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ELECTRICAL QUADRUPOLE TRANSITIONS IN SEMICONDUCTOR QUANTUM WELLS-BREAKDOWN OF SELECTION RULES

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Electric quadrupole transitions, both interband and intraband, in symmetric rectangular semiconductor quantum wells, are investigated. Transitions that are dipole forbidden are found to be allowed as quadrupole transitions, and among them the interband transitions in wide wells have highest intensities.

It is well known that a semiconductor quantum well (e.g. a thin *GaAs* layer embedded in *AlGaAs* bulk) supports a number of quantized electron and hole energy levels in direction perpendicular to the well plane. Their existance leads to interesting optical properties of the structure, e.g. the multiple-step like interband, and line-like intraband absorption spectrum. In symmetric wells the definite parity of electron and hole wavefunctions introduces selection rules: only odd-odd and even-even interband, and odd-even and even-odd intraband transitions are allowed, when evaluated within the electric-dipole interaction approximation /1,2/. In free atoms and molecules the dipole-forbidden transitions are often found to be allowed as magnetic dipole, electric quadrupole, etc. ones, and it is of interest to explore the same thing in semiconductor quantum wells, in view of the fact that well width is usually a considerable fraction of the wavelength of light.

The light-electron interaction Hamiltonian is given by

$$H' = \frac{e}{2m_o} (\vec{p}\vec{A} + \vec{A}\vec{p}) \tag{1}$$

where m_o is the free electron mass, $\vec{p}$ the momentum operator, and $\vec{A}$ is the magnetic vector potential, given by

$$\vec{A} = A_o \vec{e} \cdot e^{i\vec{k}\vec{r}} = A_o \vec{e}(1 + \vec{k}\vec{r} + \dots) \tag{2}$$

where $\vec{k}$ is the photon wavevector.

The first term in the expansion corresponds to electric dipole (D) interaction, and leads to the above selection rules, while the second $(\vec{k}\vec{r})$ corresponds to magnetic dipole (M) and electric quadrupole (Q) interactions, i.e.

$$H'_{M,Q} = \frac{eA_o}{2m_o} \left[(\vec{p}\vec{e})(\vec{k}\vec{r}) + (\vec{k}\vec{r})(\vec{e}\vec{p})\right] \quad . \tag{3}$$

Using the well-known vector relations, (3) may be recast as

$$H'_{M,Q} = \frac{eA_o}{2m_o} \{ \left[(\vec{k}\mathrm{x}\vec{e})(\vec{r}\mathrm{x}\vec{p})\right] + \\ + \frac{1}{2}\left[(\vec{k}\vec{r})(\vec{e}\vec{p})+(\vec{e}\vec{r})(\vec{k}\vec{p})+(\vec{e}\vec{p})(\vec{k}\vec{r})+(\vec{k}\vec{p})(\vec{e}\vec{r})\right] \} \tag{4}$$

where the first term is the magnetic dipole H'_M, and the second the electric quadrupole H'_Q interaction. Among a number of terms in (4), it is only

$$H'_{Qzz} = \frac{eA_o}{2m_o} k_z e_z (zp_z + p_z z) \tag{5}$$

that can induce transitions that are dipole forbidden (z axis is taken to be perpendicular to the well plane). Interband and intraband transitions will now be treated separately.

a) Interband transitions

Using $[p_z, z] = -i\hbar$ we get $zp_z + p_z z = 2zp_z - i\hbar$ and, since the constant term cannot cause transitions, the transition matrix element is given by

$$M^Q = \frac{eA_o}{m_o} k_z e_z \int \Psi^*_{en} \, zp_z \Psi_{hl} dz \tag{6}$$

where $\Psi_{en} = u_{oe}\psi_{en}$ is the complete wavefunction of electron in quantized state n, equal to the product of Bloch wavefunction and envelope wavefunction ψ_{en} (and similarly for holes). Using the method of separate integration of slowly varying (envelope) and rapidly varying (Bloch) functions, the integral in (6) may be transformed into $p_{cv} \cdot M^Q_{env}$, where p_{cv} is the Kane matrix element and M^Q_{env} is the quadrupole envelope matrix element, given by

$$M^Q_{env} = k_z e_z \int \psi^*_{en} \, z\psi_{hl} \, dz \tag{7}$$

For comparison, the dipole envelope matrix element is given by

$$M^D_{env} = \int \psi^*_{en}\, \psi_{hl}\, dz . \qquad (8)$$

We shall here, for simplicity, adopt the infinite barrier model of quantum well, with odd level *(m)* wavefunctions given by $A\cos(m\pi z/L)$, and even level *(j)* ones by $A\sin(j\pi z/L)$, where L is the well width, and $A=(2/L)^{1/2}$ the normalizing constant. Now (7) and (8) may be calculated analytically

$$M^D_{env} = \frac{2L}{\pi^2}\left[\frac{1}{(n+l)^2} + \frac{1}{(n-l)^2}\right]\cdot e_z k_z;\; M^D_{env} = \delta_{n,e} . \qquad (9)$$

