TWO-PHOTON ABSORPTION IN INVERTED SEMICONDUCTORS AND THEIR SIZE-QUANTIZED FILMS

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The TPA of light in semiconductors with inverted band structure and their size quantized films is theoretically considered. The general formula for TPA coefficient for the possible geometries in the framework of two-band model are obtained. It was shown that in the film case the TPA spectrum exhibits the sharp peaks due to the size quantization of carriers energies.

Currently, much attention has given to the linear and nonlinear optical properties of the quantum nanostructures such as semiconductor quantum wells, wires and dots [1-3]. These properties depends sensitively on the transition matrix elements between the eigenstates of the carriers, size of nanostructure and band gap of bulk semiconductor.

Consequently, a consideration of the optical effects in such structures demands an account of the real energy spectrum and wave functions.

A large number of semiconductor compounds have an inverted band structure, where the conduction band (c) and heavy hole band (h) are degenerated at the $\vec{k}=0$ point. The typical examples are α-Sn, HgTe, $Hg_{1-x}Cd_xTe$ ($x < 0.15$), $Hg_{1-x}Mn_xTe$ ($x<0.07$). Since the bands (c) and (h) have the same parity, the dipole transition (h)→(c) is forbidden [4]. In such situation the two-photon absorption may be effective.

Two-photon absorption (TPA) in semiconductors with normal band structure has been considered in [5]. The reference of the papers that are concerned with TPA right up till 1972 are contained in the rather well detailed paper of Lee and Fan [6], in which $A^{III}B^{V}$ compounds are considered. In [7-8] TPA in InSb type narrow gap semiconductors has been studied.

In present paper TPA connected with (h)→(c) transitions in semiconductors with inverted band structure and their size-quantized films is investigated. The case of intrinsic semiconductor at the temperature $T = 0$ is considered. In other words it is supposed that the valence bands are entirely filled up, but conduction band is empty. The general expression for the absorption coefficient of photon of energy $h\nu_1$ in presence of photons of energy $h\nu_2$ has the form [9]:

$$K_2(h\nu_1) = \frac{e^4}{n_1 n_2 c^2 m^4 \nu_1 \nu_2^2} \sum_{f,i} \left| \sum_t \left\{ \frac{\vec{e}_2 \vec{M}_{ft} \vec{e}_1 \vec{M}_{ti}}{E_t - E_i - h\nu_1} + \frac{\vec{e}_1 \vec{M}_{ft} \vec{e}_1 \vec{M}_{ti}}{E_t - E_i - h\nu_2} \right\} \right|^2 \delta\left[E_f - E_i - h(\nu_1 + \nu_2)\right] \quad (1)$$

$$\vec{M}_{ft} = \langle f|\vec{P}|t\rangle, \qquad \vec{M}_{ti} = \langle t|\vec{P}|i\rangle$$

Here e is the electron charge, c is the light velocity, n_1 and n_2 are the refraction coefficients corresponding to the frequencies ν_1 and ν_2; $\vec{e}_1$, $\vec{e}_2$ are the corresponding polarizations, E_i, E_f, E_t are the energies of initial, intermediate and final states of electron, $|i\rangle$, $|f\rangle$, $|t\rangle$, are the wave functions, respectively.

In fig.1a the band structure of inverted semiconductor and possible TPA process are shown.

I. Spectrum and wave functions for arbitrary k in a parabolic approximation can be obtained from the expressions (19) of the work [10] if $\varepsilon_g \to \infty$, i.e. $\varepsilon << \varepsilon_g$, where ε is the energy counted from extremum of bands and ε_g is the energetic distance between extrema of (1) and (h) bands:

$$\varepsilon_\mu(\vec{k}) = \varepsilon_{\mu o} \pm \frac{h^2 k^2}{8\pi^2 m_\mu} , \qquad (2)$$

