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THE INTERSUBBAND OPTICAL ABSORPTION COEFFICIENT OF THE QD WITH ACCEPTOR IMPURITY UNDER APPLIED ELECTRIC FIELD

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Abstract. In this work, a spherical quantum dot (QD) under the influence of an external electric field is investigated. A multi-band model of the valence band is applied. The influence of off-center acceptor impurity, electric field and QD size dispersion on the absorption coefficient during intersubband transitions between hole states has been analyzed. The results show that an electric field combined with an off-center impurity induces the appearance of two distinct absorption bands corresponding to different magnetic quantum numbers. The intensity of absorption bands depends on the direction and strength of the electric field, and significant differences are observed between fields of opposite polarity. It is important to note that there is a critical field strength that restores the degeneracy of the energy levels, narrowing the broadband absorption tail for systems with small or large dispersion sizes. This research aims to improve the understanding and optimization of the optical properties of nanomaterials.

Keywords: acceptor impurity, electric field, hole energy spectrum, multiband hole model, level splitting

КОЕФІЦІЄНТ МІЖПІДЗОННОГО ОПТИЧНОГО ПОГЛИНАННЯ КВАНТОВОЇ ТОЧКИ З АКЦЕПТОРНОЮ ДОМІШКОЮ В ПРИКЛАДЕНОМУ ЕЛЕКТРИЧНОМУ ПОЛІ

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Анотація. У роботі досліджено сферичну квантову точку (КТ) під впливом зовнішнього електричного поля. Застосовано багатозонну модель валентної зони. Проаналізовано вплив нецентральної акцепторної домішки, електричного поля та дисперсії розміру КТ на коефіцієнт поглинання при міжпідзонних переходах між дірковими станами. Результати показують, що електричне поле в поєднанні з нецентральною домішкою викликає появу двох чітких смуг поглинання, що відповідають різним магнітним квантовим числам. Інтенсивність смуг поглинання залежить від напрямку і напруженості електричного поля, причому значні відмінності

спостерігаються між полями протилежної полярності. Важливо відзначити, що існує критична напруженість поля, яка відновлює виродження енергетичних рівнів, звужуючи широкосмуговий хвіст поглинання для систем з малими або великими розмірами дисперсії. Це дослідження спрямоване на покращення розуміння та оптимізацію оптичних властивостей наноматеріалів.

Ключові слова: акцепторна домішка, електричне поле, енергетичний спектр дірок, багатозонна діркова модель, розщеплення рівнів

Introduction

The modern nanotechnologies make it possible to create various nanostructures with predefined properties. Among them, quantum dots (QDs) attract the most attention. These studies are of interest to both theorists and experimentalists because they have unique optical properties [1–3]. Various factors influence the optical properties of QDs, including the shape of the QDs, the material of the QDs and their surrounding environment, and the presence of impurities, both donor and acceptor types. The application of an electric field causes a shift of the energy levels of the QDs, which can be experimentally observed and used in the development of optoelectronic devices. This requires more detailed theoretical research.

Currently, many theoretical works are devoted to the calculation of linear and nonlinear optical properties of optoelectronic devices. This serves as a theoretical basis for the manufacture of devices with predetermined optical properties [4–6].

Most of the authors in their works investigate donor impurities. They are placed in the centers of QDs [7–9]. This allows to use a singleband model and simplify theoretical calculations. Similar studies were carried out for multilayer quantum dots in an electric field [10, 11]. It is shown that a change in the external field causes a change in the density of the probability of finding electrons in different shells. In the work [12], the energy spectrum, wave functions, and binding energies of an electron with a donor impurity ion located at the center of a multilayer spherical QD consisting of a core and two spherical shells were investigated. It was shown that changing the parameters significantly affects both the energy spectrum and the absorption coefficient. The works [7–16] are shown the optical properties depend on the position of the donor hydrogen impurity and external fields. It has been demonstrated that shifting the impurity from the center of a spherical quantum dot also causes the splitting of electronic levels (for degenerate states). These impurity shifts cause a shift in the absorption band to the low-energy region [13].

