Proceedings of the 10th World Congress on Recent Advances in Nanotechnology (RAN 2025)
Barcelona, Spain - April 7 - 9, 2025
Paper No. ICNNFC 103
DOI: 10.11159/icnnfc25.103

Exciton Stark Shift in CdSe Nanoplatelet

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Abstract **-** In the effective mass approximation, the effect of a planar external uniform electric field on excitonic states in a CdSe nanoplatelet (NPL) is considered. It is shown that an external field deforms the Coulomb-like potential of electron-hole attraction in a NPL, which ultimately leads to the destruction of the exciton as a bound state. Computational calculations show the degree of deformation of the electron-hole potential and the change in the spatial distribution of the probability density (exciton wave function) under the influence of an external field. Curves are also shown that illustrate the degree of shift in the position of the main exciton energy level (Stark shift) under the influence of an external field. In the region of weak fields, the magnitude of the energy shift has a quadratic dependence on the external field strength. For strong fields, this dependence on the field is linear. It is also shown that the effect of an external field on excitonic states in a NPL strongly depends on the number of atomic monolayers in the direction of size quantization of charge carriers. It is also shown that the effect of an external field on excitonic states in a nanoplate strongly depends on the number of monolayers in the direction of size quantization of charge carriers. As the number of monolayers increases, the effect of the external field increases, which is a consequence of a decrease in the exciton binding energy with increasing of NPL thickness.

Keywords**:** nanoplatelet, exciton, size quantization, electric field, Stark shift;

1. Introduction

Along with many low-dimensional semiconductors, various quasi-two-dimensional quantum nanostructures with anomalously strong size quantization in one direction - semiconductor nanoplatelets (NPLs) - have also been intensively studied in the last two decades. NPLs based on compounds II-VI (CdS, CdSe, CdTe) are the most intensively studied (see, for example, Refs. [1-4]). Semiconductor NPLs exhibit a number of unique properties, especially in the field of optical phenomena, which are fundamentally impossible not only in massive samples, but also in quantum-sized structures of the previous generation: quantum films, quantum wires and quantum dots [1-7]. One of the most important factors that shape and determine the optical properties of semiconductor NPLs is the specificity of excitonic states in these structures (see, for example, Refs. [1- 4,8-11]). First of all, these are the large values of exciton binding energy in NPLs compared not only to bulk samples, but also compared to conventional quantizing films of the previous generation. For example, the exciton binding energy $E_{b,ex}^{3D}$ in a bulk CdSe sample is 15 meV, in a quantized film-quantum well $E_{b,ex}^{QF} \sim$ 60 meV, and in NPL this energy already reaches hundreds of meV $E_{b,ex}^{2D} \sim$ 200- 300 meV at room temperature [2,10,12]. Moreover, as theoretical calculations and the corresponding experiment show, the value of $E_{b,ex}^{2D}$ significantly depends on the number of monolayers (ML) in the NPL: with a decrease in the number of monolayers, $E_{b,ex}^{2D}$ increases [2, 10,12].

For real practical application of a number of unique properties of nanoplatelets that have been identified so far, it is necessary to study external influences that make it possible to change these properties in a controlled manner. One of the powerful modulating external factors that significantly affects the states of charge carriers in a semiconductor is, as it is known, a uniform electric field. Due to the important role noted above that excitons play in the formation of the optical properties of NPLs, it is of particular interest to study the effect of an external uniform electric field specifically on excitonic states in NPLs. It is clear that, due to the presence of a strong size quantization anomaly in NPLs, the effect of the field on a 2D exciton in a nanoplatelet will be completely different when the field is directed along the strong quantization axis (z) compared to the case when it is directed in the NPL plane. In Ref. [13] in detail the case is examined

when the external uniform field is directed along the axis of strong quantization of charge carriers (z) in CdSe NPL. It is shown that the thus directed electric field has a correlating weakening effect on the electron-hole attraction also in the NPL plane. Changes in the linear and nonlinear optical absorption spectra that occur in CdSe NPL due to the action of an electric field lateral to the plane are presented.

A number of works have also considered the effect of a planar uniform electric field on the excitonic states of a number of quasi-two-dimensional structures made of various materials (see, for example, Refs. [14-18]). In these works, the exciton Stark effect, the dissociation of excitons under the influence of an external field, the influence of the field on the optical absorption spectrum of the sample, etc. were examined experimentally and theoretically.

In the present work, the effect of an external in-plane uniform electrostatic field on exciton states in CdSe NPLs of various thicknesses is considered theoretically. In the model we use, we assume that the band structure and, in particular, the effective mass of the exciton does not change under the action of an external electric field.

