PHYSICS AND CHEMISTRY OF SOLID STATE
V. 22, No. 4 (2021) pp. 786-791
Section: Physics

DOI: 10.15330/pcss.22.4.786-791

Vasyl Stefanyk Precarpathian National University

ФІЗИКА І ХІМІЯ ТВЕРДОГО ТІЛА
Т. 22, № 4 (2021) С. 786-791
Фізико-математичні науки

PACS: 78.20.Ci, 62.23.Eg, 78.67.-n ISSN 1729-4428

B.A. Lukiyanets, D.V. Matulka

Optical properties of semiconductor “anisotropic” quantum dot

Lviv Polytechnic National University, Lviv, Ukraine, bohdan.a.lukiyanets@lpnu.ua, dariya.v.matulka@lpnu.ua

The optical properties of an “anisotropic” semiconductor nanodot – a nanoscale object in the form of a rectangular parallelepiped - with sides $a \neq b \neq c$ are considered. Such dimensions are closely related to the values of the effective masses of the electron. The analysis of the spectral dependence of the absorption coefficient $\alpha(\omega)$ under different degrees of "anisotropy" and under different polarizations of the electromagnetic wave is car ried out. The cases of the most intense optical transitions, i.e. between electronic states separated by the Fermi level, are analyzed. The obtained results indicate that 1) $\alpha(\omega)$ is of line structure, and 2) the positions of the peaks of $\alpha(\omega)$ in identical optical transitions in the isotropic nanodot and in the “anisotropic” ones coincide qualitatively. However, different masses in the “anisotropic” nanodot lead to a shift to the left or right of the peaks relative to identical peaks in the isotropic nanodot with simultaneous splitting of its degenerate peaks. Such shifts and their magnitudes are determined both by the degree of anisotropy (i.e. by the ratio between the effective masses), and by the polarization of light. It is pointed out that modern achievements in the creation of ordered semiconductor materials with nanoobjects of different shapes and sizes in nanostructures allows us to consider polarized electromagnetic wave as an effective factor in achieving the desired physical characteristics.

Key words: quantum dot, anisotropy, polarized light, absorption coefficient.

Received 15 September 2021; Accepted 20 December 2021.

Introduction

Nanoobjects are extreme objects of complex researches in modern science and technology. The reason for this is their unique physical properties caused by the phenomena of dimensional quantization, high manufacturability. Even nowadays, such objects are of wide practical application in electronics, photonics, medicine, agriculture, and others. Their potential is far from exhausted. Therefore, further progress is possible only due to finding the conditions for optimal use of nanoobjects, due to deep understanding of the processes in them, due to the search for new effects. The results of optical studies of objects are useful for such searches, and therefore optical studies are often used to study the physical properties of nanoobjects of different sizes, shapes, semiconductor or metallic nature, etc. In particular, the study of the effects of the shapes of nanoobjects and those of their sizes on their physical characteristics are still relevant. During almost 20 last years, intensive studies of such effects were focused on the optical characteristics of two-dimensional structures - QW and superlattices [1,2].

The QW-structure of GaAs-AlGaAs is, in fact, a bulk structure in which a thin layer of GaAs semiconductor is positioned between two AlGaAs-layers. The GaAs-AlGaAs interface is smooth since the corresponding GaAs and AlGaAs lattice constants are almost equal.

The AlGaAs band gap is higher than that of GaAs, and therefore the electron is confined within the GaAs layer. From this viewpoint such a structure can be considered as 2D one. In general, the GaAs-AlGaAs electron structure consists of discrete levels caused by nanodimensionality of the GaAs-layer, each of which contains its 2D zone of electron behaviour in the plane of the layers. As for the superlattices, they represent 3D structures of periodically reiterating QW – structures.

Therefore, instead of discrete electron states along the quantization axis, the electron states, due to electron tunneling, will be thin allowed zones.

In [1,2], change of optical characteristics with change of layer thickness, and in [3] those of with change of sizes of metal nanoparticles (diameter D = 10–40 nm) or of their forms in different crystal matrices are analyzed.