To study hole and acceptor states in spherical quantum dots, more complex multiband models in the effective mass approximation should be used. For example, the Luttinger model [17, 18] in the spherical approximation [19]. The study [20] examined hole and exciton states in an external electric field using the multiband effective mass model. It was demonstrated that the ground hole state in a spherical QD split under the influence of an electric field, a phenomenon not observed in the single-band model where the ground state remains unsplit. In addition, the ground state of the impurity splits when the acceptor impurity is at a distance D from the center of the spherical quantum dot [21]. This splitting significantly influences various properties, particularly the optical characteristics of QDs. Hence, in spherical QDs, both an applied electric field and an off-center acceptor impurity result in the splitting of hole energy levels.

The paper [22] presents a model for calculating the energy levels of holes in a spherical quantum dot with a non-central impurity in an external electric field. A non-central acceptor impurity causes splitting of levels due to the violation of spherical symmetry. However, if the electric field is applied in the direction opposite to the displacement of the impurity from the center, there is such a value of the field at which the splitting of the levels disappears (degenerate, as in the case of a central impurity or its absence). This means that changing the magnitude and direction of the applied electric field can reduce

the level splitting, which will reduce the additional absorption bands and the width of the long-wavelength absorption or luminescence peak.

Measuring the interband absorption coefficient of light allows for a detailed study of the optical properties of quantum dot. The optical characteristics of many devices created based on bulk and nanoscale semiconductors significantly depend on impurities. The aim of our work is to investigate the effect of off-center acceptor impurity, electric field and QD size dispersion on the absorption coefficient in intersubband transitions between hole states. This research is aimed at improving the understanding and optimization of the optical properties of nanomaterials.

Hole energy spectrum

Let's consider a heterostructure with a spherical semiconductor quantum dot of radius a. We choose structures where the band gap width and spin-orbit interaction are big enough. The heterosystem *GaAs/AlAs* meets these requirements. Therefore, for calculating hole states, we will use a so-called four-zone approximation [17–20], neglecting the corrugation of isoenergetic surfaces in k-space

$$\begin{aligned}\hat{\mathbf{H}} &= \frac{1}{2}\left(\gamma_1+\frac{5}{2}\gamma\right)\hat{\mathbf{p}}^2-\gamma\left(\vec{\hat{\mathbf{p}}}\vec{\hat{\mathbf{J}}}\right)^2+U(r)+V_{el}(\vec{r})+V\left(\vec{r},\vec{D}\right)= \\ &= \hat{\mathbf{H}}^{(0)}+V_{el}(\vec{r})+V\left(\vec{r},\vec{D}\right),\end{aligned} \tag{1}$$

where $\vec{\hat{\mathbf{p}}}$ is the momentum operator, $\vec{\hat{\mathbf{J}}}$ is the spin operator (j = 3/2); γ_1, γ are the Luttinger parameters which are connected with the heavy and light hole effective masses

$$m_{hh}=\frac{m_0}{\gamma_1-2\gamma},\ m_{lh}=\frac{m_0}{\gamma_1+2\gamma}. \tag{2}$$

The energy axis in the valence band is directed 'downwards', meaning the valence band maximum transitions to a minimum. The confinement potential for the hole was chosen as a spherical rectangular potential well.

$$U(r)=\begin{cases}0, & r\le a,\\ U_0, & r>a.\end{cases} \tag{3}$$

Potential energy of the hole in the external electric field F_{el} has form

$$V_{el}(\vec{r})=-\vec{d}\cdot\vec{F}_{el}=-e\vec{r}\vec{F}_{el}\ , \tag{4}$$

where $\vec{d}$ is dipole momentum, e is elementary charge. We consider electric field direction along z axis. Interaction of a hole with an acceptor impurity ion is expressed by

$$V\left(\vec{r},\vec{D}\right)=-\frac{Ze^2}{\varepsilon\left|\vec{r}-\vec{D}\right|} \tag{5}$$

where ε is dielectric permittivity of QD, Z=1 or Z=0 (if there is no impurity in the QD). We consider cases where the impurity can be located on the z axis and electric field is directed along z axis.

