Electronic Spectrum of a Quasi-2D Semiconductor in Strong Electromagnetic Field

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The change in the shape of the spectra of a quasi-2D semiconductor under the action of resonant irradiation of the frequency Ω, which is determined by the condition $0 \leq \hbar \cdot \Omega - E_g \ll E_g$, is considered. The studies showed three types of such a changes. They were analysed in the two cases: a) depending on Ω at fixed parameters of the quasi-2D semiconductor bands (effective masses m_c, m_h of electrons and holes in the plane of layers, overlap integrals β_c, β_h) and b) vice versa, when the band parameters change at fixed Ω. The decisive role of β_c, β_h in the manifestation of one or another of the three types of the spectrum shape change is indicated.

Keywords: Quasi-2D Semiconductor, Electronic Spectrum, Resonant Electromagnetic Irradiation.

1. INTRODUCTION

In nature, there are a number of so-called low-dimensional structures or layered crystals that have pronounced anisotropic properties. Such structures are packs of mono-atomic planes (packets) whose atoms are interconnected by covalent or ionic-covalent bonds, whereas the connection between the packets is created by much weaker, namely van der Waals, bond. Therefore, sometimes such structures are called quasi-two-dimensional (quasi-2D) structures. Among them, the most widespread are graphite, transition metal dichalcogenides (TMD) MX_2 (M: Mo, Ta, Ti, W, Nb, Sn, Zr, Hf, V; X: S, Se, Te), compounds of A_3B_6-type (A: Ga, In; B S, Se, Te), and others. As an example, in Fig. 1 packets S–Mo–S of the layered crystal MoS_2 are shown. The structure of the well-known representative of the layered crystals of A_3B_6-type GaSe completely coincides with the structure of MoS_2 crystals after replacing the layer of metal atoms M by a Ga–Ga double layer.
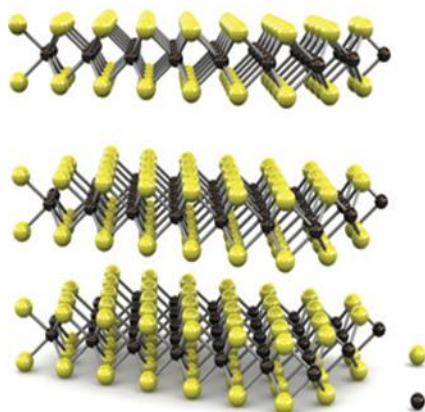

Fig. 1 – MoS2 crystal cells

Different types of bonds are the cause of pronounced anisotropy concerning physical characteristics of quasi-two-dimensional crystals, which anisotropy can be widely varied by intercalation [1, 2], by external compression [3], etc. Therefore, such structures were (and remain) objects of intensive scientific researches and their practical use in nanoelectronics, spintronics, and optoelectronics [4-6]; and especially, they are promising in solving the current scientific and technological problem – the creation of high-capacity electrical energy storage [7].
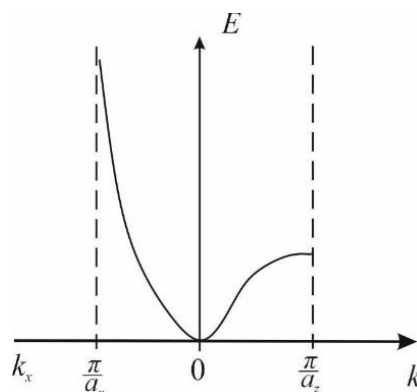

Fig. 2 – Dispersion law of an electron in a quasi-2D crystal

The interest in layered crystals renewed sharply after the discovery of graphene in 2004 [8]. Graphene is a exfoliated nanosheet of graphite. (A similar to it structure – graphdiyne – was synthesized in 2019 [9]). The first studies of graphene have showed extaordinary electrical and thermal conductivities, the possibility of many of its practical applications for high-speed electronic and optical devices, for hybrid materials based on graphene, for energy generation and storage.

Structure and chemical bonds of graphene are similar to those of TMD or A_3B_6 layered crystals. Long before the discovery of graphene, individual or few layers separated from TDM or from layered crystals were obtained [10, 11].

