theta,\phi | k,l,l_z \rangle = (2/\pi)^{1/2} i^l h_l(kr) Y_l^{l_z}(\theta,\phi) , \tag{2}$$

where $h_l = j_l + i n_l$ is a spherical Hankel function and $Y_l^{l_z}$ is a spherical harmonic. Second, appropriate Clebsch-Gordan weightings of the resulting Bloch-envelope product kets are taken to form the total-angular-momentum basis kets as follows:

$$|k,F,F_z,l_B,j,l\rangle = \sum_{j_z + l_z = F_z} C^{F,F_z}_{j_z,l_z} |j,j_z,l_B\rangle |k,l,l_z\rangle , \tag{3}$$

where $|k,F,F_z,l_B,j,l\rangle$ is a total-angular-momentum ket

and $\{C^{F,F_z}_{j_z,l_z}\}$ are the Clebsch-Gordan coefficients for the space under consideration. It is important to note that the zone-center total-angular-momentum states (i.e., $|0,F,F_z,l_B,j,l\rangle$) will be energy eigenstates only when the $2j+1$ states $\{|j,j_z,l_B\rangle\}$ appearing in the summation are degenerate. In such cases (the energy bands in the $\Gamma_6, \Gamma_7, \Gamma_8$ system are an example), the corresponding projection of $\mathbf{H}$ is as given by Eq. (1). In general, however, the crystal field will split the degeneracy of the $2j+1$ states $\{|j,j_z,l_B\rangle\}$ and the zone-center total-angular-momentum states corresponding to particular quantum numbers (l_B,j,l) will not be energy eigenstates. In the present example the effect of such bands can be included by introduction of appropriate Luttinger coupling terms in the system Hamiltonian.[1] With Luttinger coupling terms, the Hamiltonian is no longer block diagonal as given in Eq. (1). It is important to note, however, that the spherical approximation (i.e., Luttinger terms γ_2 and γ_3 are set equal[4-6]) returns the block-diagonal form for $\mathbf{H}$. In the language of group theory, the states $\{|0,F,F_z,l_B,j,l\rangle\}$ form a basis for representations of the cubic double group which are, in general, reducible. In the approximation $\gamma_2 = \gamma_3$ the symmetry of the Hamiltonian is artificially raised to that of the full rotation group so that these representations become irreducible.

Now consider the $F = \frac{1}{2}$ subspace. It is constructed from spherical envelope states having integer-valued angular-momentum quantum numbers, and from zone-center Bloch states having angular-momentum quantum numbers $j = \frac{1}{2}$ (Γ_6), $j = \frac{3}{2}$ (Γ_8), and $j = \frac{1}{2}$ (Γ_7). Using standard rules or a table of Clebsch-Gordan coefficients, it is straightforward to show that the complete $F = \frac{1}{2}$ subspace is twelve dimensional, comprising two six-dimensional subspaces corresponding to $F_z = \pm \frac{1}{2}$. $\mathbf{H}$ is block diagonal in this subspace, comprising two six-dimensional projections, $\mathbf{H}_{1/2, \pm 1/2}$. These matrices further reduce to two 3×3 projections owing to parity conservation. (This follows immediately from the selection rule $l'_B = l_B \pm 1$ and $l' = l \pm 1$, which is easily verified for the $\mathbf{k} \cdot \mathbf{p}$ operator, and the fact that the total-angular-momentum eigenstates are states of definite parity.) These matrices have the identical forms given below. The corresponding j and l quantum numbers and the band index associated with the basis vectors are shown explicitly on the columns of the matrix (i.e., $|j,l\rangle_{c,c'}$). In addition, $\pi = 0$ (1) indicates even (odd) parity:

$$
\mathbf{H}^{\pi}_{1/2, \pm 1/2} =
\begin{array}{ccc}
|\frac{1}{2},\pi\rangle_c & |\frac{3}{2},\pi+1\rangle_{c'} & |\frac{1}{2},|\pi-1|\rangle_{c'} \\
\end{array}
\begin{pmatrix}
E_c + \hbar^2 k^2/2m & (\frac{2}{3})^{1/2}\beta & (\frac{1}{3})^{1/2}\beta \\
(\frac{2}{3})^{1/2}\beta^* & E_v - (\gamma_1 + 2\gamma_2)k^2/2 & -\sqrt{2}\gamma_2 k^2 \\
(\frac{1}{3})^{1/2}\beta^* & -\sqrt{2}\gamma_2 k^2 & E_v - \Delta - \gamma_1 k^2/2
\end{pmatrix} , \tag{4}
$$

where modified Luttinger terms[7] have been included by unitary transformation of the conventional Luttinger Hamiltonian. In this expression, $\beta = -ipk$, where p is the Kane matrix element.[8] In addition, E_c, E_v, and $E_v - \Delta$ denote the

conduction-band, heavy-hole (light-hole) band, and split-off band energies at the zone center, respectively. m is the free-electron mass.