Quantum dots (0D systems) occupy a special place among nanoobjects due to their extraordinary properties, high controllability in their manufacture [4] (in particular, today the use of the nanodots can be considered as an easy way to obtain photovoltaic devices with high power conversion efficiency (see [5]). This is the reason of widespread practical use of the nanodots. Optical research in them was the subject of a number of papers (see [6]). Below we analyze the optical properties of an “anisotropic” semiconductor quantum dot under the action of polarized light.

I. Model. Calculation of Absorption Coefficient

Consider such optical studies to establish the manifestations of the shapes and the sizes of nanoobjects on their physical characteristics. We analyze the fundamental characteristic of optical research – the absorption coefficient $\alpha(\omega)$ in a semiconductor nanoobject. According to the theory of semiconductors [7], the formula for $\alpha(\omega)$ takes the form:

$$\alpha(\omega)=\frac{4\pi^2e^2}{ncm_0\omega\,\Omega}\sum_{if}\left|\vec{e}\vec{p}_{fi}\right|^2\left[f(E_i)-f(E_f)\right]\delta(E_f-E_i-\hbar\omega) \tag{1}$$

Here n is the refractive index of the medium, m_0 is the mass of electron, Ω is the volume of the crystal, ω is the angular frequency of the electromagnetic wave. Summation in (1) is to be made over all states in the optical transitions - from the initial state, E_i, in the valence band to the final state, E_f, in the conduction band. The multiplayer $\left[f(E_i)-f(E_f)\right]$ describes the occupation of the corresponding states ($f(E)$ is the Fermi-Dirac distribution; in the case of T = 0 K, the valence band is completely filled, and the conduction band is completely empty). The delta function $\delta(E_f-E_i-\hbar\omega)$ indicates that the states must be separated by an energy equal to the photon energy, according to the law of energy conservation.

The squared momentum matrix element $\left|\vec{e}\vec{p}_{fi}\right|^2$ describes the transition between states, caused by the $\vec{e}$ -polarized light. In 3D space (x, y, z) (or, otherwise, (1, 2, 3)) the momentum matrix element under the e_α -polarized light radiation is

$$p_{fis}=\iiint dx_\alpha dx_\beta dx_\gamma \psi_f^*\frac{\hbar}{i}\frac{\partial}{\partial x_s}\psi_i, \tag{2}$$

where ψ is the wave function of s-state; $s\in\alpha,\beta,\chi\in$ (1, 2, 3), but $\alpha\neq\beta\neq\chi$.

We choose a parabolic potential for both the conduction band and the valence band

$$V(x_\alpha x_\beta x_\chi)=V(x_\alpha)+V(x_\beta)+V(x_\chi)=\sum_{\alpha=1,2,3}\frac{m_0\omega_c}{2}x_\alpha^{\,2}\frac{m_\alpha}{m_0}, \tag{3}$$

where m_α is the effective mass of electron $m_{\alpha c}$ ($m_{\alpha v}$) in conduction band (of a hole in valence band)).

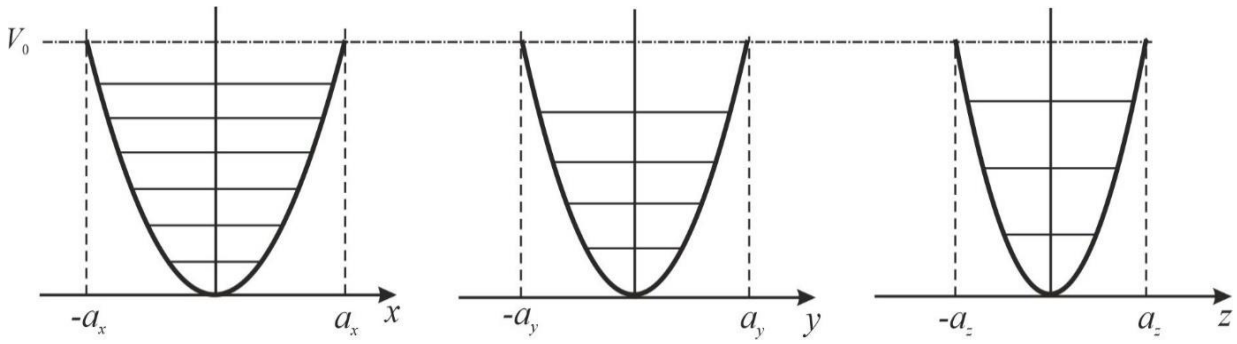

Fig.1. Potential energy in three crystallographic directions.

