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Interband Transitions in Quantum Wells

Przejścia międzypasmowe w studniach kwantowych

Межзонные переходы в квантовых ямах

Dedicated to
Stanisław Szpikowski on occasion
of his 60th birthday

1. INTRODUCTION

In the last few years much theoretical and experimental work has been done on the electronic and optical properties of microelectronic system whose thickness is of the order of the de Broglie wavelength of electrons [1]. In such quantum well structures the electrons and holes are confined in their motion perpendicular to the plane of the active layer, leading to the quantization effects. Quantum size effect has been observed in the interband ab-

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sorption of a number of materials, including GaAl [2], InSb [3],[4], PbTe [5], [6], PbSe and PbS [7], Bi [8], [9]. These materials (except GaAs) have a small energy gap E_g. Thus correct description of the interband transitions has to take into account the mixing of the conduction and valence band states (c-v mixing). In the bulk crystal this mixing manifests itself in:

(i) nonparabolicity of the conduction and valence band,

(ii) k-dependence of the interband matrix elements.

The purpose of this note is to investigate consequences of the c-v mixing for the interband matrix elements in the presence of the size quantization. Our approach is very similar to that developed in [10] where the intersubband transitions have been considered.

2. ENERGIES AND WAVE FUNCTIONS

Assume, like in [10] that the two-band effective mass Hamiltonian for determining the envelope function Ψ and the corresponding energy $\mathcal{E}$ has a Dirac-like form

$$\mathcal{H} = v\vec{\alpha}\cdot\vec{p} + mv^2\beta + U(z) , \tag{1}$$

where α_i and β are 4x4 Dirac matrices, $v^2 = E_g/(2m)$, m is the isotropic effective mass and $U(z)$ is the slowly varying layer potential (the jump of potential at interfaces may be taken into account by appropriate boundary conditions for Ψ [9], [11]).

From (1) one finds that effective mass equation may be written in the form

$$\left[\mathcal{E} - E_g/2 - U(z)\right]\varphi = v\,\vec{\sigma}\cdot\vec{p}\,\chi , \tag{2a}$$

$$\left[\mathcal{E} + E_g/2 - U(z)\right]\chi = v\,\vec{\sigma}\cdot\vec{p}\,\varphi , \tag{2b}$$

where φ and χ are the upper and lower part of the four-component function Ψ, respectively. The energy $\mathcal{E}$ is measured from the middle of the energy gap.

To obtain the simple analytical solution of (2) we assume that

$$\mathcal{E} - E_g/2 \ll E_g \tag{3a}$$

for the states in the conduction band and

$$|\mathcal{E} + E_g/2| \ll E_g \tag{3b}$$

for the states in the valence band.

Using the procedure similar to that in the relativistic theory [12] one finds the following expression for the electron energy in the conduction (c) and valence (v) band:

$$\mathcal{E}_{\substack{cns \\ (v)}}(\vec{k}_\parallel) = \pm E_{\substack{cns \\ (v)}}(\vec{k}_\parallel) \pm E_g/2 \tag{4}$$

where the energy $E_{lns}(\vec{k}_\parallel)$ is obtained from the effective mass equation with the "relativistic" corrections

$$H^l F_{ln\vec{k}_\parallel s} = E_{lns}(\vec{k}_\parallel) F_{ln\vec{k}_\parallel s}, \qquad (l = c, v) \tag{5}$$

with

$$H^l = H_0^l + H_1^l = \left[p^2/(2m) + U^l(z)\right] + \\ + \left[-\frac{p^4}{4m^2E_g} + \frac{\hbar}{2mE_g}\vec{\sigma}\cdot(\vec{\nabla}U^l(z)\times\vec{p}) + \frac{\hbar^2}{4mE_g}\nabla^2U^l(z)\right]. \tag{6}$$

Here $U^c(z) = U(z)$, $U^v(z) = -U(z)$, $n=1,2,\ldots$ is the subband quantum number, $s = \pm 1/2$ is the spin quantum number and $\vec{k}_\parallel = (k_x, k_y, 0) \equiv (k_\parallel \sin\varphi, k_\parallel \cos\varphi, 0)$.

In quantum well the layer potential is usually symmetric. Thus each level is doubly degenerated with respect to the spin quantum number ($E_{lns}(\vec{k}_\parallel) = E_{ln}(k_\parallel)$) and the function $F_{ln\vec{k}_\parallel s}$ may be written in the form

$$F_{ln\vec{k}_\parallel s}(\vec{r}) = e^{i\vec{k}_\parallel\cdot\vec{r}} F_{ln}(z)\,|s\rangle \tag{7}$$

where $|+\frac{1}{2}\rangle = \begin{pmatrix}1\\0\end{pmatrix}$, $|-\frac{1}{2}\rangle = \begin{pmatrix}0\\1\end{pmatrix}$.

