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ELECTRIC QUADRUPOLE TRANSITIONS IN SEMICONDUCTOR QUANTUM WELLS

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Interband and intraband electric quadrupole transitions in semiconductor quantum wells are analysed. All dipole forbidden transitions are found to be quadrupole allowed, and among them the interband transitions in wide wells have the highest intensities.

I. INTRODUCTION

It is well-known that a semiconductor quantum well (e.g. a thin GaAs layer nbedded in an AlGaAs bulk) support a number of quantized electron and hole ıergy levels perpendicular in direction to the well plane. Their existence leads to teresting optical properties of the structure, e.g. the multiple-step like interband, ıd line-like intraband absorption spectrum. In symmetric wells the definite parity electron and hole wave functions introduces selection rules: only odd-odd and ven-even interband, and odd-even and even-odd intraband transition are allowed, hen evaluated within the electric-dipole interaction approximation [1—3]. In free toms and molecules the dipole-forbidden transitions are often found to be allowed s magnetic dipole, electric quadrupole, etc., and it is of interest to investigate the ıme thing in semiconductor quantum wells, in view of the fact that the well width ussually a considerable fraction of the wavelength of light.

II. METHOD AND RESULTS

In the electron-light interaction Hamiltonian $H' = (e/2m_0)(\boldsymbol{pA} + \boldsymbol{Ap})$, with m_0 the free electron mass and p the momentum operator, the magnetic vector otentials is $\boldsymbol{A} = A_0\boldsymbol{\epsilon}\exp(\mathrm{i}\boldsymbol{kr}) = A_0\boldsymbol{\epsilon}(1 + \boldsymbol{kr} + \cdots)$, where the first term in the xpansion corresponds to the electric-dipole (D), and the second to magnetic dipole

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(M) and electric quadrupole (Q) interactions ($\boldsymbol{k}$ is the photon wave vector). Using the well-known vector relations, the last two interactions may be recast as [1]:

$$H'_{MQ} = \frac{eA_0}{2m_0}\Big\{[(\boldsymbol{k}\times\boldsymbol{\varepsilon}).(\boldsymbol{r}\times\boldsymbol{p})] + \\ + \frac{1}{2}[(\boldsymbol{kr}).(\boldsymbol{\varepsilon p}) + (\boldsymbol{\varepsilon r}).(\boldsymbol{kp}) + \boldsymbol{\varepsilon p}).(\boldsymbol{kr}) + (\boldsymbol{kr}).(\boldsymbol{\varepsilon r})]\Big\} \tag{1}$$

the first term being the magnetic dipole H'_M and the second the electric quadrupole H'_Q interaction Hamiltonian. Among a number of terms in (1) only

$$H'_{Qzz} = \frac{eA_0}{2m_0}k_z\varepsilon_z(zp_z + p_z z) \tag{2}$$

can induce transitions which are dipole forbidden in QW's (the z axis is taken to be perpendicular to the QW plane). Interband and intraband transitions will now be treated separately.

II.a INTERBAND TRANSITIONS

Using $[p_z, z] = -i\hbar$, i.e. $zp_z + p_z z = 2zp_z - i\hbar$, the transition matrix element is

$$M_Q = \frac{eA_0}{m_0}k_z\varepsilon_z \int \Psi^*_{en_1} z p_z \Psi_{hn_2} \mathrm{d}z, \tag{3}$$

where the full wave function $\Psi = u_0\psi$ is the product of the Bloch wave function u_0 and the envelope function ψ, and the indices $e(h)$ denote the electrons (holes) and n_1, n_2 the numbers of the quantized levels involved in the transition. Using the method of separate integration of rapidly varying (Bloch) and slowly varying (envelope) functions, (3) may be transformed into $p_{cv}.M_{Qenv}$, where p_{cv} is the Kane matrix element, and M_{Qenv} the quadrupole envelope matrix element, given by

$$M_{Qenv} = k_z\varepsilon_z \int \psi^*_{en_1} z \psi_{hn_2} \mathrm{d}z. \tag{4}$$

For comparison, the dipole envelope matrix element is $M_{Denv} = \int \psi^*_{en_1}\psi_{hn_2}\mathrm{d}z$. For estimation, it will suffice to consider an infinite barrier model of QW, where (4) may be simply calculated to be

$$M_{Qenv} = \frac{2L}{\pi^2}\left[\frac{1}{(n_1+n_2)^2} + \frac{1}{(n_1-n_2)^2}\right].\varepsilon_z k_z, \tag{5}$$

where L is the well width, while within this simple model $M_{Denv} = \delta_{n_1,n_2}$. The term $\varepsilon_z k_z$ in (5) has a maximum of $k/2$ (because $\boldsymbol{\varepsilon}.\boldsymbol{k} = 0$) when light falls at a 45° incidence angle to the QW plane, and is zero for the perpendicular or the in-plane incidence. Now, using $k = \bar{n}\omega/c$, where $\bar{n}$ is the refractive index, and $\hbar\omega = E_g + E_{en_1} + E_{hn_2}$, $E_{e,hn} = n^2\hbar^2\pi^2/2L^2m_{e,h} + \hbar^2k_t^2/2m_{e,h}$, where E_g is the band gap of the well material, $m_{e,h}$ the electron and hole effective masses, and k_t the in-plane component of the wave vector of the electron and the hole involved in the transition, we get