The energy states in it are [8]

$$E_{\nu_1\nu_2\nu_3} = \hbar\omega\sqrt{\frac{m_0}{m_1}}\left(\nu_1+\frac{1}{2}\right)+\hbar\omega\sqrt{\frac{m_0}{m_2}}\left(\nu_2+\frac{1}{2}\right)+\hbar\omega\sqrt{\frac{m_0}{m_3}}\left(\nu_3+\frac{1}{2}\right) \tag{4}$$

(here ν_α are quantum numbers, ν_α = 0, 1, 2, 3… ..) whose wave functions are the following:

$$\psi_{\nu_1\nu_2\nu_3}\left(\xi_1,\xi_2,\xi_3\right)=\prod_{\alpha=1,2,3}\frac{1}{\sqrt{2^{\nu_\alpha}\nu_\alpha!\sqrt{\pi}}}\exp(-\frac{\xi_\alpha^2}{2})H_{\nu_\alpha}\left(\xi_\alpha\right) \tag{5}$$

($H_{\nu_\alpha}\left(\xi_\alpha\right)$ is the Hermite polynomial; $\xi_\alpha = x\left(\frac{\hbar}{m_0\omega}\frac{m_0}{m_\alpha}\right)^{-1/2}$).

We apply the above mention results for the case of a nanoobject – "anisotropic" quantum dot - in the form of a rectangular parallelepiped with nanosized sides $2a\times 2b\times 2c$. The depth of the parabolic well V_0, its size and effective mass are related by the rationalship $a_\alpha = \sqrt{\frac{2V_0}{\omega_c^2}\frac{1}{m_\alpha}}$, i.e. the size of the well is larger for particles with smaller effective masses.

Let us estimate the probability of whereabout at the edge of the well of a ground state electron and that of the lowest excited state electron. For example, in a well of $V_0 \approx$ 1 eV (in this case the width of the well is 10 nm) such probability of whereabout of the ground state electron of the mass m_0, according to (5) is of $P_0\left(x=5\right)\equiv\left|\psi_0\right|^2 \sim 10^{-25}$. Thus, we can assume with high accuracy that the electron of the ground or weakly excited states is, surely, in the nanoobject.

The formula for squared momentum matrix element $\left|\vec{e}\vec{p}_{fi}\right|^2$ for the transition caused by the action of e_α-polarized light is of the form:

$$\left|p_{fi}\right|^2 = (\int dx_\alpha\psi^*_{cv_\alpha}\left(\xi_\alpha\right)\frac{\hbar}{i}\frac{\partial}{\partial x_\alpha}\psi_{vv'_\alpha}\left(\xi_\alpha\right)\int dx_\beta\psi^*_{cv_\beta}\left(\xi_\beta\right)\psi_{vv'_\beta}\left(\xi_\beta\right)\int dx_\chi\psi^*_{cv_\gamma}\left(\xi_\chi\right)\psi_{vv'_\chi}\left(\xi_\chi\right))\times$$
$$(\int dx_\alpha\psi^*_{cv_\alpha}\left(\xi_\alpha\right)\frac{\hbar}{i}\frac{\partial}{\partial x_\alpha}\psi_{vv'_\alpha}\left(\xi_\alpha\right)\int dx_\beta\psi^*_{cv_\beta}\left(\xi_\beta\right)\psi_{vv'_\beta}\left(\xi_\beta\right)\int dx_\chi\psi^*_{cv_\gamma}\left(\xi_\chi\right)\psi_{vv'_\chi}\left(\xi_\chi\right)) \tag{6}$$

The integration in the integrals of (6) must be performed over the volume of the nanoobject, but the above considerations indicate that for the ground and weakly excited states there exists a possibility of extending the integration with high accuracy to infinite space.