Note that the function $F_{ln}(z)$ has a well defined parity.

The four component eigenvector of Hamiltonian (1) has the form

$$|cn\vec{k}_n\sigma\rangle = \begin{pmatrix} \varphi_{cn\vec{k}_n\sigma} \\ \left[1 - \frac{E_{cn}(\vec{k}_n) - U(z)}{E_g}\right] \frac{\vec{\sigma}\cdot\vec{p}}{2mv} \varphi_{cn\vec{k}_n\sigma} \end{pmatrix} \tag{8}$$

for the conduction band and

$$|vn\vec{k}_n\sigma\rangle = \begin{pmatrix} \left[1 - \frac{E_{vn}(\vec{k}_n) + U(z)}{E_g}\right] \frac{-\vec{\sigma}\cdot\vec{p}}{2mv} \chi_{vn\vec{k}_n\sigma} \\ \chi_{vn\vec{k}_n\sigma} \end{pmatrix} \tag{9}$$

for the valence band.

Here

$$\varphi_{cn\vec{k}_n\sigma} = \left(1 - \frac{p^2}{4mE_g}\right) F_{cn\vec{k}_n\sigma} , \tag{10a}$$

$$\chi_{vn\vec{k}_n\sigma} = \left(1 - \frac{p^2}{4mE_g}\right) F_{vn\vec{k}_n\sigma} . \tag{10b}$$

We assume that function $F_{ln\vec{k}_n s}$ (see eq. (5)) is normalized and that the penetration of the wavefunction into the interface barrier regions may be neglected.

3. CONDUCTIVITY TENSOR

When light is polarized in the x_i direction the power absorbed in a quantum well is proportional to $\mathrm{Re}\sigma_{ii}^{2D}(\omega)$ where $\ddot{\sigma}^{2D}(\omega)$ is the frequency dependent two-dimensional conductivity tensor. For direct interband transitions

$$\mathrm{Re}\,\sigma_{ii}^{2D}(\omega) = \sum_{nn'} \sigma_{ii}^{nn'}(\omega) \tag{11}$$

where

$$\sigma_{ii}^{nn'}(\omega) = \frac{e^2\pi}{\omega} \sum_{\vec{k}_\parallel ss'} f(E_{vn'}(\vec{k}_\parallel))[1-f(E_{cn}(\vec{k}_\parallel))] \cdot |\langle cn\vec{k}_\parallel s|v_i|vn'\vec{k}_\parallel s'\rangle|^2 \delta(E_{nn'}(\vec{k}_\parallel)-\hbar\omega). \tag{12}$$

Here $f(E)$ is the Fermi occupation function, $E_{nn'}(\vec{k}_\parallel) = E_{cn}(\vec{k}_\parallel) - E_{vn'}(\vec{k}_\parallel)$ and $\vec{v}$ is the velocity operator which for two-band model is defined by

$$\vec{v} = \frac{\partial \mathcal{H}}{\partial \vec{p}} = v\vec{\alpha} \,. \tag{13}$$

Assume for simplicity that the valence band states and conduction band states involved in the transitions are completely filled and empty, respectively. Since $E_{cn}^{(v)}(\vec{k}_\parallel) = E_{cn}^{(v)}(k_\parallel)$, we find without any difficulty

$$\sigma_{ii}^{nn'}(\omega) = \frac{e^2\pi}{\omega} 2\, \overline{M}_{nn'}^{(i)}(k_\parallel = k_\parallel^{nn'}) \Big]_{nn'}^{cv}(\omega) \tag{14}$$

where

$$\overline{M}^{(i)}_{nn'}(k_\parallel) = \frac{1}{2\pi}\int_0^{2\pi} d\varphi\, M^{(i)}_{nn'}(\vec{k}_\parallel) =$$

$$= \frac{1}{2\pi}\int_0^{2\pi} d\varphi \left(\frac{1}{2}\sum_{\sigma\sigma'} |\langle cn\vec{k}_\parallel\sigma|v_i|vn'\vec{k}_\parallel\sigma'\rangle|^2\right) \qquad (15)$$

is the averaged value of the squared velocity matrix element and

$$\rho^{cv}_{nn'}(\omega) = \int \frac{d\vec{k}_\parallel}{(2\pi)^2}\,\delta(\mathcal{E}_{nn'}(k_\parallel) - \hbar\omega) = \frac{1}{2\pi}\,\frac{k_\parallel^{nn'}\,\theta(\hbar\omega - \mathcal{E}_{nn'}(0))}{\left|\frac{\partial}{\partial k_\parallel}\mathcal{E}_{nn'}(k_\parallel)\right|_{k_\parallel = k_\parallel^{nn'}}} \qquad (16)$$

with $k_\parallel^{nn'}$ defined by relation

$$\mathcal{E}_{nn'}(k_\parallel = k_\parallel^{nn'}) = \hbar\omega \qquad (17)$$

is the joint density of states between the size-quantized levels $|cn\rangle$ and $|vn'\rangle$. $\theta(x)$ denotes the unit step function.