To determine the energy spectrum of a hole, we expressed the hole's wave function ψ as a series expansion in terms of the system of functions ψ_i^0 that are solutions to the Schrödinger equation with the Hamiltonian $\hat{\mathbf{H}}^{(0)}$ (the Hamiltonian that describes a hole in a QD without impurity and electric field):

$$\psi=\sum_i c_i\psi_i^0\ . \tag{6}$$

The wave function can be expressed as a product of the eigenfunctions of the total momentum $\vec{\hat{\mathbf{F}}}=\vec{\hat{\mathbf{L}}}+\vec{\hat{\mathbf{J}}}$ and the radial functions [23, 24]:

$$\psi^{(0)}_{f,M}(r,\theta,\varphi)=\sqrt{2f+1}\sum_{l=f-j}^{f+j}(-1)^{l-j+M}\,R_l^f(r)\sum_{m_l}\sum_{m_j}\begin{pmatrix} l & j & f\\ m_l & m_j & -M\end{pmatrix}Y_l^{m_l}(\theta,\varphi)\,\chi_{m_j}, \qquad (7)$$

where $\hbar^2 f(f+1), \hbar^2 l(l+1), \hbar M, \hbar m, \hbar m_j$ are the eigenvalues of operators $\hat{\mathbf{F}}^2, \hat{\mathbf{L}}^2, \left(\hat{\mathbf{L}}_z+\hat{\mathbf{J}}_z\right)$, $\hat{\mathbf{L}}_z, \hat{\mathbf{J}}_z$ respectively; χ_{m_j} are the spin functions, Y_l^m are the spherical harmonic functions, which are the eigenfunctions of $\hat{\mathbf{L}}^2$, $\begin{pmatrix} l & j & f\\ m_l & m_j & -M\end{pmatrix}$ are the 3-j symbols.

For the convenience of writing formulas, we will further use the atomic sys system of Hartree units ($\hbar$= 1, m_e = 1, e = 1).

In the case F_{el} = 0 and Z = 0 we have obtained the radial equation like in [23, 24] for $f \geq 3/2$

$$\frac{1}{2}\gamma_1\begin{pmatrix} -(1+\mu C_1)\hat{\Delta}_l & \mu C_2\hat{\mathbf{B}}^-_{l+1}\hat{\mathbf{B}}^-_{l+2}\\ \mu C_2\hat{\mathbf{B}}^+_{l+1}\hat{\mathbf{B}}^+_{l} & -(1+\mu C_3)\hat{\Delta}_{l+2}\end{pmatrix}\begin{pmatrix} R_l^f(r)\\ R_{l+2}^f(r)\end{pmatrix}+U(r)\begin{pmatrix} R_l^f(r)\\ R_{l+2}^f(r)\end{pmatrix}=E^{(0)}\begin{pmatrix} R_l^f(r)\\ R_{l+2}^f(r)\end{pmatrix} \qquad (8)$$

with coefficients

$$\begin{aligned} C_1 &= C_1(f,l)=\sqrt{5}(-1)^{3/2+l+f}\begin{Bmatrix} l & l & 2\\ 3/2 & 3/2 & f\end{Bmatrix}\sqrt{\frac{2l(2l+1)(2l+2)}{(2l+3)(2l-1)}}\\ C_2 &= C_2(f,l)=\sqrt{30}(-1)^{3/2+l+f}\begin{Bmatrix} l+2 & l & 2\\ 3/2 & 3/2 & f\end{Bmatrix}\sqrt{\frac{(l+1)(l+2)}{2l+3}}\\ C_3 &= -C_1, \quad (C_1)^2+(C_2)^2=1, \qquad C_2/\mu>0, \qquad \mu=\frac{2\gamma}{\gamma_1}\end{aligned} \qquad (9)$$

and operators

$$\hat{\mathbf{B}}^+_l=-\frac{\partial}{\partial r}+\frac{l}{r}, \quad \hat{\mathbf{B}}^-_l=\frac{\partial}{\partial r}+\frac{l+1}{r}, \quad \hat{\Delta}_l=\frac{\partial^2}{\partial r^2}+\frac{2}{r}\frac{\partial}{\partial r}-\frac{l(l+1)}{r^2}. \qquad (10)$$