$$\varphi_{c\vec{k}\uparrow}(\vec{r}) = \left[\frac{1}{2}\frac{k_+}{k} u_4 + \frac{k_y}{k} u_5 + \frac{\sqrt{3}}{2}\frac{k_-}{k} u_6\right] e^{i\vec{k}\vec{r}}$$

$$\varphi_{h\vec{k}\uparrow}(\vec{r}) = \left[\frac{1}{2}\frac{k_+}{k} u_3 + \frac{\sqrt{3}}{2}\frac{k_-}{k} u_5 + \frac{k_z}{k}\frac{k_-^2}{k_\perp^2} u_6\right] e^{i\vec{k}\vec{r}} , \qquad (3)$$

$$\varphi_{1\vec{k}\uparrow}(\vec{r}) = u_2 e^{i\vec{k}\vec{r}}$$

$$\varphi_{\mu k\downarrow}(\vec{r}) = \hat{R}\varphi_{\mu\vec{k}\uparrow}(\vec{r}) , \quad \hat{R} = \hat{K}\hat{I} , \quad \mu \equiv (c, h, 1) \qquad (4)$$

Here K is the time-reversal operator and I is the space-inversion operator, $k_\pm = k_x \pm ik_y$, $k_\perp^2 = k_x^2 + k_y^2$, u_1 to u_6 are the Bloch amplitudes:

$$u_1 = |iS\downarrow\rangle, \qquad u_2 = |iS\uparrow\rangle,$$

$$u_3 = \frac{1}{\sqrt{2}}|(X - iY)\downarrow\rangle, \qquad u_6 = \frac{1}{\sqrt{2}}|(X + iY)\uparrow\rangle,$$

$$u_4 = \sqrt{\frac{2}{3}}|Z\downarrow\rangle + \frac{1}{\sqrt{6}}|(X - iY)\uparrow\rangle, \qquad u_5 = \sqrt{\frac{2}{3}}|Z\uparrow\rangle - \frac{1}{\sqrt{6}}|(X + iY)\downarrow\rangle,$$

where m_c, m_h, m_l are the effective masses of the conduction electrons, heavy holes and light holes, respectively X, Y, Z represent the functions transforming like atomic p-functions and S transforms like an atomic S-function.

In the bulk case for the isotropic crystal of cubic symmetry there are two different geometries zz and xz. Using the spectrum and the wave functions (2)-(4) we get for TPA coefficient following expressions:

$$K_2^{zz}(\nu_1) = \frac{256\pi^5}{45}\frac{e^4P^4}{n_1n_2c^2h}\left(\frac{2m_{ch}}{h^2}\right)^{3/2}\left[\frac{1}{A_o} + \frac{1}{B_o}\right]^2\frac{\left[h(\nu_1 + \nu_2)\right]^{1/2}}{h\nu_1(h\nu_2)^2}, \tag{5a}$$

$$K_2^{xz}(\nu_1) = \frac{352\pi^5}{45}\frac{e^4P^4}{n_1n_2c^2h}\left(\frac{2m_{ch}}{h^2}\right)^{3/2}\left[\frac{1}{A_o^2} + \frac{1}{B_o^2} + \frac{2}{11}\frac{1}{A_oB_o}\right]\frac{\left[h(\nu_1 + \nu_2)\right]^{1/2}}{h\nu_1(h\nu_2)^2}, \tag{5b}$$

where $P^2=3h^2\varepsilon_g/(16\pi^2m_c)$ is Kane's constant,

$$A_o = -\frac{m_{ch}}{m_{lh}^-}\left[h(\nu_1 + \nu_2)\right] - \varepsilon_g - h\nu_1, \qquad \frac{1}{m_{ch}} = \frac{1}{m_c} + \frac{1}{m_h},$$

$$B_o = -\frac{m_{ch}}{m_{lh}^-}\left[h(\nu_1 + \nu_2)\right] - \varepsilon_g - h\nu_2, \qquad \frac{1}{m_{lh}^-} = \frac{1}{m_l} - \frac{1}{m_h},$$