$$M_{Qenv} = \frac{\bar{n}}{\hbar c\pi^2}\left[\frac{1}{(n_1+n_2)^2} + \frac{1}{(n_1-n_2)^2}\right]\times \\ \times\left[E_gL + \frac{\hbar^2\pi^2}{2L}\left(\frac{n_1^2}{m_e} + \frac{n_2^2}{m_h}\right) + \frac{\hbar^2k_t^2}{2}\left(\frac{1}{m_e} + \frac{1}{m_h}\right).L\right], \tag{6}$$

which obviously reaches a minimum at some $L = L_{min}$. With values $E_g = 1.424$ eV, $m_e = 0.067m_0$, $m_{hh} = 0.45m_0$ (heavy holes), $m_{eh} = 0.083$ (light holes), $\bar{n} = 3.6$ for GaAs, L_{min} is typically $20-40$ Å, and high values of M_{Qenv} should be expected for large L, especially if $n_1 - n_2 = \pm 1$ ($n_1 - n_2 = \pm 3$ wil decrease M_{Qenv} approx. tenfold). Thus M^2_{Qenv} for the $2e-1hh$ transition at the absorption edge ($k_t = 0$) is calculated to be, e.g. 2×10^{-3} and 5.6×10^{-3} for $L = 150$ Å and 250 Å QW's respectively. These values are comparable with M^2_{Denv} for the same parity different index transitions (like $1e-3hh$), which are not strictly forbidden in real, finite barrier structures [2] and should be within the range of measurability.

II.b. INTRABAND TRANSITIONS

Using $p_z = (im_0/\hbar)[H_0, z]$ in (2), and proceeding along the same lines as above, the transition matrix element is now found to be $M_Q = -ieA_0\omega.M_{Qenv}$,

$$M_{Qenv} = \frac{\varepsilon_z k_z}{2}\int \psi^*_{n_1} z^2 \psi_{n_2}\mathrm{d}z \tag{7}$$

and $M_D = -ieA_0\omega.M_{Denv}$, $M_{Denv} = \varepsilon_z\int\psi^*_{n_1}z\psi_{n_2}\mathrm{d}z$ where wave functions are either all electron or all hole ones. For the infinite barrier model we get

$$M_{Qenv} = \frac{\varepsilon_z k_z}{2}\cdot\frac{2L^2}{\pi^2}\left[\frac{1}{(n_1-n_2)^2} - \frac{1}{(n_1+n_2)^2}\right] \tag{8}$$

while M_{Denv} is the same as M_{Qenv} in the interband case (eqn. (5)) with the factor k_z ommitted (note that n_1 and n_2 now have the same parity for Q and the opposite parity for D transitions). For intraband transitions (e.g. in the conduction band) $\hbar\omega = E_{en_1} - E_{en_2} = (\hbar^2\pi^2/2m_eL^2)(n_1^2 - n_2^2)$ and the maximum value of (8) with $\varepsilon_z k_z = k/2$ and $k = \bar{n}\omega/c$, is

$$\bar{M}_{Qenv} = \frac{\bar{n}\hbar}{cm_e}\cdot\frac{n_1n_2}{n_1^2 - n_2^2} \tag{9}$$

and is independent of L. However, $M_{Denv} \sim L$, so M_{Qenv} becomes progressively negligible with increasing L. On the other hand, a large enough L must be taken in a real, finite-barrier structure to provide at least three bound levels (the 1—2 transition is dipole allowed), and, e.g., the GaAs QW in $Al_{0.5}Ga_{0.5}As$ should be at least 110 Å wide. The numerical factor with n_1 and n_2 in (9) is the largest for $n_1 - n_2 = 2$. Thus, taking $L = 150$ Å and $\bar{n} = 3.1$ we find $M_{Qenv} = 0.067$ Å for the 1—3 transition, while $M_{Denv} = 33.75$ Å for the 1—2 transition, the ratio of their squares being very small ($\sim 4 \times 10^{-6}$).

III. CONCLUSION

All dipole forbidden transitions in symmetric QW's are quadrupole allowed, but among them only the interband transitions, with level indices differing by one have high enough intensities to be dectectable.

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ЕЛЕКТРИЧЕСКИЕ КВАДРУПОЛЬНЫЕ ПЕРЕХОДЫ В КВАНТОВОЙ ЯМЕ ПОЛУПРОВОДНИКА

В работе сделан анализ межполосных и внутриположных електрических квадрупольных переходов в квантовой яме полупроводников. Поскольку все дипольныйе переходы оказываются запрещенными, и квадрупольные разрешенными, в широкой яме имеют внутреполосные переходы, повышенную интенсивность.