Analysis of the momentum matrix element in Eq (1) indicates that it is nonzero in the case when both $\nu_\beta+\nu'_\beta$ and $\nu_\gamma+\nu'_\gamma$ are even and at the same time $\nu_\alpha+\nu'_\alpha$ is odd. Since the most intense transitions are those the lowest electron states in the valence band to their states in the conduction band, we consider the transitions $\psi_{v\nu_1\nu_2\nu_3}\rightarrow\psi_{c\nu'_1\nu'_2\nu'_3}$ from the ground state of the hole (ν_1,ν_2,ν_3) to the lowest electron states (ν'_1,ν'_2,ν'_3), namely, under the polarization $\vec{e}=\left(e_x,0,0\right)$ we consider the following transitions: 1) $\left(0,0,0\right)\rightarrow\left(1,0,0\right)$, 2) $\left(0,1,0\right)\rightarrow\left(1,1,0\right)$, 3) $\left(0,0,1\right)\rightarrow\left(1,0,1\right)$, 4) $\left(0,1,1\right)\rightarrow\left(1,1,1\right)$.

For example, in the case of the e_α polarization

$$\left|p_{v000,c100}\right|^2 = \frac{16\hbar^2\left(m_{ac}m_{av}\right)}{\left(m_{ac}+m_{av}\right)^3\left(m_{\beta c}+m_{\beta v}\right)\left(m_{\gamma c}+m_{\gamma v}\right)} \tag{7}$$

(here and below the masses are written m_0 units).

In general case, the frequency of transition $\psi_{v\nu_1\nu_2\nu_3}\rightarrow\psi_{c\nu'_1\nu'_2\nu'_3}$ under the action of e_α-polarized electromagnetic wave is determined from the equation

$$\hbar\omega = E_g+\hbar\omega_c(\frac{1}{\sqrt{m_{ac}}}\left(\nu_\alpha+\frac{1}{2}\right)+\frac{1}{\sqrt{m_{av}}}\left(\nu'_\alpha+\frac{1}{2}\right)+\left(\frac{1}{\sqrt{m_{\beta c}}}+\frac{1}{\sqrt{m_{\beta v}}}\right)\left(\nu_\beta+\frac{1}{2}\right)+\left(\frac{1}{\sqrt{m_{\gamma c}}}+\frac{1}{\sqrt{m_{\gamma v}}}\right)\left(\nu_\gamma+\frac{1}{2}\right)) \tag{8}$$

Note that non-zero transitions (7) are in the case of $\nu_\alpha+\nu'_\alpha=(2n+1)$ (n=0, 1, 2....).

Let us analyze the most intense of absorption coefficients $\alpha(\omega)$ for the four above mentioned transitions in the case of equality of the effective masses of electrons and holes, i.e. $m_{sc}=m_{sv}=m_s\left(s=\alpha,\beta,\gamma\right)$.

From (7) it follows that

$$\left|p_{v000,c100}\right|^2 = \frac{\hbar^2}{2m_\alpha m_\beta m_\gamma},$$

and the transition 1) $(0,0,0)\to(1,0,0)$ due to the light polarization $\vec{e}=(e_x,0,0)$, according to (8), will occur when

$$\hbar\omega = E_g + \hbar\omega_c\left(\frac{2}{\sqrt{m_\alpha}}+\frac{1}{\sqrt{m_\beta}}+\frac{1}{\sqrt{m_\gamma}}\right)$$

Then the formula for absorption coefficient $\alpha(\omega)$ at such frequency takes the form:

$$\alpha(\omega) = const\frac{1}{2m_\alpha m_\beta m_\gamma}\frac{1}{E_g+\hbar\omega_c\left(\frac{2}{\sqrt{m_\alpha}}+\frac{1}{\sqrt{m_\beta}}+\frac{1}{\sqrt{m_\gamma}}\right)},$$

where $const=\frac{4\pi^2\hbar^2e^2}{ncm_0\Omega}$.