In search of desired characteristics, intensive research has recently been focused on mono- and multilayer graphene or TMD-structures. Significant differences have been found in the band spectra of mono- and bi-layered compounds as compared to those of bulk (quasi-2D) crystals. For example, bulk MoS_2 is a semiconductor with an indirect band gap of 1.2 eV with negligible photoluminescence – becomes a direct band gap semiconductor with a band gap of 1.8 eV and

with a strong photoluminescence in the case of a individual monolayer [12, 13]. The band gap of 2.3 eV in tri-layer ε-GaSe becomes a band gap of 3.3 eV in monolayer ε-GaSe, while for quasi-2D-crystals it is 1.91 eV. When passing through a certain critical number of layers, the effective mass of holes at point Γ changes its sign from positive to negative. Successful practical application of the peculiar properties of 2D crystals or their cleaved fragments is possible only on the basis of thorough knowledge of their electronic states. This explains a significant number of, in particular, theoretical studies using various methods (in particular, DFT and its modifications) of such states. In particular, the peculiar properties of graphene as 2D structure described by the relativistic Dirac equation were discovered. It was found that the band structure in the vicinity of point K can have a conical shape – Dirac cones. At this point electrons are zero-mass particles, i.e., they become massless fermions instead of the usual quasiparticles in solids. Such a conclusion raises a number of questions [14]. First, how justified is the model of graphene as an infinite 2D crystal, bearing in mind that real 2D crystals are thermodynamically unstable [15]? Second, how justified is the application of Dirac's theory to the structure of graphene with its small nuclear charge (+6e), capable of causing significant relativistic effects [14]. However, the cone-shaped bands observed in graphene photoemission experiments can be described within the framework of conventional non-relativistic DFT calculations, rather than with the help of massless fermions according to the Dirac equation

Given the importance of knowing the band spectrum and its properties for electronics, comprehensive studies of the spectrum, in particular, its response to external factors, are relevant.Below we will consider one of the mechanisms of band spectrum reconstruction in quasi-2D crystals.

2. EFFECT OF AN ELECTROMAGNETIC WAVE ON AN ANISOTROPIC SEMICONDUCTOR WITH PARABOLIC ZONES

Let us consider the absorption by a semiconductor of a monochromatic electromagnetic wave whose an electric field intensity, in general, has the following form:

$$\vec{E}(\vec{r},t) = \vec{e} E_0 \exp\left[i\left(\vec{q}\vec{r} - \Omega t\right)\right] \qquad (1)$$

where $\vec{e}$ is the unit vector of light polarization, $\vec{q}$ is its wave vector, and Ω is its frequency

Let the stationary states and wave functions of an ideal isotropic semiconductor be known:

$$\hat{H}_0 \varphi_{c,v}(\vec{r}) = E_{c,v} \varphi_{c,v}(\vec{r}) \qquad (2)$$

where

$$E_{c\vec{k}} = \frac{E_g}{2} + \frac{\hbar^2 k^2}{2m_c}, \qquad (3)$$

$$E_{v\vec{k}} = -\frac{E_g}{2} + \frac{\hbar^2 k^2}{2m_v}. \qquad (4)$$

Here (3), (4) are the dispersion laws of an electron in terms of the effective masses m_c, m_v in conduction band and in valence band, respectively, referenced from the middle of the band gap E_g.

The effect of electromagnetic irradiation of a semiconductor is described by the Hamiltonian

$$\hat{H}_1 = -\frac{e}{mc}\vec{A}\hat{\vec{p}} \Rightarrow i\frac{e}{mc}\vec{e}\hat{\vec{p}}E_0 \exp\left[i\left(\vec{q}\vec{r} - \Omega t\right)\right] \qquad (5)$$

where

$$\hat{\vec{p}} = -i\hbar\left(\vec{e}_x \frac{\partial}{\partial x} + \vec{e}_y \frac{\partial}{\partial y} + \vec{e}_z \frac{\partial}{\partial z}\right)$$

is the momentum operator,

$$\vec{A} = \frac{ic}{\Omega}\vec{E}$$

is the vector potential, and *e, m* are the charge of the electron and its mass, respectively.