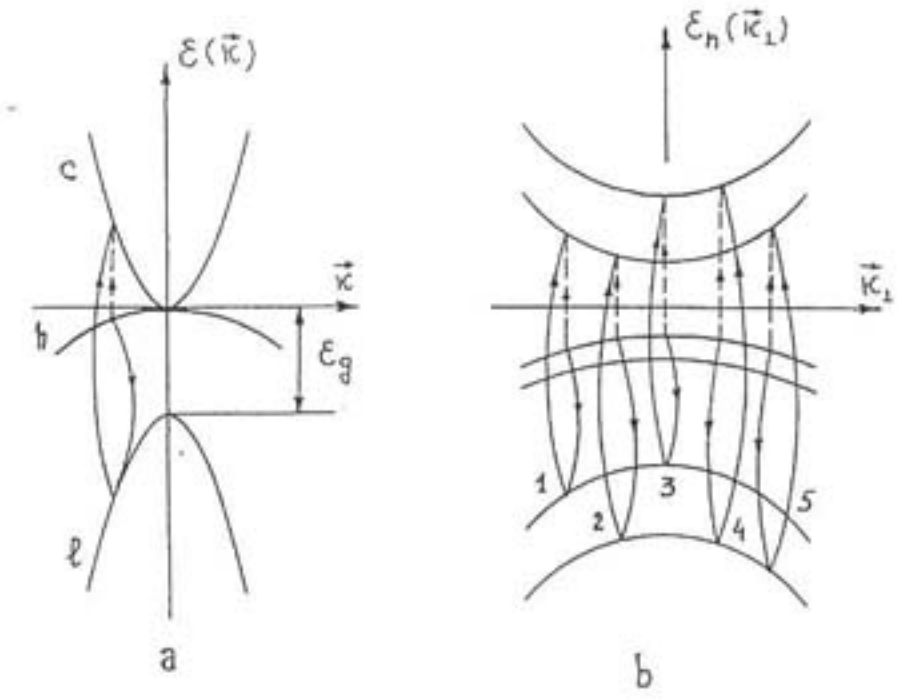

Fig.1. Band structure of HgTe near the Γ point of the Brillouin zone and possible TPA processes: a) bulk crystal case; b) size quantized film case.

II. In a size quantized films of the semiconductors with a normal band structure TPA has been investigated in [11]. However the author does not take into account the dependence of matrix element on wave vector $\bar{k}$. Taking into account this we found correct result with the concrete polarization dependences.

In the thin semiconductor film when thickness comparable with the de-Broglie wavelength, the wave vector $\bar{k}$ in this direction becomes discrete, i.e. $k_z=\pi n/d$, $n=1,2,$ and consequently the energy quantizes also. This lead to the elimination of degeneracy and the gap ε_h^o appears in zero

gap structure, that is equal to the energetic distance between the first size subbands of the heavy hole and conduction bands. In the interval $h\nu_1, h\nu_2 < \varepsilon_h^\circ < h(\nu_1+\nu_2)$ the one photon absorption is forbidden both by symmetry and by energy. In this case TPA can gives an essential information about the energy spectrum at the vicinity of $\bar{k}_\perp = 0$. There is four different geometries in the thin film case. That are zz, zx, xx and xy. For calculation of TPA coefficient we have used the spectrum and wave functions, that are obtained from the solution of Schredinger equation at $\bar{k}\cdot\bar{p}$ approximation for the potential well with the infinitily deep well [12]. We assume that the surfaces of the layer a represented by high potential walls at the planes $z=0$ and $z=Na=d$, where $N(N>>1)$ is the number of crystal layers in the films. We also assume that $\varphi\left(\overline{r_\perp}, z=0\right) = \varphi\left(\overline{r_\perp}, z=d\right) = 0$. Then the spectrum and wave functions may be written in the form:

