Acceptor and band states in quantum wells in multiband model

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Abstract. We developed the framework for numerical calculation of the valence band dispersion in quantum well structures and the energy spectrum of the shallow acceptor in QWs. We used the finite-difference method to quantize the Luttinger-Kohn Hamiltonian and the decomposition of the impurity potential over the eigenstates of the Luttinger-Kohn Hamiltonian with the QW profile to obtain the acceptor states energies and wavefunction in $\vec{k}$-space. Developed approach allows one to calculate the energies of the acceptor states with spherical symmetry for an arbitrary QW potential profile with the coordinate-dependent material parameters.

1. Introduction

The development of compact and effective solid-state sources of terahertz (THz) radiation is an important problem which is not completely solved yet. A number of THz [1, 2] and GHz [3, 4, 5] radiation sources has been already developed, but each of them has one or more significant drawbacks. The most promising quantum cascade lasers fully fabricated in Russia [6, 7] recently was demonstrated, however the experimental samples operate in mid-IR range. The shallow impurity ionization energy in semiconductors corresponds to the THz frequency range, therefore optical transitions of charge carriers from and to shallow impurity states can be used to obtain THz radiation. To date, several mechanisms of impurity-assisted THz radiation generation in doped semiconductors have been proposed. Stimulated THz radiation associated with intracenter transitions of nonequilibrium charge carriers in silicon doped with shallow impurities [8] was detected under intense intraband optical excitation by CO$_2$ laser. Concept of THz radiation sources based on carrier transitions from and to impurity states under the conditions of interband optical pumping was first demonstrated in [9, 10] for doped bulk semiconductors of n-GaAs and p-Ge. In Refs. [11, 12] the THz radiation associated with transitions of nonequilibrium electrons from the first electron subband and excited donor states to the ground state of the donor in quantum wells (QWs) was detected and investigated experimentally.

The possibility of THz radiation generation in bulk materials can be associated with optical transitions of charge carriers between resonant and localized states of shallow impurities under conditions of external factors which lower the symmetry of the crystal, for example, under conditions of uniaxial strain [13]. Resonant states of shallow impurities can also arise in p-type
semiconductor structures with built-in mechanical strain [14], and in semiconductor structures with QWs due to size quantization effects. In this case, the shallow impurity spectrum is modified due to the spatial confinement.

It was shown, that the acceptor impurities in QW have some unique advantages related to possibility of the population of excited acceptor states due to strong polar optical phonon scattering of free holes [15]. Simultaneously, this scattering processes cannot populate the ground acceptor states because the binding energy is higher then the optical phonon energy. However, authors of [15] used the hydrogen-like impurity model to describe the energy spectrum of acceptors in QW. This is a rude first-cut approximation where the complicated valence band structure can not be taken into account.

In this work we develop the framework for numerical calculation of the valence band dispersion low in quantum well structures and the energy spectrum of the shallow acceptors in QWs taking into account the complicated valence band structure. It can be considered as the first step to experimental observation of THz emission associated with shallow acceptors in QWs.

2. Finite-difference Luttinger-Kohn Hamiltonian

It is well known, that the valence band of $A_3B_5$ materials can be well described within the effective mass approximation with the $4\times4$ Luttinger-Kohn Hamiltonian that can be expressed in a block-diagonal form (see, for example, [16]). The eigenstates of upper and lower blocks have the same energy, so one can consider first only the upper block

$$
\begin{bmatrix}
P + Q & \tilde{R} \\
\tilde{R}^\dagger & P - Q
\end{bmatrix}
$$

in the basis $|1\rangle \langle 2\rangle^T$ of heavy and light hole states, where

$$
P \pm Q = \frac{1}{2}(\gamma_1 \pm \gamma_2)(k_x^2 + k_y^2) + \frac{1}{2}(\gamma_1 \mp 2\gamma_2)k_z^2,
$$

$$
\tilde{R} = |R\rangle - i|S\rangle, \tilde{R}^\dagger = |R\rangle + i|S\rangle,
$$

$$
|R\rangle = \sqrt{3} \gamma_2 \sqrt{\gamma_2^2 (k_x^2 - k_y^2)^2 + (\gamma_3 - 2\gamma_2) k_x^2 k_y^2},
$$

$$
|S\rangle = \sqrt{3} \gamma_3 k_z \sqrt{k_x^2 + k_y^2},
$$

and $\gamma_1, \gamma_2, \gamma_3$ are the Luttinger parameters with the $\hbar^2/m_0$ multiplier included inside.