The eigenvalues of the Hamiltonian in Eq. (4) are identical to three of four eigenvalues found for this system using the normal $\mathbf{k} \cdot \mathbf{p}$ formalism. They give the bulk dispersion relations associated with the conduction band, the light-hole band, and the split-off band. As a result of the preceding analysis, these eigenvalues are fourfold degenerate in the $F = \frac{1}{2}$ subspace. The fourth, "missing," bulk eigenvalue, the heavy-hole band, does not emerge in

this space. The first signs of the heavy-hole band appear in the $F = \frac{3}{2}$ subspace. Proceeding as before, it can be shown that this space is spanned by 32 vectors. These vectors break down into four eight-dimensional subspaces corresponding to $F_z = \frac{3}{2}, \frac{1}{2}, -\frac{1}{2}, -\frac{3}{2}$. The Hamiltonian is block diagonal with respect to these subspaces and its projection further reduces owing to parity conservation. Consequently, eigenenergies are eightfold degenerate in this subspace. The odd-parity 4×4 projections of the Hamiltonian into the $F = \frac{3}{2}$ subspace appear below:

$$\mathbf{H}_{3/2, F_z}^{F=\frac{1}{2}} = \begin{bmatrix} \quad | \frac{1}{2}, 1 \rangle_c & \quad | \mathbf{LH} \rangle & \quad | \mathbf{HH} \rangle & \quad | \frac{1}{2}, 2 \rangle_c \\[4pt] E_c + \hbar^2 k^2/2m & (\frac{2}{3})^{1/2}\beta & 0 & (\frac{1}{3})^{1/2}\beta \\[4pt] (\frac{2}{3})^{1/2}\beta^* & E_v - (\gamma_1 + 2\gamma_2)k^2/2 & 0 & \sqrt{2}\gamma_2 k^2 \\[4pt] 0 & 0 & E_v - (\gamma_1 - 2\gamma_2)k^2/2 & 0 \\[4pt] (\frac{1}{3})^{1/2}\beta^* & \sqrt{2}\gamma_2 k^2 & 0 & E_v - \Delta - \gamma_1 k^2/2 \end{bmatrix}. \tag{5}$$

Here, LH and HH denote light hole and heavy hole, respectively, F_z assumes the values $\frac{3}{2}, \frac{1}{2}, -\frac{1}{2}, -\frac{3}{2}$, and, subsequent to computing the projections, a unitary transformation has been applied on the inner 2×2 system with resulting basis-vector definitions $|\mathbf{LH}\rangle = (| \frac{3}{2}, 0\rangle_v - | \frac{3}{2}, 2\rangle_v)/\sqrt{2}$ and $|\mathbf{HH}\rangle = (| \frac{3}{2}, 0\rangle_v + | \frac{3}{2}, 2\rangle_v)/\sqrt{2}$. Equation (5) contains a 3×3 matrix which yields bulk eigenvalues identical to those found with the $F = \frac{1}{2}$ Hamiltonian projections (i.e., conduction, light-hole, and split-off bands). The isolated 1×1 system with Eq. (5) gives the dispersion relation characteristic of the heavy-hole band.

Calculation of quantum-dot band structure using this formalism is, with some minor exceptions, identical to standard heterostructure envelope calculations. Eigenvectors of the Hamiltonian are computed interior and exterior to the quantum dot. Boundary conditions are applied to arrive at a relation between spherical wave numbers. This condition is combined with the known energy dispersion relations to determine the eigenvalues. The only procedural differences are first, the use of spherical Hankel waves rather than plane waves, and second, the need in the present case for a boundary condition at the origin. The latter is the requirement that the wave function be regular at the origin.