In the one band effective mass approximation the expression for the joint density of states takes the form

$$\rho^{cv(p)}_{nn'}(\omega) = \frac{m}{4\pi\hbar^2}\,\theta(\hbar\omega - \mathcal{E}^{(p)}_{nn'}) \qquad (18)$$

where $\mathcal{E}^{(p)}_{nn'} = E_g + E^{(p)}_{cn} + E^{(p)}_{vn'}$. The energy $E^{(p)}_{in}$ is obtained from the one band effective mass equation

$$\left[p_z^2/(2m) + U^i(z)\right] F^{(p)}_{in} = E^{(p)}_{in} F^{(p)}_{in}\,. \qquad (19)$$

For our purposes it will be sufficient to approximate $\rho^{cv}_{nn'}(\omega)$ by

$$\rho^{cv}_{nn'}(\omega) = \frac{m}{4\pi\hbar^2}\,\theta\left[\hbar\omega - \mathcal{E}_{nn'}(0)\right]. \qquad (20)$$

From (8-10) and (13) we find

$$\langle c\vec{k}_\parallel\sigma|\vec{v}|vn'\vec{k}_\parallel\sigma'\rangle =$$

$$=\left\langle \varphi_{cn\vec{k}_\parallel\sigma}\left| v\vec{\sigma}-\frac{1}{4m^2v}\left[1-\frac{E_{cn}(k_\parallel)-U(z)}{E_g}\right]\right.\right.\left.\left[1-\frac{E_{vn'}(k_\parallel)+U(z)}{E_g}\right](\vec{\sigma}\cdot\vec{p})\vec{\sigma}(\vec{\sigma}\cdot\vec{p})\right|\left.\chi_{vn'\vec{k}_\parallel\sigma'}\right\rangle$$

$$=\left\langle F_{cn\vec{k}_\parallel\sigma}\left| v\vec{\sigma}-\frac{v}{mE_g}\vec{p}(\vec{p}\cdot\vec{\sigma})\right|F_{vn'\vec{k}_\parallel\sigma'}\right\rangle+O(E_g^{-2}) \qquad (21)$$

In obtaining (21) we have used the relation $\sigma_\mu\sigma_\nu+\sigma_\nu\sigma_\mu=2\delta_{\mu\nu}$. Inserting (21) into (15) we find

$$\bar{M}^{(i)}_{nn'}(k_\parallel)=\bar{M}^{(i)(1)}_{nn'}(k_\parallel)+\bar{M}^{(i)(2)}_{nn'}(k_\parallel) \qquad (22)$$

where

$$\bar{M}^{(i)(1)}_{nn'}(k_\parallel)=v^2|\langle F_{cn}|F_{vn'}\rangle|^2\cdot$$

$$\left[1-(\delta_{ix}+\delta_{iy})\frac{\hbar^2k_\parallel^2}{mE_g}-\delta_{iz}\frac{2}{mE_g}\frac{\langle F_{cn}|p_z^2|F_{vn'}\rangle}{\langle F_{cn}|F_{vn'}\rangle}\right]+O(E_g^{-2}) \qquad (23)$$

and

$$\bar{M}^{(i)(2)}_{nn'}(k_\parallel)=\left(\frac{v\hbar k_\parallel}{\sqrt{2}mE_g}\right)^2|\langle F_{cn}|p_z|F_{vn}\rangle|^2(1+\delta_{iz})+O(E_g^{-3}). \qquad (24)$$

For comparison, in the parabolic approximation $\bar{M}^{(i)}_{nn'}(k_\parallel)=v^2\langle F^{(p)}_{cn}|F^{(p)}_{vn'}\rangle$.

4. DISCUSSION

Transitions induced by $\bar{M}^{(i)(1)}_{nn'}$ obey, like in the one band efective mass approximation, the selection rule $\Delta n = 0,2,4,\ldots$ (parity allowed transitions). The conduction and valence band

mixing reduces the intensity of the $\Delta n=0,2,4,\ldots$ transitions. This reduction is order of E_g^{-1} and depends on the polarization of the light. Equations (11), (14), (20) and (23) show that at the threshold for the transitions between the subbands in the conduction and valence bands whose quantum numbers n differ by an even integer, there will be a step in the absorption spectrum (more exactly in $\sigma_{ii}^{nn}(\omega)$).