The common approach to quantize this Hamiltonian in $z$ direction in the quantum well potential $V(z)$ is the replacement of the $z$ component of the wavevector (that is no longer a good quantum number due to the break of the translational symmetry with the $V(z)$) in (1) with the differential operator $-i \frac{d}{dz}$. This leads to a system of differential equations of the form

$$
\begin{bmatrix}
\frac{d}{dz} \left( A_1 \frac{d}{dz} \right) & B \frac{d}{dz} \\
- \frac{d}{dz} \left( A_2 \frac{d}{dz} \right) & \frac{d}{dz} \left( A_2 \frac{d}{dz} \right)
\end{bmatrix}
\begin{bmatrix}
C_1 + V(z) \\
|R|
\end{bmatrix}
\begin{bmatrix}
|R| \\
C_2 + V(z)
\end{bmatrix}
\begin{bmatrix}
F_{11} \\
F_{12}
\end{bmatrix}
= \mathcal{E}
\begin{bmatrix}
F_{11} \\
F_{12}
\end{bmatrix}
$$

with respect to the energy of the valence band subbands $\mathcal{E} = \mathcal{E}(\vec{k}_||) (\vec{k}_|| = \{k_x, k_y\})$ and envelope
functions of the hole state in subbands \( F_{1,1}(z)|1\rangle + F_{2,1}(z)|2\rangle \), where

\[
A_1(z) = -\frac{1}{2}(\gamma_1(z) - 2\gamma_2(z)), \\
A_2(z) = -\frac{1}{2}(\gamma_1(z) + 2\gamma_2(z)), \\
B(z) = -\sqrt{3}\gamma_3(z)\sqrt{k_x^2 + k_y^2}, \\
C_1(z) = \frac{1}{2}(\gamma_1(z) + \gamma_2(z))(k_x^2 + k_y^2), \\
C_2(z) = \frac{1}{2}(\gamma_1(z) - \gamma_2(z))(k_x^2 + k_y^2).
\]

Applying the central-weighted finite-difference scheme similar to one used in Ref. [17] to this equations, one obtains the block-three-diagonal matrix

\[
\mathbf{H}_0 = \begin{bmatrix}
M_{11} & N_{1+} & 0 & 0 & \cdots & 0 & 0 \\
N_{1-} & M_{22} & N_{2+} & 0 & \cdots & 0 & 0 \\
0 & N_{31} & M_3 & N_{3+} & \cdots & \cdots & \cdots \\
0 & 0 & \cdots & \cdots & \cdots & \cdots & \cdots \\
0 & 0 & \cdots & 0 & N_{n-2} & M_{n-2} & N_{n-2} \\
0 & 0 & \cdots & 0 & 0 & N_{n-1} & M_{n-1} \\
0 & 0 & \cdots & 0 & 0 & 0 & N_n \\
\end{bmatrix}
\]

representing the Hamiltonian with the quantum well profile, were the \( 2 \times 2 \) \( N^\pm \) matrices in the \( i^{th} \) row

\[
M_i = -\frac{1}{2\Delta z^2} \begin{bmatrix}
A_1(z_{i+1}) + 2A_1(z_i) + A_1(z_{i-1}) & 0 & A_2(z_{i+1}) + 2A_2(z_i) + A_2(z_{i-1}) & 0 \\
0 & C_1(z_i) + V(z_i) & |R(z_i)| & 0 \\
A_2(z_{i+1}) + 2A_2(z_i) + A_2(z_{i-1}) & 0 & C_2(z_i) + V(z_i) & |R(z_i)| \\
\end{bmatrix}
\]

\[
N_i^\pm = \frac{1}{2\Delta z^2} \begin{bmatrix}
A_1(z_{i+1}) + A_1(z_i) & 0 & A_2(z_{i+1}) + A_2(z_i) & 0 \\
0 & -(B(z_{i+1}) + B(z_i)) & 0 & B(z_{i+1}) + B(z_i) \\
\end{bmatrix}
\]

operates on the \( [F_{11}(z_i), F_{21}(z_i)]^T \) and \( [F_{12}(z_{i+1}), F_{22}(z_{i+1})]^T \) vectors respectively.

Now one can directly calculate the eigenvalues and eigenvectors of the \( \mathbf{H}_0 \) at different in-plane wavevector values to obtain the subband dispersion and envelope functions.