$$\varepsilon_{\mu n_\mu}\left(\vec{k}_\perp\right) = \varepsilon_{\mu o} \pm \frac{h^2 k_\perp^2}{8\pi^2 m_\mu} \pm \varepsilon_\mu^\circ n_\mu^2 \qquad (6)$$

$$\varphi_{\mu\vec{k}_\perp n_\mu \sigma_\mu}(\vec{r}) = A_{\mu\vec{k}_\perp n_\mu\sigma_\mu}(\vec{r})\sin\left(\frac{\pi n_\mu}{d} z\right) + B_{\mu\vec{k}_\perp n_\mu\sigma_\mu}(\vec{r})\cos\left(\frac{\pi n_\mu}{d} z\right) \qquad (7)$$

Here $A_{\mu\vec{k}_\perp n_\mu\sigma_\mu}(\vec{r})$ and $B_{\mu\vec{k}_\perp n_\mu\sigma_\mu}(\vec{r})$ are the combination of the Bloch amplitudes, $\sigma_\mu = (\uparrow\downarrow)$ and $\mu \equiv (ch1)$. Note that $B_{\mu\vec{k}_\perp n_\mu\sigma_\mu}(\vec{r}) = 0$ for $z=(0;d)$.

Using (7) one may calculate the matrix elements in (1). As seen from fig.1b and (7), the presence of the size levels leads to the possibility of appearance of three types processes: (i) $\Delta n_{ch}=0$ ($\Delta n_{1h}=\Delta n_{1c}=0$ or $n_c=n_h\neq n_1$, processes 1 and 2);(ii) $\Delta n_{ch}=2s+1$,where $s=0,\pm1,\pm2,\ldots$($\Delta n_{1h}=2s+1$, $\Delta n_{c1}=0$ or $\Delta n_{1h}=0$, $\Delta n_{c1}=2s+1$, processes 3 and 4); (iii) $\Delta n_{ch}=2s$ ($\Delta n_{1h}=2s+1$, $\Delta n_{c1}=2s'+1$, processes 5), where $\Delta n_{\mu'\mu}=n_{\mu'}-n_\mu$.

The final expressions for TPA coefficient has the form ($\nu_1=\nu_2=\nu$):

$$K_2(h\nu) = \frac{32\pi^5}{3}\frac{e^4P^4}{n^2c^2h}\frac{m_{ch}}{dh^2}\sum_{n_c n_h} f(\nu)\frac{\theta(\Lambda_{ch})}{(h\nu)^3}, \qquad (8)$$

where for the different possible geometries $f(\nu)$ has the following form:

$$f^{zz}(\nu) = 4\left(\frac{\xi_{1c}}{A_o}\right)^2\delta_{n_c n_h} + 8\xi_{1c}\xi_{2c}\frac{\left|T_{n_c n_h}\right|^2}{A_2^2}, \qquad (9a)$$

$$f^{xx}(\nu) = \left[\frac{7}{2}\left(\frac{\varepsilon_{1c}}{A_o}\right)^2 + \frac{\varepsilon_{1c}\varepsilon_{2c}}{A_o}\sum_{n_1}\frac{\left|T_{n_c n_1}\right|^2}{A}\right]\delta_{n_c n_h} + \varepsilon_{2c}\varepsilon_{2h}\left[\frac{\varepsilon_{1c}}{\varepsilon_{2c}}\left(\frac{5}{A_1^2}+\frac{1}{A_2^2}-\frac{3}{A_1A_2}\right)\left|T_{n_c n_1}\right|^2 + \left|\sum_{n_1}\frac{1}{A}T_{n_c n_1}T_{n_1 n_h}\right|^2\right], \qquad (9b)$$

$$f^{xz}(\nu) = 4\left(\frac{\varepsilon_{1c}}{A_o}\right)^2\delta_{n_c n_h} + \varepsilon_{2c}\varepsilon_{2h}\left[\frac{\varepsilon_{1c}}{2\varepsilon_{2c}}\left(\frac{1}{A_1^2}+\frac{1}{A_2^2}\right)\left|T_{n_c n_h}\right|^2 + \left|\sum_{n_1}\frac{1}{A}T_{n_c n_1}T_{n_1 n_h}\right|^2\right], \qquad (9c)$$