Let us present the final Hamiltonian

$$\hat{H} = \hat{H}_0 + \hat{H}_1$$

in the representation of secondary quantization basing on the operator wave function

$$\hat{\psi}(\vec{r},t) = \sum_{\vec{k}}\left[a_{\vec{k}c}\varphi_{\vec{k}c}(\vec{r})\exp\left(-i\frac{E_c t}{\hbar}\right) + a_{\vec{k}v}\phi_{\vec{k}v}(\vec{r})\exp\left(-i\frac{E_v t}{\hbar}\right)\right] (6)$$

where $a_{\vec{k}c}^+(a_{\vec{k}c})$, $a_{\vec{k}v}^+(a_{\vec{k}v})$ are creation operators (annihilation) of an electron in the conduction band and valence band, respectively. Then

$$\hat{H} = \sum_{\vec{k}}\left(E_c a_{\vec{k}c}^+ a_{\vec{k}c} + E_h b_{\vec{k}h}^+ b_{\vec{k}h}\right) + \\ + \sum_{\vec{k}\vec{k}'}\left(\lambda_{\vec{k}\vec{k}'} a_{\vec{k}c}^+ b_{\vec{k}'h}^+ \exp\left(-i\Omega t\right) + h.c.\right) \qquad (7)$$

where the first two terms describe the electron and hole dispersion laws $(b_{\vec{k}h}^+(b_{\vec{k}h}))$ the hole creation (annihilation) operator) with

$$E_{h\vec{k}} = \frac{E_g}{2} + \frac{\hbar^2 k^2}{2m_h}; \ (m_v = -m_h).$$

The third term in (7) describes the effect of electron and hole mixing by an electromagnetic wave:

$$\lambda_{\vec{k}\vec{k}'} = \int \varphi_{\vec{k}c}^*(\vec{r})\hat{H}_1 \varphi_{\vec{k}'h}(\vec{r})d\vec{r} \qquad (8)$$

Thus, the Hamiltonian (7) is time-dependent. This dependence can be eliminated by unitary transformation [16] and time-independent zones can be obtained:

$$\varepsilon_{1,2}(\vec{k}) = \pm\frac{\alpha_0 k^2}{4} + \sqrt{\left(\xi - \frac{\hbar\Omega}{2}\right)^2 + |\lambda|^2}, \qquad (9)$$

where

$$\xi = \frac{\alpha_1 k^2}{2} + \frac{E_g}{2}, \quad \alpha_0 = \frac{\hbar^2}{2}\left(\frac{1}{m_c} - \frac{1}{m_h}\right), \quad \alpha_1 = \frac{\hbar^2}{2a^2}\left(\frac{1}{m_c} + \frac{1}{m_h}\right).$$

Equation (9) describes zones of quasiparticles of the electron-hole pair type with the reference point $-\dfrac{\hbar\Omega - E_g}{2}$.

3. THE EFFECT OF ELECTROMAGNETIC IRRADIATION OF A QUASI-2-DIMENSIONAL SEMICONDUCTOR

In [16], changes in the electronic spectrum (9) caused by the resonant interaction were considered. The condition of such interaction is

$$0 < \hbar\Omega - E_g << E_g \qquad (10)$$

Taking into account the uniqueness of the quasi-2D crystal, we will consider a similar problem for it. In fact, any quasi-2D crystal is three-dimensional. In the XOY planes of the isotropic layers with real filling of the zone, the electronic spectrum can be with satisfactory accuracy represented by a 2D parabolic law with effective mass m_c, m_v:

$$E_{IIch} \equiv \frac{E_g}{2} + \frac{\hbar^2 k_{II}^2}{2m_{ch}} \qquad (11)$$

where $\vec{k}_{II}(k_x, k_y)$ is the 2D-quasi-momentum.

Along the normal to the layers, with the increasing of filling of the zones, deviations from parabolicity in the dispersion law become quite noticeable. Then, in the strong coupling approximation, taking into account the electronic overlap between adjacent layers $\beta_{ch}^{vv+1} = \beta$
(v is the layer's number), the longitudinal dispersion law takes the following form:

$$E_{ch}(k_z) = \beta_{ch}\left[1 - \cos(k_z)\right] \qquad (12)$$

Thus, the width of the allowed zone is equal to $2^{\beta c}$ and $2^{\beta h}$. In the case of a single zone model the transition of the Fermi level through such a value causes the Lifshitz transition, in which the closed isoenergy zone turns into an open one.

The total dispersion law is the sum of (11) and (12). For example, Fig. 2 shows it for the conduction zone. As the zones are filled, the dispersion laws along the normal to the layers become noticeably different from the parabolic ones.

Let us analyse a reconstructed spectrum of a quasi-2D crystal caused by electromagnetic radiation. Here we can use (9) taking into account the following renormalised parameters in it:

$$\frac{\alpha_0 k^2}{2} \rightarrow \frac{\alpha_0 k_{II}^2}{2} + \beta_0 \cdot \left(1 - \cos(k_z)\right), \qquad (13)$$

$$\frac{\alpha_1 k^2}{2} \rightarrow \frac{\alpha_1 k_{II}^2}{2} + \beta_1 \cdot \left(1 - \cos(k_z)\right) \qquad (14)$$

where $\beta_0 \rightarrow (\beta_c - \beta_v), \quad \beta_1 \rightarrow (\beta_c + \beta_v)$.