If f=1/2, the radial equation takes the form

$$-\frac{1}{2}\gamma_1(1+\mu)\left(\hat{\Delta}_l R_l^{1/2}(r)\right)+U(r)R_l^{1/2}(r)=ER_l^{1/2}(r). \qquad (11)$$

If $r \leq a$, the solution of (8) can be expressed by the sum of the Bessel functions of the first kind

$$\begin{aligned} R_l^{f(I)}(r) &= A_1^{(I)}\frac{(C_1-1)\mathrm{J}_{l+1/2}\left(k_{hh}^{(I)}r\right)}{C_2\sqrt{r}}+A_2^{(I)}\frac{(C_1+1)\mathrm{J}_{l+1/2}\left(k_{lh}^{(I)}r\right)}{C_2\sqrt{r}},\\ R_{l+2}^{f\,(I)}(r) &= A_1^{(I)}\frac{\mathrm{J}_{l+5/2}\left(k_{hh}^{(I)}r\right)}{\sqrt{r}}+A_2^{(I)}\frac{\mathrm{J}_{l+5/2}\left(k_{lh}^{(I)}r\right)}{\sqrt{r}},\end{aligned} \qquad (12)$$

where $k_{hh}^{(I)}=\sqrt{2m_{hh}^{(I)}E}$, $k_{lh}^{(I)}=\sqrt{2m_{lh}^{(I)}E}$.

If $r > a$, the solution can be expressed in terms of the modified Bessel functions of the first and second kind

$$R_l^{f\,(II)}(r) = A_1^{(II)} \frac{\left(-C_1+1\right)\mathrm{K}_{l+1/2}\left(k_{hh}^{(II)} r\right)}{C_2\sqrt{r}} + A_2^{(II)} \frac{-\left(C_1+1\right)\mathrm{K}_{l+1/2}\left(k_{lh}^{(II)} r\right)}{C_2\sqrt{r}},$$

$$R_{l+2}^{f\,(II)}(r) = A_1^{(II)} \frac{\mathrm{K}_{l+5/2}\left(k_{hh}^{(II)} r\right)}{\sqrt{r}} + A_2^{(II)} \frac{\mathrm{K}_{l+5/2}\left(k_{lh}^{(II)} r\right)}{\sqrt{r}}, \qquad (13)$$

where $k_{hh}^{(II)} = \sqrt{2m_{hh}^{(II)}(U_0 - E)}$, $k_{lh}^{(II)} = \sqrt{2m_{lh}^{(II)}\left(U_0 - E\right)}$.

Using the boundary conditions and normalized condition, the dispersion equation has been received and energy spectrum of hole has been defined in the case of $F_{el} = 0$ and Z = 0. Quantum numbers l and f are connected by the rule of adding angular moments $\hat{\vec{\mathbf{F}}} = \hat{\vec{\mathbf{L}}} + \hat{\vec{\mathbf{J}}}$. For fixed l and f solution of the dispersion equation are numbered by n. Therefore the wave function depend on one mere quantum number n:

$$\psi_{f,M}^{(0)}(r,\theta,\varphi) \equiv \psi_{f,M;n}^{(0)}(r,\theta,\varphi).$$

In this way, the energy of the holes was determined. The calculation shows than hole energy levels split by total magnetic quantum number $|M| = 1/2,\ 3/2$ when there is the off-central impurity or/and the electric field is applied (for example, see Fig.1). Their combined influence