Here

$$\Lambda_{ch} = 2h\nu - \varepsilon_c^\circ n_c^2 - \varepsilon_h^\circ n_h^2,$$

$$A = -\frac{m_{ch}}{m_{1h}}\Lambda_{ch} - \varepsilon_g - \varepsilon_1^\circ n_1^2 + \varepsilon_h^\circ n_h^2 - h\nu, \qquad \varepsilon_{1\mu} = \frac{\Lambda_{ch}}{\Lambda_{ch} + \varepsilon_{ch}^\circ n_\mu^2}, \qquad (10)$$

$$A_1 = A\delta_{n_c n_1}, \quad A_2 = A\delta_{n_1 n_h}, \qquad \varepsilon_{2\mu} = \frac{h^2/\left(8\pi^2 m_{ch} d^2\right)}{\Lambda_{ch} + \varepsilon_{ch}^\circ n_\mu^2},$$

$\theta(x)$ is the step function, ε_v° are the energies of the first size subbands,

$$T_{n_\mu n_\mu} = \left[1 - (-1)^{n_\mu' + n_\mu}\right] \frac{2n_\mu' n_\mu}{\left(n_\mu'\right)^2 - n_\mu^2}$$

The curves in Fig.2 show the dependences of TPA coefficient on parameter $\alpha=h\nu/\varepsilon_g$ in the case of zz and xx polarizations for d=180 Å, ε_g=0,28 eV, $m_1=m_c$=0,029m_o, m_h=0,4m_o (HgTe). The estimations show, that in the bulk case when $h\nu$=0,117 eV for HgTe in zz geometry $K_2 \approx$ 0,357 cm/MW. It is seen that the curves for K_2 in the zz and xx are qualitatively different, TPA starts from the threshould, which corresponds to the energy distance between the first subbands of the conduction and heavy hole bands $\left(\varepsilon_{ch}^\circ\right)$. The arrows indicate the transitions with the selection rules Δn_{ch}=-2 (arrows 2, 6, 12), Δn_{ch} =-4 (arrows 3, 7, 13), Δn_{ch} =-6 (arrows 5, 9), Δn_{ch} =1 (arrows 4, 11) and Δn_{ch} =2 (arrows 10).