Let us consider the higher zone (9) taking into account such redefinitions. From the condition of the extremum $\dfrac{\partial \varepsilon_1(\vec{k})}{\partial k_s} = 0$, we obtain the equation:

$$k_s\left(\alpha_0 + \frac{\alpha_1 \xi}{\sqrt{\left(\xi - \frac{\hbar\Omega}{2}\right)^2 + |\lambda|^2}}\right) = 0, \quad (s = x, y) \qquad (15)$$

and from the condition of the extremum $\dfrac{\partial \varepsilon_1(\vec{k})}{\partial k_z} = 0$ we obtain the equation:

$$\sin(k_z)\left(\beta_0 + \frac{\beta_1 \xi}{\sqrt{\left(\xi - \frac{\hbar\Omega}{2}\right)^2 + |\lambda|^2}}\right) = 0 \qquad (16)$$

From (9), (10), it follows that there are possible the following extremum points:

a) $\vec{k} = (0,0,0)$ which coincides with a extremum point of the dispersion law of an non-irradiated $\varepsilon_c(\vec{k})$ of semiconductor, i.e. electromagnetic irradiation does not change its form.

b) $k_z = 0$ and k_x, k_y are thes solutions of the equation

$$\alpha_0 + \frac{\alpha_1 \xi}{\sqrt{\left(\xi - \frac{\hbar\Omega}{2}\right)^2 + |\lambda|^2}} = 0 \qquad (17)$$

After transformations this equation takes the following form:

$$\frac{\hbar^2}{2}\frac{m_c + m_h}{m_c m_h}k_s^2 = \hbar\Omega - E_g - \frac{\lambda(m_h - m_c)}{\sqrt{m_h m_c}}, \quad (s = x, y) \quad (18)$$

c) $k_x = k_y = 0$ and k_z is determined according to equation (10), which can be reduced to the form:

$$\cos(k_z) = 1 - \frac{\hbar\Omega - E_g}{\beta_c + \beta_h} + \frac{\lambda(\beta_c - \beta_h)}{\sqrt{\beta_c \beta_h}} \qquad (19)$$

From the condition (5) and the equation (11), it follows that $k_x, \ k_y \neq 0$ is possible only when

$$E_g + \frac{m_h - m_c}{\sqrt{m_h m_c}}\lambda \leq \hbar\Omega \leq E_g + \frac{m_h - m_c}{\sqrt{m_h m_c}}\lambda + \frac{\hbar^2 \pi^2}{2}\frac{m_h + m_c}{m_h m_c} \quad (20)$$

(In (13), there was assumed the parabolicity of the

zone in space $(k_x k_y)$ over the entire Brillouin zone).

Similarly, from equation (19) it follows that $k_z \neq 0$ in the Brillouin zone is possible only when

$$E_g + \frac{(\beta_c - \beta_h)}{\sqrt{\beta_c \beta_h}} \lambda \leq \hbar \Omega \leq E_g + \frac{(\beta_c - \beta_h)}{\sqrt{\beta_c \beta_h}} \lambda + 2 \cdot (\beta_c + \beta_h) \quad (21)$$

We introduce the notations: $E_{\min} = E_g + \frac{m_h - m_c}{\sqrt{m_h m_c}} \lambda$,

$E_{\max} = E_{\min} + \frac{m_h + m_c}{m_h m_c} \left(\frac{\pi}{a}\right)^2$, $\widetilde{E}_{\min} = E_g + \frac{\beta_c - \beta_h}{\sqrt{\beta_c \beta_h}} \lambda$,

$\widetilde{E}_{\max} = \widetilde{E}_{\min} + 2(\beta_c + \beta_h)$. Since the width of the allowed zones in space (k_x, k_y) is greater than that along k_z, then $E_{\max} > \widetilde{E}_{\max}$.

Depending on the proportions among $m_h, m_c, \beta_c, \beta_h$, there are the possible frequencies Ω of resonant electromagnetic irradiation at which minimuma of the zones are achived at $\vec{k} \neq (0,0,0)$, i.e. the shape of the spectra of the non-irradiated sample is changed. There are three specific ranges of change of Ω:

1) $\frac{\beta_c - \beta_h}{\sqrt{\beta_c \beta_h}} < \frac{m_h - m_c}{\sqrt{m_h m_c}}$: for $\widetilde{E}_{\max} < E_{\min}$.