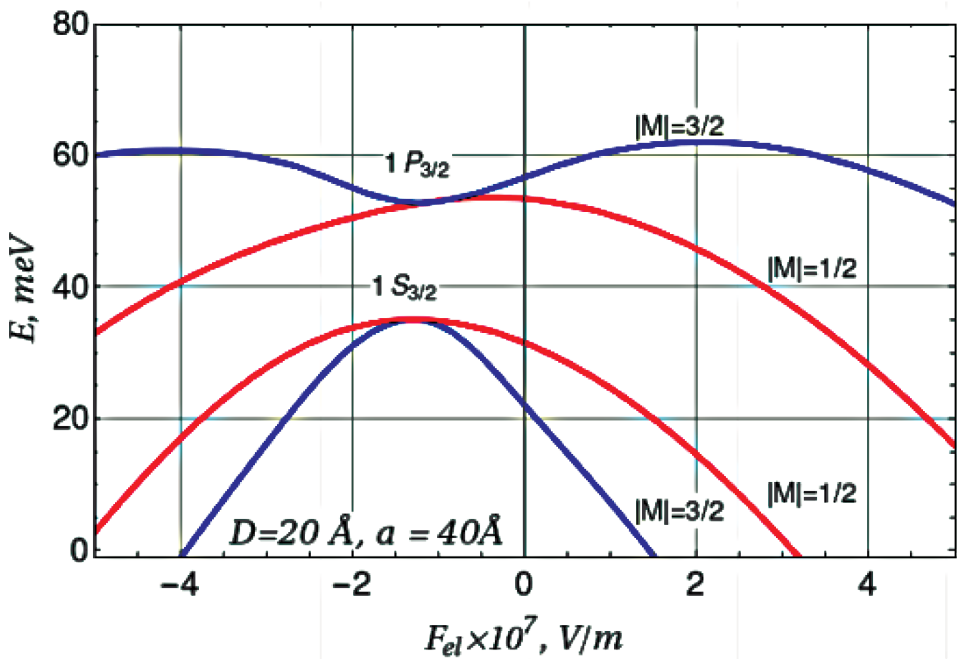

Figure 1. Hole energies of $1S_{3/2}$ and $1P_{3/2}$ states as the function of the electric field in QD heterostructure *GaAs/AlAs.* QD radius is 40 Å, the acceptor ion of is located at the distance *D*=20 Å form the QD center.

can lead to both an increase in the splitting of energy levels and a decrease in the splitting [22].

All of this will be reflected in the optical absorption coefficient.

Optical absorption

The coefficient of linear optical absorption due to the interlevel optical transition causes by a linear polarized wave is written in the form [25, 26]:

$$\alpha_{1,2}(\omega) = \omega\sqrt{\frac{\mu_0}{\varepsilon_0\varepsilon}}\frac{N\left|d_{1,2}\right|^2\hbar\Gamma}{\left(E_2 - E_1 - \hbar\Gamma\right)^2 + \left(\hbar\Gamma\right)^2}, \quad (14)$$

where ω is the frequency of electromagnetic wave, μ_0 is the magnetic constant. The electron density in the QD N is the QD concentration. E_1 and E_2 are energies of the initial and final states. $d_{1,2}$ is the matrix element of the dipole transition. $\hbar\Gamma$ is the relaxation rate caused by the electron-phonon interaction and some other scattering factors. If $\hbar\Gamma \to 0$ we got

$$\alpha_{1,2}(\omega) = \omega\pi\sqrt{\frac{\mu_0}{\varepsilon_0\varepsilon}}\sigma\left|d_{1,2}{}^2\right|\delta(E_2 - E_1 - \hbar\omega). \quad (15)$$

In practice, sets of QDs arranged in a matrix are obtained. Regardless of the growth method used, any set of QDs can always be characterized by a size dispersion. Let the distribution of QDs be described by a Gaussian function:

$$g(\tilde{s}, \bar{a}, a) = \frac{1}{\tilde{s}\sqrt{2\pi}}\exp\left(\frac{-(a-\bar{a})^2}{2\tilde{s}^2}\right), \quad (16)$$

where a is the radius of the quantum dot (variable), $\tilde{s}$ is the half-width of the distribution (16), which can be expressed through the average radius $\bar{a}$ and the value of σ, representing the size variation of the quantum dots in percentage: $s = \bar{a}\sigma/100$. Considering the size dispersion of the quantum dots (17), the absorption coefficient of the quantum dot system is written as:

$$\alpha_{1,2;system}(\omega) = \omega\pi\sqrt{\frac{\mu_0}{\varepsilon_0\varepsilon}}N\int g(\tilde{s}, \bar{a}, a)\left|d_{mn}\right|^2\delta(E_2(a) - E_1(a) - \hbar\omega)da.$$