As a first example, we will compute the $F = \frac{1}{2}$ subspace ground-state eigenenergy of a quantum dot, including only the conduction and light-hole bands (i.e., neglecting coupling to the split-off band). Results are compared with those from a one-band effective-mass calculation. In Fig. 1 (refer to left vertical axis) we present the calculated confinement energy of the quantum dot relative to the confinement energy predicted using the one-band effective-mass model. This energy difference is plotted as a function of quantum-dot radius. Two material systems are considered: a large-band-gap system

[GaAs(Al$_{0.3}$Ga$_{0.7}$As)] in which agreement between the one-band and multiband models is expected, and a narrow-band-gap type-II system [InAs(GaSb)] in which multiband effects in the quantum dot should be important. This, in fact, is the first calculation of band structure in a narrow-band-gap quantum dot. The states in these systems are twofold degenerate. Band energy constants for GaAs(AlGaAs) are taken from Ref. 9, and for InAs(GaSb) are taken from Ref. 10.

The one-band effective-mass Hamiltonian assumes a parabolic bulk dispersion relation and it neglects mixing of the zone-center conduction-band wave function with
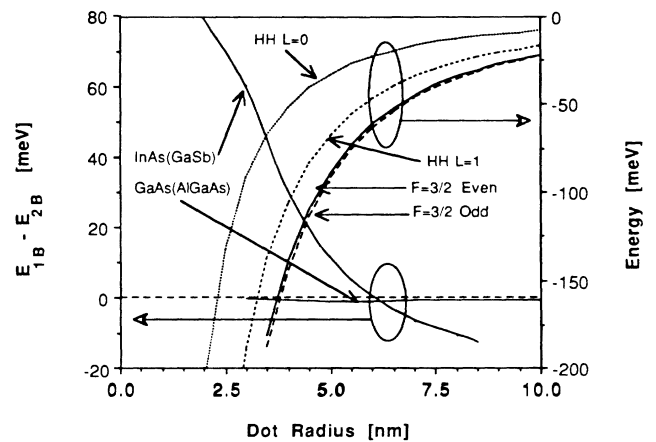

FIG. 1. Left axis: Difference between the quantum-dot ground-state energies found using a one-band effective-mass model and the total-angular-momentum formalism (two material systems are considered). Right axis: Confinement energy for the two lowest-energy valence-band states in a GaAs quantum dot plotted vs dot radius. Both one-band and multiband results are presented.

other bands for nonzero wave numbers. Both assumptions break down for small energy gaps and for strong confinement by the quantum-dot potential. In comparison against the multiband model, the ground-state binding energies are in excellent agreement for all quantum-dot radii in the large-band-gap system. For the narrow-band-gap system, however, large differences appear, which become more pronounced for stronger confinement (i.e., smaller radii).

As a second example, we consider the Γ_8 valence bands in a GaAs quantum dot. Confinement-energy calculations for the two lowest-energy valence states are presented in Fig. 1 (refer to right vertical axis) using both a one-band model and the multiband total-angular-momentum formalism. The one-band model utilizes a simple effective-mass equation for the heavy holes with states characterized by envelope angular momentum L (i.e., HH $L = 0$ and 1 are presented). It can be seen to underestimate the confinement energy at a given dot radius. This happens because it neglects mixing of the light-hole and heavy-hole bands induced by the quantum-dot potential. The one-band model also predicts a larger separation between the lowest-energy states than predicted with the multiband theory. The lowest-energy multiband eigenstates are mixtures of $|\text{LH}\rangle$ and $|\text{HH}\rangle$ having the envelope parity indicated in the figure [note that the odd-parity case emerged in consideration of Eq. (5)].

In conclusion, we have presented a formalism for solution of band structure in quantum-dot heterostructures. It is based on a reformulation of multiband envelope theory in terms of a basis set of total zone-center Bloch and envelope angular momentum. In this basis, the $\mathbf{k} \cdot \mathbf{p}$ operator is block diagonal. We have derived the $F = \frac{1}{2}$ and $F = \frac{3}{2}$ projections of this Hamiltonian for the eight-band $\Gamma_6, \Gamma_7, \Gamma_8$ system and have demonstrated the formalism by calculating the $F = \frac{1}{2}$ band structure in GaAs(AlGaAs) and InAs(GaGb) quantum dots and the $F = \frac{3}{2}$ band structure in a GaAs quantum dot. Finally, we note that in systems exhibiting cylindrical symmetry (i.e., quantum wires), a formalism based on eigenstates of F_z can be employed to simplify calculations in a manner similar to that discussed here for quantum dots.

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