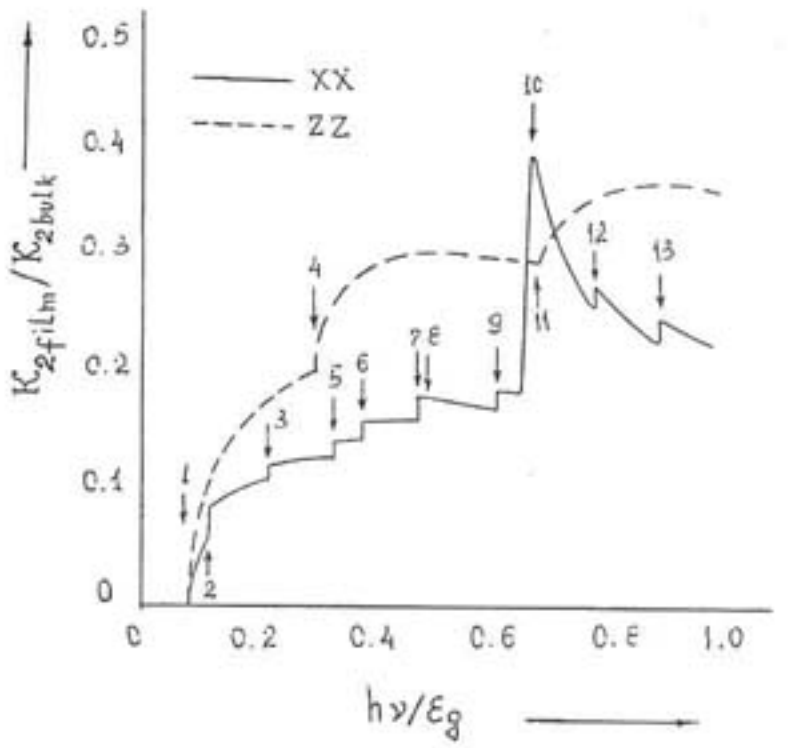

Fig.2. The ratio of the two-photon absorption coefficients (K_{2film}) in a size quantized films of semiconductors with inverted band structure to that in bulk (K_{2bulk}) is shown as a function of the normalized photon energy $\alpha=h\nu/\varepsilon_g$. The parameters used in the calculation are characteristic of HgTe.

[1] *S.V. Gaponenko.* FTP, 1996, v.30, № 4, 557.

[2] *Al.L.Efros, M.Rosen, M. Kuno, M. Nirmal, D.J. Norris, M.Bawendi.* Phys.Rev. B, 1996-I, v.54, №7, 4843.

[3] *A.V.Fedorov, A.V.Baranov.* Phys.Rev.B, 1996-II, v.54, № 12, 8627.

[4] *N.B. Brandt, V.V. Moshalkov.* Adv. in Physics, 1984, v.33, № 3.

[5] *N.G. Basov, A.Z. Geesyuk, I.G.Zubarev, V.A. Katulin, O.N. Krokhin.* Zh. Eksp. Teor. Fiz.., 1996, v.50, 551/ Sov. Phus. - JETP, 1996, v.23, 366/.

[6] *C.C.Lee, H.Y.Fan.* Phys. Rev.B.,1974, v.9, № 8, 3502.

[7] *A.M.Johnston, G.R. Pidgeon, J. Dempsey.* Phys. Rev. B., 1980, v.22, 825.

[8] *T.G. Ismailov, M.A. Bagirov.* Phys. Stat. Sol. (b) 1992, 17, 371.

[9] *F. Bassani, G. Pastori Parravicini.* Electronic States and Optical Transitions in Solids, Pergamon Press, 1975.

[10] *M.A. Mekhtiev, T.G. Ismailov.* Phys. Stat. Sol. (b), 1980, v.99, 507.

[11] *M. Zaluzny.* Thin Solid Films, 1978, v.55, 243.

[12] *H.N. Spector.* Phys. Rev. B., 1987, v.35, № 11, 5876.

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İNVERS YARIMKEÇİRİCİLƏRDƏ VƏ ONLARIN ÖLÇÜYƏ GÖRƏ KVANTLANMIŞ NAZİK TƏBƏQƏLƏRİNDƏ İKİFOTONLU UDULMA

İnvers yarımkeçiricilərdə və onların ölçüyə görə kvantlanmış nazik təbəqələrində ikifotonlu udulma hadisəsinə baxılmışdır. İki zonalı Keyn modeli daxilində ikifotonlu udulma əmsalı üçün işığın mümkün olan bütün polyarizasiyalarını nəzərə alan formul alınmışdır. Göstərilmişdir ki, nazik təbəqə halında ikifotonlu udulma spektri zərrəciklərin enerjisinin ölçüyə görə kvantlanmasının nəticəsi olan sərt piklərə malikdir.

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ДВУХФОТОННОЕ ПОГЛОЩЕНИЕ В ИНВЕРСНЫХ ПОЛУПРОВОДНИКАХ И ИХ РАЗМЕРНО-КВАНТОВАННЫХ ПЛЕНКАХ

Рассмотрено двухфотонное поглощение света в инверсных полупроводниках и их размерно-квантованных пленках. В рамках двухзонной модели Кейна получена общая формула для коэффициента двухфотонного поглощения, учитывающая всевозможные геометрии эксперимента. Показано, что в случае пленки спектр ДФП содержит резкие пики, обусловленные размерным квантованием энергий носителей.

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