After integrating and considering the properties of the delta-function, the following result is obtained:

$$\alpha_{1,2;system}(\omega) = \omega\pi\sqrt{\frac{\mu_0}{\varepsilon_0\varepsilon}}N\sum_i \frac{\left|d_{12}(a_{0i})\right|^2 g(\tilde{s}, \bar{a}, a_{0i})}{\frac{d}{da}\left(E_2(a) - E_1(a) - \hbar\omega\right)\Big|_{a=a_{0i}}}. \quad (17)$$

In the spherical symmetry case (when $F_{el} = 0$ and Z = 0) the energies of both the ground state $1S_{3/2}$ and first exited state $1P_{3/2}$ is quadruple degenerate. But when $Z \neq 0$ or $F_{el} \neq 0$ the energy levels are split by $|M| = 1/2, 3/2$. Therefore, there are two possible transitions between those levels. According to the selection rules transition is allowed when

$$d_{(1S_{3/2},|M|=3/2);(1P_{3/2},|M|=3/2)} \neq 0, \quad d_{(1S_{3/2},|M|=1/2),(1P_{3/2},|M|=1/2)} \neq 0$$

Therefore due to splitting, there are two absorption band (between states with $|M|3/2 \to 3/2$ and $1/2 \to 1/2$) instead of one in the absence of an electric field and a central impurity.

We have calculated the optical absorption coefficients for various values and directions of the electric field, specifically for $-3\cdot10^7$, $-1\cdot10^7$ and $1\cdot10^7$ V/m, in the presence of an acceptor at a distance of D=20 Å. The results show that the presence of an electric field, combined with the impurity, leads to the existence of two absorption bands corresponding to $|M|$ = 1/2 and 3/2. As in the case without an electric field and with a non-central acceptor impurity, the absorption bands corresponding to transitions between states with $|M|$ = 3/2 are larger than those corresponding to transitions between states with $|M|$ = 1/2. The distances between the bands for the cases