Here are the frequency Ω ranges (their geometric representation is given below) in which the shape of the electronic spectrum is changed:

$\hbar\Omega$	$\left[\widetilde{E}_{\min}, \widetilde{E}_{\max}\right]$	$\left[\widetilde{E}_{\max}, E_{\min}\right]$	$\left[E_{\min}, E_{\max}\right]$
extremum points	$(0,0,k_z)$	–	$(k_x,0,0)$ $(0,k_y,0)$
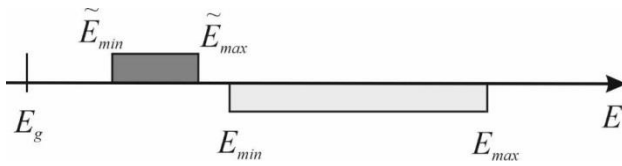
2) $\frac{\beta_c - \beta_h}{\sqrt{\beta_c \beta_h}} < \frac{m_h - m_c}{\sqrt{m_h m_c}}$ for $\widetilde{E}_{\min} < E_{\min} < \widetilde{E}_{\max}$.

Here are the frequency Ω ranges (their geometric representation is given below) in which the shape of the electronic spectrum is changed:

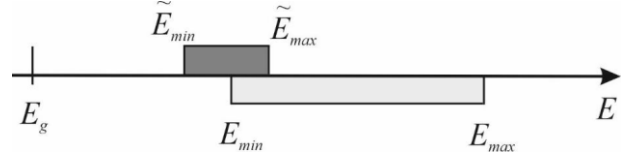| $\hbar\Omega$	$\left[\widetilde{E}_{\min}, E_{\min}\right]$	$\left[E_{\min}, \widetilde{E}_{\max}\right]$	$\left[\widetilde{E}_{\max}, E_{\max}\right]$
extremum points	$(0,0,k_z)$	$(0,0,k_z)$, $(k_x,0,0)$ $(0,k_y,0)$	$(k_x,0,0)$, $(0,k_y,0)$

3) $\frac{\beta_c - \beta_h}{\sqrt{\beta_c \beta_h}} > \frac{m_h - m_c}{\sqrt{m_h m_c}}$ for $\widetilde{E}_{\min} < E_{\min}$.

Here are the frequency Ω ranges (their geometric representation is given below) in which the shape of the electronic spectrum is changed:
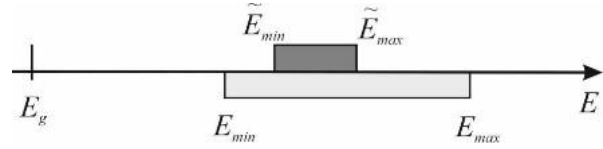
$\hbar\Omega$	$\left[\widetilde{E}_{\min}, E_{\min}\right]$	$\left[\widetilde{E}_{\min}, \widetilde{E}_{\max}\right]$	$\left[\widetilde{E}_{\max}, E_{\max}\right]$
extremum points	$(k_x,0,0)$, $(0,k_y,0)$	$(0,0,k_z)$, $(k_x,0,0)$, $(0,k_y,0)$	$(k_x,0,0)$, $(0,k_y,0)$

A similar analysis can be carried out for the lower zone in (9) and obtain similar conclusions to those obtained above.

It should be noted once again that quasi-2D crystals are sharply anisotropic crystals, which is confirmed, in particular, by their mechanical characteristics [17, 18]. Table 1 shows the elastic constants along the normal to the layers (C_{33}), in the layers (C_{11}), and the degree of anisotropy C_{33}/C_{11} for graphite and for some well-known layered crystals such as GaS, GaSe, InSe.

Table 1 – Elastic constants of the quasi-2D crystals

	C (graphite)	GaS	GaSe	InSe
$C_{11}, 10^{10}\,\text{Pa}$	106	15.7	10.3	7.3
$C_{33}, 10^{10}\,\text{Pa}$	3.7	3.6	3.4	3.6
C_{11}/C_{33}	28.6	4.4	3.0	2.0

Table 2 shows the pressure dependence of the elastic moduli of the same crystals at $T = 300$ K.