Similar considerations are valid for the $\vec{e}=(0,e_y,0)$ and $\vec{e}=(0,0,e_z)$ polarizations of light. Numerical calculations of $\alpha(\omega)$ in the case of the e_x-, e_y-, e_z- polarizations were performed in two nanodots under the assumption of equality of the effective masses of electrons and holes ($m_{sc}=m_{sv}=m_s\ (s=\alpha,\beta,\gamma)$). The effective masses in the nanodot 1 were $m_1=1.1$ $m_2=1.2$, $m_3=1.3$, and in the nanodot 2 - $m_1=0.9$, $m_2=1.0$, $m_3=1.1$ ($E_g=0.2\,eV$, $\hbar\omega_c=0.1\,eV$). Obtained and, for comparison, the analogical ones of the isotropic nanoobject (i.e. when $m_1=m_2=m_3=m_0$) are presented in Fig.2.

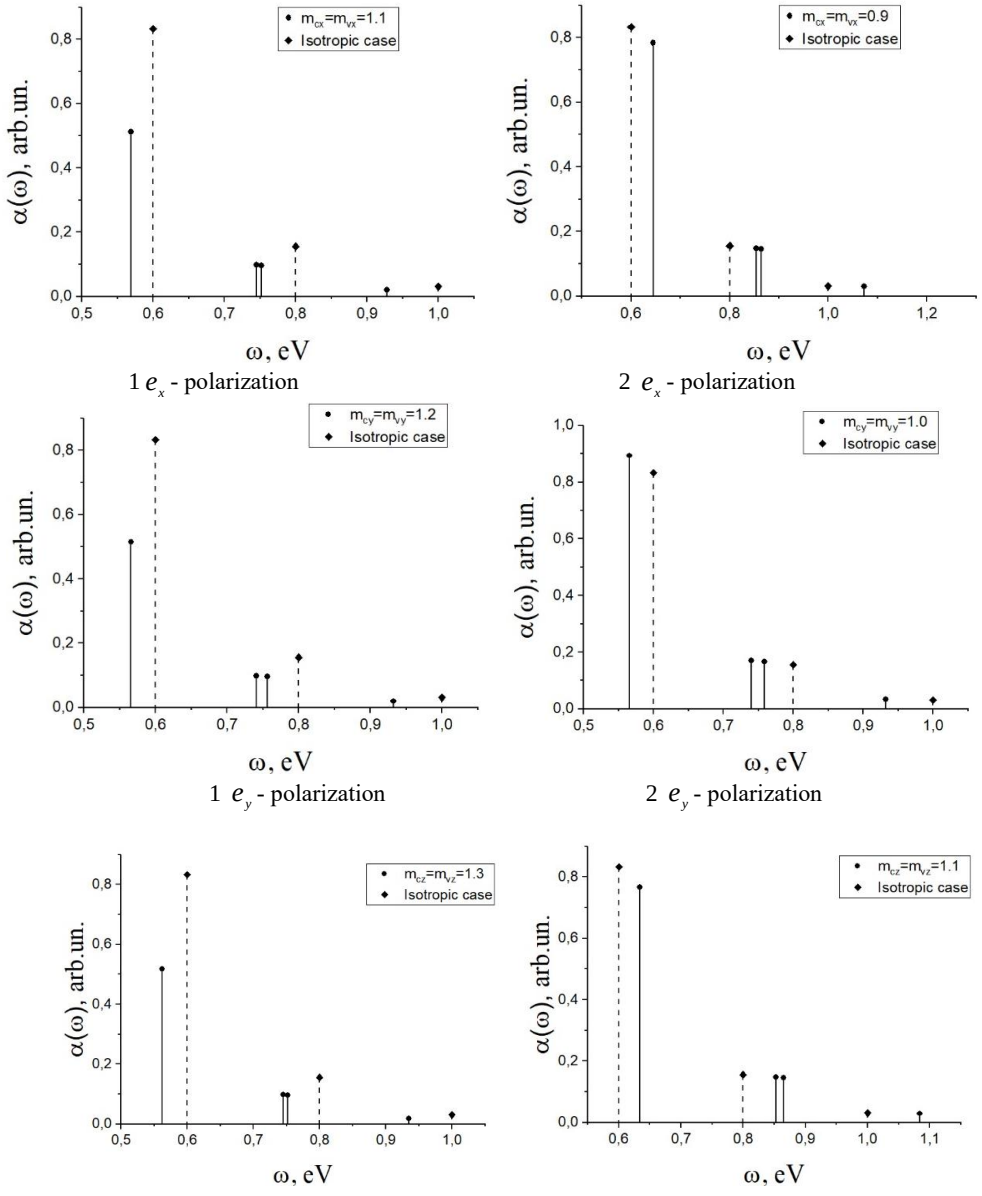

Fig. 2. Spectral polarization dependence $\alpha(\omega)$ (in *const* – units; $const=\frac{4\pi^2\hbar^2e^2}{ncm_0\Omega}$) in two nanodots whose effective masses are m_x = 1.1, m_y = 1.2, m_z = 1.3 for (1) and m_x = 0.9, m_y = 1.0, m_z = 1.1 for (2).