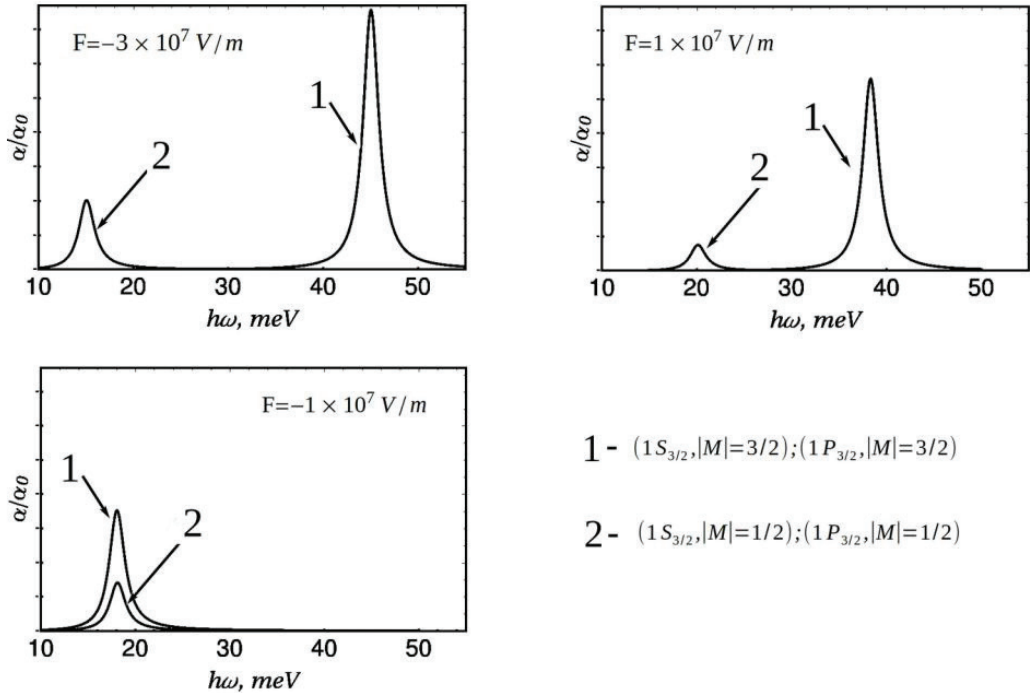

Figure 2. Optical absorption coefficient caused by the interlevel hole transition between allowed states for different electric fields. QD radius is 40 Å, the acceptor ion of is located at the distance *D*=20 Å form the QD center, QD dispersion 5%.

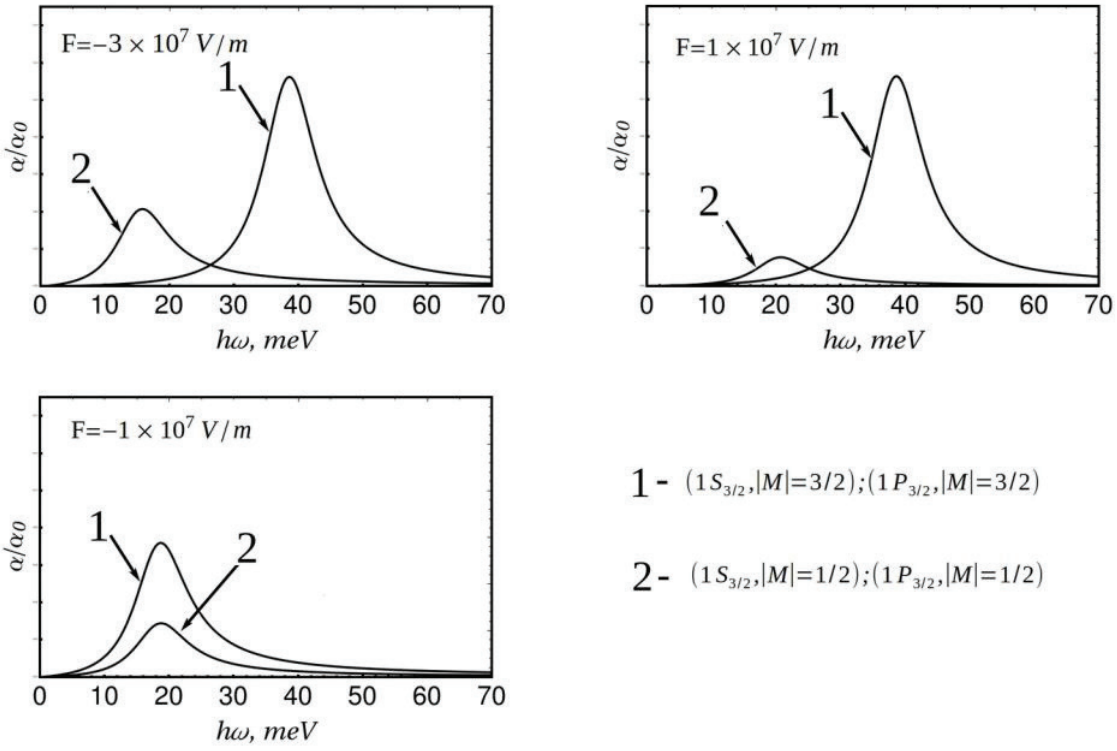

Figure 3. Optical absorption coefficient caused by the interlevel hole transition between allowed states for different electric fields. QD radius is 40 Å, the acceptor ion of is located at the distance *D*=20 Å form the QD center, QD dispersion 20%.