Table 2 – Baric dependences of the quasi-2D crystal elastic modulus

		C (graphite)	GaS	GaSe	InSe
$\frac{1}{C_{kk}}\frac{\partial C_{kk}}{\partial P}$, $10^{-11}\,\text{Pa}^{-1}$	C_{11}	4	8	8	11
	C_{33}	26	63	56	50
	C_{33}/C_{11}	5.5	7.9	7.0	4.5

Important conclusions can be drawn from these tables. From Table 1 it follows that:

a) the elastic moduli C_{11} in the plane of the layers of these crystals are significantly higher (30 times higher than those for graphite and on average 3 times

higher for other crystals);

b) the modulus C_{33} normal to the layers of these crystals are almost of the same values, while the moduli C_{11} in the plane of the layers are different for different crystals. Thus, the ratio C_{11}/C_{33} determines the degree of anisotropy of the quasi-2D crystals.

Table 2 shows that the baric dependences $\partial C_{ii|}/\partial P$ of the elastic constants in the same quasi-2D crystals for C_{33} are much higher than those for C_{11}. This fact allows us to neglect the effect of pressure in the domains of ionic-covalent interaction (in the layers) in the zero approximation and consider the pressure as a factor of the change of van der Waals gaps, and hence with a change in the overlap integral. Thus, the relations between $\dfrac{\beta_c - \beta_h}{\sqrt{\beta_c \beta_h}}$ and $\dfrac{m_h - m_c}{\sqrt{m_c m_h}}$, which determine the emergence of the cases 1-3, depend only on β_c and β_h.

Similar conclusions about the role of β_c and β_h in the emergence of the cases 1-3 are hold good for the intercalation of layered crystals. There are a number of external factors that can significantly change of β_c, β_h, in particular, intercalation. It should be noted that intercalation is a unique phenomenon of reversible insertion of foreign atoms into van der Waals gaps. Among the many studies of intercalation that remain relevant today [1, 19], we will turn to those that are interesting from the point of view of our consideration. In the 70s of the last century, interest in layered crystals as materials with possible superconductivity based on the exciton mechanism was revived [20]. Among the studies of this phenomenon, the work [21] is particularly important for us. In it superconductivity in 50 created by the authors intercalated TMD TaS_2 and $NbSe_2$ were studied. Organic and inorganic molecules were chosen as intercalant. In all the cases, the van der Waals gap increased to a different extent depending on the nature of the intercalant. Thus, during intercalation of $2H-TaS_2$ by octadecylamine, the gap value increased to 50 Å (in the pure $2H-TaS_2$ the gap value is ~ 3 Å)

Thus, intercalation by changing the geometrical dimensions of the van der Waals gaps leads to a change in β_c, β_h. It should be born in mind not only about the geometric manifestation of intercalation, but also about the nature of intercalated atoms, which themselves can change the the van der Waals interaction

4. CONCLUSION

The obtained results indicate that by changing the resonant frequency Ω it is possible to achieve extrema of spectrum of the quasi-2D semiconductor beyond the point $\vec{k} = (0,0,0)$, namely, only at the points $(0,0,k_z)$ or $(k_x,0,0)$, $(0,k_y,0)$ and simultaneously at these points. The ranges in which such points may occur depend on the proptions among the parameters β_c, β_h, m_h, m_c. In any case, the range of change of Ω where the extremum point $(0,0,k_z)$ occurs is much less than that in the case of $(k_x,0,0)$, $(0,k_y,0)$

There is an alternative approach possible: with a fixed frequency Ω, the analogous changes in the shape of the spectrum can be achieved by changing the overlap integrals β_c, β_h in the crystal by means to intercalation, axial or hydrostatic compression.

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Електронний спектр квазі-2D напівпровідника в сильному електромагнітному полі

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Розглянуто зміну форми спектрів квазі-2D напівпровідника під дією резонансного опромінення з частотою Ω, яка визначається умовою $0 \leq \hbar \cdot \Omega - E_g \ll E_g$. Дослідження показали три типи таких змін.

Їх проаналізовано у двох випадках: а) залежно від Ω при фіксованих параметрах зон квазі-2D напівпровідника (ефективних мас m_c, m_h, електронів і дірок у площині шарів, інтегралів перекриття β_c, β_h) і б) навпаки, при зміні параметрів зон при фіксованих Ω. Вказано на вирішальну роль β_c, β_h у прояві того чи іншого з трьох типів зміни форми спектра.

Ключові слова: Квазі-2D напівпровідник, Електронний спектр, Резонансне електромагнітне опромінення.