The second term in (22) can be the origin of direct interband transitions with selection rule $\Delta n=1,3,5,\ldots$ (parity forbidden transitions). Intensity of these transitions depends upon the polarization of the radiation field relative to the direction of carrier confinement ($\mathfrak{M}_{nn}^{(z)(2)}/\mathfrak{M}_{nn}^{(x)(2)} = 2$) and is much smaller than intensity of the $\Delta n = 0$ transitions. Since $\mathfrak{M}_{nn}^{(i)(2)}$ is proportional to k_z^2, $\sigma_{ii}^{nn}(\omega)$ (for $\Delta n=1,3,5,\ldots$) increases continuously with the photon energy. Thus identification of the parity forbidden transitions in the interband absorption will be extremely difficult.

We expect that more suitable for the observation of the breaking of the $\Delta n=0,2,4$ selection rule for the interband transitions should be the inelastic tight scattering by intersubband excitation (for details see [13]). However, we must remember that our results have been obtained in the dipole approximation. If we go beyond this approximation the selection rule Δn even is relaxed also in the absence of the c-v mixing. In a quadrupole approximation the averaged squared optical matrix element is given by [14]:

$$\overline{M}'_{nn'} = \overline{M}^{(p)}_{nn'} + \overline{M}'^{(2)}_{nn'} \tag{26}$$

with

$$\overline{M}'^{(2)}_{nn'} = v^2 q_z^2 \left| \langle F_{cn}^{(p)} | z | F_{vn'}^{(p)} \rangle \right|^2 \tag{27}$$

where q_z is the "z" component of the "effective" wave vector of the EM radiation defined by

$$q_z = \frac{\omega}{c}\sqrt{\varepsilon}\, f(\theta) = \frac{\omega}{c}\sqrt{\varepsilon}\left(\sqrt{1-\frac{\varepsilon_0}{\varepsilon}\sin^2\theta}\right) \tag{28}$$

Here ε and ε_0 are the background dielectric constants of the layer and the ambient phase, respectively, θ is the angle of incidence of the light.

In order to have a rough estimation of $\bar{M}^{(1)}_{nn'}/\bar{M}^{(2)}_{nn'}$ for the parity forbidden transitions we assume that $U(z) = 0$, $F^{(p)}_{\nu n} = F_{\nu n}$ and $E^{(p)}_{\nu n} = E_{\nu n}$. Then, after some manipulation we obtain

$$\frac{\bar{M}^{(1)(2)}_{\nu n'}(k_u)}{\bar{M}^{(2)}_{nn'}(k_u)} = \frac{4mc^2(1+\delta_{iz})}{\mathcal{E}E_g f(\theta)}\left(\frac{E_{nn'}}{E_g}\right)^3\left(\frac{\hbar^2k_u^2}{2mE_{nn'}}\right) =$$

$$= 2\cdot 10^6\,\frac{m(m_0)(1+\delta_{iz})}{E_g(eV)\,\mathcal{E}\,f(\theta)}\left(\frac{E_{nn'}}{E_g}\right)^3\left(\frac{\hbar^2k_u^2}{2mE_{nn'}}\right) \qquad (29)$$

where $E_{nn'} = |E^{(p)}_{cn} - E^{(p)}_{cn'}|$.

From (27) we see that relative intensity of the transitions induced by $\bar{M}^{(1)(2)}_{nn'}$ and $M^{(2)}_{nn'}$ very strongly depends on the parameter $\gamma = E_{nn'}/E_g$. When γ is not too small ($\gamma \gtrsim 0.1$) (what is usually the case in the narrow band gap semiconductor layer) $\bar{M}^{(2)}_{nn'}$ plays a dominant role only if $\hbar^2k_u^2/(2m) \ll f(\theta)E_{nn'}$, i.e. at the threshold for $|vn\rangle \longrightarrow |cn'\rangle$ transitions.

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STRESZCZENIE

W pracy badano wpływ nieparaboliczności pasm w masywnym krysztale na widmo absorpcji międzypasmowej w studniach kwantowych. Obliczenia wykonano w dwupasmowym przybliżeniu masy efektywnej.

РЕЗЮМЕ

В работе исследовалось влияние непараболичности зон в массивном кристалле на спектр межзонного поглощения в квантовых ямах. Расчеты выполнены в двухзонном приближении эффективной массы.