$F = -3\times10^7$ and $F = 1\times10^7$ V/m, are significant. If the size dispersion of the quantum dot is small, around 5% (see Fig. 2), these bands are clearly distinguishable. However, with larger dispersion of 20% (see Fig. 3), these absorption bands "overlap", making it practically difficult (and often impossible) to identify them experimentally (especially when considering temperature). Nevertheless, even without an applied electric field, a non-central impurity splits the states, and two bands exist, even if they are poorly identifiable. This blurring, however, broadens the broadband "tail" of absorption, which is often undesirable. For any arbitrary impurity position, there is always a specific electric field value that restores the degeneracy of the energy levels. For instance, at D=20 Å and a=40 Å, such a field value is approximately -1×10^7. In this case, the absorption bands coincide, and overall, the broadband "tail" of absorption becomes narrower, even with small or large size dispersion.

Conclusion

The results of our calculations demonstrate that the presence of an electric field, combined with a non-central impurity, leads to the emergence of two distinct absorption bands corresponding to different magnetic quantum numbers. The magnitude of the absorption bands depends on the direction and strength of the electric field, with significant differences observed between fields of opposite polarity. Notably, there exists a critical field strength that restores the degeneracy of energy levels, narrowing the broadband absorption tail for systems with small or large size dispersions. The proposed models can also be extended to the case of arbitrary electric field orientation relative to the impurity displacement, which will be addressed in future studies. This extension will provide further insights into the control of optical properties in quantum dots under electric fields.

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THE INTERSUBBAND OPTICAL ABSORPTION COEFFICIENT OF THE QD WITH ACCEPTOR IMPURITY UNDER APPLIED ELECTRIC FIELD

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Summary

Quantum dots (QDs) are nanostructures with unique optical properties. They are of interest to both theorists and experimentalists. The optical characteristics of quantum dots are influenced by various factors, including the shape of the QDs, the QD material, their environment, and impurities of donor or acceptor type. In this work, a spherical quantum dot under the influence of an external electric field is investigated. A multi-band model of the valence band is applied. The influence of off-center acceptor impurity, electric field and QD size dispersion on the absorption coefficient during intersubband transitions between hole states has been analyzed. The results show that an electric field combined with an off-center impurity induces the appearance of two distinct absorption bands corresponding to different magnetic quantum numbers. The intensity of absorption bands depends on the direction and strength of the electric field, and significant differences are observed between fields of opposite polarity. It is important to note that there is a critical field strength that restores the degeneracy of the energy levels, narrowing the broadband absorption tail for systems with small or large dispersion sizes. This research aims to improve the understanding and optimization of the optical properties of nanomaterials.

Keywords: acceptor impurity, electric field, hole energy spectrum, multiband hole model, level splitting

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КОЕФІЦІЄНТ МІЖПІДЗОННОГО ОПТИЧНОГО ПОГЛИНАННЯ КВАНТОВОЇ ТОЧКИ З АКЦЕПТОРНОЮ ДОМІШКОЮ В ПРИКЛАДЕНОМУ ЕЛЕКТРИЧНОМУ ПОЛІ

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Реферат

Квантові точки (КТ) – це наноструктури з унікальними оптичними властивостями. Вони цікавлять як теоретиків, так і експериментаторів. На оптичні характеристики квантових точок впливають різні фактори, зокрема форма КТ, матеріал КТ, їхнє оточення та домішки донорного чи акцепторного типу. У роботі досліджено сферичну квантову точку під впливом зовнішнього електричного поля. Застосовано багатозонну модель валентної зони. Проаналізовано вплив нецентральної акцепторної домішки, електричного поля та дисперсії розміру КТ на коефіцієнт поглинання при міжпідзонних переходах між дірковими станами. Результати показують, що електричне поле в поєднанні з нецентральною домішкою викликає появу двох чітких смуг поглинання, що відповідають різним магнітним квантовим числам. Інтенсивність смуг поглинання залежить від напрямку і напруженості електричного поля, причому значні відмінності спостерігаються між полями протилежної полярності. Важливо відзначити, що існує критична напруженість поля, яка відновлює виродження енергетичних рівнів, звужуючи широкосмуговий хвіст поглинання для систем з малими або великими розмірами дисперсії. Це дослідження спрямоване на покращення розуміння та оптимізацію оптичних властивостей наноматеріалів.

Ключові слова: акцепторна домішка, електричне поле, енергетичний спектр дірок, багатозонна діркова модель, розщеплення рівнів