Discussion, conclusions

Fig.2 presents the results of calculations of the absorption coefficient $\alpha(\omega)$ in a semiconductor nanoobject in the form of a rectangular parallelepiped with a parabolic confinement for partial cases. However, their qualitative characteristics remain valid in other specific cases. In particular,

- in all cases, $\alpha(\omega)$ is of line structure, which is a consequence of the discrete spectrum of the carriers;
- the positions of the peaks of $\alpha(\omega)$ in identical transitions in the anisotropic nanodot shift to the left or to the right relative to their position in the isotropic nanodot. Such shifts and their magnitudes are determined both by the degree of anisotropy (i.e. by the ratio between the effective masses), and by the polarization of light.
- in an isotropic nanoobject, regardless of the polarization of light, $\alpha(\omega)$ takes the same value in the transitions $(0,1,0)\to(1,1,0)$, $(0,0,1)\to(1,0,1)$. In the case of an anisotropic nanoobject, such a doublet splits by a certain value which depends on the degree of anisotropy and of the polarization of light.

If we compare the electron states in the "anisotropic" quantum dot with those in nano-heterostructures, then in the first case they are strictly discrete, while in the second case discreteness is weakly blurred by continuous electron states in the plane of the layers. It is this difference that under certain conditions provides a clear experimental observation of the described results in "anisotropic" quantum dots. It is known that in real structures there is always blurring of levels. In addition to natural blurring, it largely depends on temperature, impurities, interaction with the environment. We considered low temperatures, without the presence of impurities. But there is another requirement to experimental observation in the case of an ensemble of "anisotropic" quantum dots. With their chaotic arrangement, with an equally probable orientation in space, the resulting polarization effect will be absent, because, in average, the ensemble has no a chosen direction. However, in recent years, purposeful creation of ordered nanostructures went on. While until recently such an ordering was observed for intercalates in graffiti [9], nowadays significant progress has been made in the technology of such nanostructures [10-14]. Review [15] presents the results of intensive research of ordered semiconductor materials in the form of nanowires, nanoparticles, nanocones, etc. in nanostructures. Non-ordinary properties, in particular, those of optical properties which can have wide practical application in photovoltaics have been revealed. The study of the angular dependence of the reflection coefficient on ordered nanosized islands of gold on sapphire substrates [16] revealed certain features of this dependence. The authors suggest that they are the result of surface plasmon-polariton excitation at the vacuum-gold interface.

Thus, such ordered nanostructures will be anisotropic, and therefore the polarized electromagnetic wave is an effective factor in achieving the desired physical characteristics

Lukiyanets B.A. – Doctor of Physical and Mathematical Sciences, Professor at Department of Applied Physics and Nanomaterials Science.
Matulka D.V. - Candidate of Technical Sciences, Associate Professor at Department of Applied Physics and Nanomaterials Science;