The term $e_z k_z$ has a maximum of $k/2$ (because $\vec{e}\cdot\vec{k}=0$) when light falls at 45° incidence angle to the well plane, and is zero for normal, or in-plane incidence. Now, using $k = \bar{n}\hbar\omega/\hbar c$, $\hbar\omega = E_g + E_{en} + E_{he}$, $E_{e,h\,j} = j^2\hbar^2\pi^2/2L^2 m_{e,h}$, where $\bar{n}$ is the structure refractive index, E_g the band gap of the well semiconductor, and $m_{e,h}$ the electron (hole) effective mass, we finally have

$$M^Q_{env} = \frac{\bar{n}}{\hbar c\pi^2}\left[\frac{1}{(n+l)^2} + \frac{1}{(n-l)^2}\right]\left[E_g L + \frac{\hbar^2\pi^2}{2L}\left(\frac{n^2}{m_e} + \frac{l^2}{m_h}\right)\right] . \qquad (10)$$

Obviously, (10) will reach a minimum at some $L=L_{(min)}$. Using the material parameters E_g = 1.424 eV, m_e = 0.067 m_o, m_{hh} = 0.45 m_o (heavy holes), m_{lh} = 0.083 m_o (light holes), $\bar{n}$ = 3.6, $L_{(min)}$ is typically 20÷40 Å, so high values of M^Q_{env} should be expected for large L. Highest values are obtained if $n-l = \pm 1$ ($n-l=\pm 3$ will decrease M^Q_{env} approx. tenfold). Thus, e.g. for L=250 Å and L=150 Å $(M^Q_{env})^2$ *(2e-1hh)* is calculated to be $5.6\cdot 10^{-3}$ and $2\cdot 10^{-3}$, respectively (while $M^D_{env}=1$), which are very high values when compared with the corresponding values in free atoms. Furthermore, these values exceed the values of M^D_{env} for different index same parity transitions (like *1e-3hh*), which are not strictly forbidden if barriers are finite and ammount to 10^{-4}-10^{-3}.

b) Intraband transitions

Using $p_z = (im_o/\hbar)[H,z]$ in (5), and following the same procedure as above, the transition matrix element is now found to be

$$M^Q = -ieA_o\omega_{nl}\cdot M^Q_{env};\; M^Q_{env} = \frac{e_z k_z}{2}\int \Psi^*_n\, z^2\, \Psi_l\, dz \qquad (11)$$

while for electric dipole interaction M^Q_{env} should be replaced by M^D_{env}, where

$$M^D_{env} = e_z \int \Psi_n^* z \Psi_l \, dz \tag{12}$$

For the infinite barrier model we may calculate

$$M^Q_{env} = \frac{e_z k_z}{2} \cdot \frac{2L^2}{\pi^2}\left[\frac{1}{(n-l)^2} - \frac{1}{(n+l)^2}\right] \tag{13}$$

while M^D_{env} is given by the same expression as interband M^Q_{env} (9) with factor k_z ommited. For intraband transitions $\hbar\omega = E_{en}-E_{el} = (\hbar^2\pi^2/2m_e L^2)(n^2-l^2)$, and maximal value of M^Q_{env} may be written as

$$M^Q_{env} = \frac{\bar{n}\hbar}{cm_e} \cdot \frac{nl}{n^2-l^2} \tag{14}$$

and is independent on well width L. However, $M^D_{env} \sim L$, so M^Q_{env} becomes progressively negligible with increasing L. On the other hand, large enough L must be taken in real (finite barrier) structures to provide at least three bound levels (the 1-2 transition is dipole allowed), e.g. the $GaAs$ well in $Al_{0.5}Ga_{0.5}As$ should be at least 110 Å wide. The numerical factor $nl/(n^2-l^2)$ in (14) is largest for $n-l=\pm 2$. Thus, taking L=150 Å and $\bar{n}$=3.1 we may calculate M^Q_{env} = 0.067 Å for 1-3 transition, while M^D_{env} = 33.75 Å for 1-2 transition, and the ratio of their squares is $4\cdot 10^{-6}$, a rather small value.

In conclusion, we found that all dipole-forbidden transitions in semiconductor quantum wells are quadrupole allowed, and among them the interband transition with electron and hole level indices differing by one have highest intensities. This leads to breakdown of strict selection rules for optical transitions in semiconductor quantum wells.

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