Figure 1 shows the computed envelope wavefunctions for first two valence band subbands of the 5-nm GaAs/Al\(_{0.3}\)Ga\(_{0.7}\)As QW with material parameters taken form [18].

3. Acceptor states and heavy-light hole mixing

For a given eigenvalues \( \varepsilon_n^{\text{imp}}(|\vec{r}|) \equiv \varepsilon_\alpha \) and eigenstates \( F_{s,\vec{r}}^{n}(z) \equiv |\alpha\rangle \) of the initial QW Hamiltonian \( \mathbf{H}_0 \) (\( n \) numbers the subbands, \( s \) numbers the basis states of Hamiltonian (1)) we can expand the solution of the problem

\[
(\mathbf{H}_0 + V_{\text{imp}}(\vec{r})) \Psi = \varepsilon \Psi,
\]
Figure 1. Calculated wavefunctions of the first (a) and second (b) valence band subbands for 5-nm GaAs/Al$_{0.3}$Ga$_{0.7}$As QW.

Figure 2. Valence band dispersion and acceptor energy levels for the same QW.

Figure 3. Expansion coefficients for the ground and first excited acceptor states.

with the impurity potential $V_{\text{imp}}$ in a series over the states $|\alpha\rangle$ of the QW without impurity:

$$\Psi = \sum_{\alpha} C_{\alpha} |\alpha\rangle. \tag{16}$$

Substituting this expansion into (15) and multiplying it on the other state $\langle \beta |$ we get the equation for the expansion coefficients and the energies of the states in impurity potential in the form

$$\left( \langle \beta | V_{\text{imp}} |\alpha\rangle + \varepsilon_{\beta} \hat{1} \right) \tilde{C} = \varepsilon \tilde{C}. \tag{17}$$

After a simple transformations similar to Ref. [19], the matrix element $\langle \beta | V_{\text{imp}} |\alpha\rangle$ for the acceptor potential

$$V_{\text{imp}}(\vec{r}) = -\frac{e^2}{\varepsilon \sqrt{\rho^2 + z^2}} \tag{18}$$
can be expressed as
\[ \langle \beta | V_{\text{imp}} | \alpha \rangle = -\frac{e^2}{\varepsilon S} \frac{1}{|\Delta k||} \xi_{mn}(k', k, \theta) \] (19)
for the states without angular dependence of expansion coefficients, where
\[ \xi_{mn}(k', k, \theta) = \int \exp(-|\Delta k||z|) \sum_s \left( F_{m|s,\vec{k}'\rangle}(z) \right)^\dagger F_{n|s,\vec{k}\rangle}(z) dz, \] (20)
and \( \Delta k|| = \sqrt{\left(k'\right)^2 + k^2 - 2k'k\cos\theta}. \) However, the angular dependence of \( C \) in \( \vec{k} \)-space can be taking into account with the irreducible representations \( e^{im\theta} \) [20] substituted inside the integral (20).

Then, the final matrix that represents equation (17) has the form
\[ E_{\beta \delta \alpha} - \frac{e^2}{\varepsilon(2\pi)} \delta k k\xi(\beta, \alpha), \] (21)
where \( \delta k \) is the discretization interval in \( \vec{k} \)-space and
\[ \xi(\beta, \alpha) \equiv \xi_{mn}(k', k) = \int_{\theta=0}^{2\pi} \frac{\xi_{mn}(k', k, \theta)}{\sqrt{(k')^2 + k^2 - 2k'k\cos\theta}} d\theta. \] (22)

Again, direct calculations of the eigenvalues and eigenvectors of this matrix gives the energies of the acceptor states in the QW and the corresponding expansion coefficients.

Figure 3 shows the valence band dispersion together with the energies of the first 3 acceptor states for 5-nm GaAs/Al\(_{0.3}\)Ga\(_{0.7}\)As QW. Figure 4 shows the expansion coefficients for the acceptor wavefunctions for the same QW. Note the non-zero contribution of the first and second subband states both to the ground and excited acceptor states, that results from the intermixing of the heavy and light hole states.

4. Conclusions
In this work we developed the framework for numerical calculations of the valence band dispersion in quantum well structures and the energy spectrum of the shallow acceptor in QWs. The results of calculations show the acceptor level structure that is compatible with the scheme of increase in population of excited acceptor states due to strong polar optical scattering of free holes proposed in [15]. However, the complicated structure of the acceptor wavefunctions should lead to non-trivial spectral and polarization dependencies of optical matrix elements of the transitions with participation of the acceptor states.

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