[1] E.Runge, Solid State Physics, 57, 149 (2003) https://doi.org/10.1016/S0081-1947(08)60180-0.
[2] R. Koole, E. Groeneveld, D. Vanmaekelbergh, A. Meijerink, C. M. Donegá, Size Effects on Semiconductor Nanoparticles. (Springer Verlag, 2014).
[3] S. Marhaba, Effect of Size, Shape and Environment on the Optical Response of Metallic Nanoparticles (2017); http://dx.doi.org/10.5772/intechopen.71574.
[4] Handbook of Nanophysics: Nanoparticles and Quantum Dots. Ed by K. D. Sattler. (CRC Press, 2011) https://doi.org/10.1201/9781420075458.
[5] F. Marangi, M. Lombardo, A. Villa, F. Scotognella. New Strategies for Solar Cells Beyond the Visible Spectral Range. https://arxiv.org/abs/2104.06657 [Submitted on 14 Apr 2021].
[6] E. Cottancin, M. Broyer, J. Lerme, M. Pellarin, Optical Properties of Metal Clusters and Nanoparticles. In: Nanoelectronics and Nanophotonics. Dots. Ed by Klaus D.Sattler. (CRC Press, 2010).
[7] J.H. Davies, The Physics of Low-dimensional Semiconductors: An Introduction (Cambridge Univ.Press, 1998).
[8] A.S. Davydov, Quantum mechanics (Pergamon Press, 1965).
[9] S.A. Safran, Solid State Physics, 40, 183 (1987), https://doi.org/10.1016/S0081-1947(08)60692-X.
[10] P. Mu, G. Zhou, C.-L. Chen, Nano-Structures & Nano-Objects 15, 153 (2018) https://doi.org/10.1016/j.nanoso.2017.09.010.
[11] J.T. Zhang, J. Ma, R.K. Kankala, Q. Yu, Shi-Bin Wang, A.Z. Chen, Appl. Bio Mater., 4(5), 4039 (2021) https://doi.org/10.1021/acsabm.1c00156 .
[12] O.V. Rudenko, A.I. Korobov, N.I. Odina, Method to form ordered nanostructures on substrate; Patent № 2421394 viewed 20.06.2011.
[13] V.A. Eremeyev, H. Altenbach, On the Eigenfrequencies of an Ordered System of Nanoobjects. in IUTAM Symposium on Modelling Nanomaterials and Nanosystems (eds.R. Pyrz and J.C. Rauhe). 123 (2008); https://doi.org/10.1007/978-1-4020-9557-3_13.

[14] Nano-objects of desire: Assembling ordered nanostructures in 3D Accessed on: Jan. 2020, [Online]. Available: https://www.sciencedaily.com/releases/2020/01/200113111042.htm.
[15] R. Yu, Q. Lin, S.F. Leung, Z. Fan. Nano Energy 1(1), 57 (2012). https://doi.org/10.1016/j.nanoen.2011.10.002.
[16] A.E. Muslimov, et al., Crystallography reports 57(3), 415 (2012); https://doi.org/10.1134/S1063774512030157.

Б.А. Лукіянець, Д.В. Матулка

Оптичні властивості “анізотропної” квантової точки

Національний університет "Львівська політехніка", Львів, Україна *bohdan.a.lukiyanets@lpnu.ua*, *dariya.v.matulka@lpnu.ua*

Розглянуто оптичні властивості “анізотропного” напівпровідникової наноточки – нанорозмірного об'єкта у вигляді прямокутного паралелепіпеда – зі сторонами $a \neq b \neq c$. Такі розміри тісно пов'язані зі значеннями ефективних мас електрона. Здійснено аналіз спектральної залежності коефіцієнта поглинання $\alpha(\omega)$ при різних ступенях “анізотропії” та при різних поляризаціях електромагнітної хвилі. Проаналізовано випадки найбільш інтенсивних оптичних переходів, тобто між електронними станами, розділеними рівнем Фермі. Отримані результати вказують на те, що 1) $\alpha(\omega)$ має лінійчасту структуру, а 2) положення піків $\alpha(\omega)$ однакових оптичних переходів ізотропних наноточках та в “анізотропних” якісно збігаються. Однак різні маси в "анізотропній" наноточці призводять до зсуву ліворуч або праворуч від піків відносно однакових піків в ізотропній наноточці з одночасним розщепленням його вироджених піків. Такі зсуви та їх величини визначаються як ступенем анізотропії (тобто співвідношенням між ефективними масами), так і поляризацією світла. Показано, що сучасні досягнення у створенні впорядкованих напівпровідникових матеріалів з нанооб’єктами різної форми та розміру в наноструктурах дозволяють розглядати поляризовану електромагнітну хвилю як ефективний фактор досягнення бажаних фізичних характеристик.

Ключові слова: квантова точка, анізотропія, поляризоване світло, коефіцієнт поглинання.