2. CdSe Nanoplatelet in the In-Plane Uniform Electrostatic Field

For the Hamiltonian of an electron and a hole interacting with each other in the presence of an in-plane field, in the most general case we can write:

$$\hat{H} = -\frac{\hbar^2\Delta_{\vec{\rho}_e}}{2\mu_{\square,e}} - \frac{\hbar^2\Delta_{\vec{\rho}_h}}{2\mu_{\square,h}} - \frac{\hbar^2\Delta_{z_e}}{2\mu_{z,e}} - \frac{\hbar^2\Delta_{z_h}}{2\mu_{z,h}} + U\left(z_e, z_h, \vec{\rho}_e, \vec{\rho}_h\right) + |e|\left(\vec{F}\cdot(\vec{\rho}_e - \vec{\rho}_h)\right)$$
$$\vec{F} = \vec{F}\left(F_x; F_y; 0\right);\ \vec{\rho}_{e,h} = \vec{\rho}_{e,h}\left(x_{e,h}; y_{e,h}; 0\right) \tag{1}$$

Here $\Delta_{\vec{\rho}_i}, \Delta_{z_i}$ are the Laplacian of electron and hole in *XY* plane and along the stacking direction, $\mu_{\square,i}$ and μ_{zi} are effective masses in *XY* plane and along the *z* axis for particle *i* = *e, h* (electron or hole) respectively, $\vec{\rho}_{e,h}$ are the in-plane *2D* radius-vectors of electron and hole, respectively, *e* is the elementary charge, and $\vec{F}$ is the in-plane electric field strength. The potential energy term of electron-hole pair can be written as:

$$U\left(z_e, z_h, \vec{\rho}\right) = U^e\left(z_e\right) + U^h\left(z_h\right) + U^e_{conf}\left(x_e, y_e\right) + U^h_{conf}\left(x_h, y_h\right) + V\left(z_e, z_h, \vec{\rho}\right)$$
$$\vec{\rho} = \vec{\rho}_e - \vec{\rho}_h \tag{2}$$

where $V\left(z_e, z_h, \vec{\rho}\right)$ is the interaction between an electron and a hole, $U^i\left(z_i\right)$ are one-particle confining potential energies appearing due to the conduction and valence band offsets, together with the self-energy correction appearing because of the mismatch of the dielectric constants in the regions of a barrier and a well [2,13]:

$$U^i\left(z_i\right) = U^i_{conf}\left(z_i\right) + U^i_{self}\left(z_i\right) \tag{3}$$

The confining potential defined by the band offset between the NPL and its surrounding material is taken as a finite potential well [2,13]:

$$U^i_{conf}\left(z_i\right) = \begin{cases} U^i_0, |z_i| > L_z/2 \\ 0, |z_i| \le L_z/2 \end{cases};\ U^i_{conf}\left(x_i, y_i\right) = \begin{cases} \infty, \left(x_i, y_i\right) \in NPL \\ 0, \left(x_i, y_i\right) \notin NPL \end{cases} \quad \left(i = e, h\right) \tag{4}$$

Here $U^i_0 \equiv U^{e,h}_0$ is the depth of the potential well for electron and hole, respectively, and $U^i_{self}\left(z_i\right)$ is self- induced potential for every type of carriers (see, for example Refs. [2,13,19]):

$$U_{\text{self}}(z) = \sum_{m=\pm1,\pm2,\ldots} \frac{\kappa^{|m|} e^2}{2\varepsilon_w \left| z - (-1)^m z + mL_z \right|}, \left| z \right| < L_z / 2,$$

$$U_{\text{self}}(z) = \frac{2\varepsilon_w}{\varepsilon_w + \varepsilon_b} \sum_{m=0}^{\infty} \frac{\kappa^{2m+1} e^2}{(\varepsilon_w + \varepsilon_b) \left| 2z + (2m+1)L_z \right|} - \frac{\kappa e^2}{2\varepsilon_b \left| 2z - L_z \right|}, \quad z > L_z / 2 \tag{5}$$

$$U_{\text{self}}(z) = U_{\text{self}}(-z), \quad z < -L_z / 2$$

where $\kappa = \frac{\varepsilon_w - \varepsilon_b}{\varepsilon_w + \varepsilon_b}$, ε_w and ε_b are the dielectric materials of the NPL material and of the spacer, respectively, and L_z is the thickness of NPL along the quantization direction. The solution of the Schrödinger equation with Hamiltonian (1) in this case can be carried out with sufficient accuracy within the framework of the adiabatic approximation. Indeed, the energy of the "fast" movement of charge carriers along the quantization z-axis will always be significantly greater than the energy of any of the carrier states in the NPL plane. Accordingly, let us imagine the total wave function of an interacting electron-hole pair in an external field with the following form:

$$\Psi\left(\vec{\rho}_e, \vec{\rho}_h, F, z_e, z_h\right) = \psi_{\perp}\left(z_e, z_h\right) \psi_{\square}\left(\vec{\rho}_e, \vec{\rho}_h, F\right) \tag{6}$$

For the wave functions $\psi_{\perp}\left(z_e, z_h\right)$ we can write now [2,13]:

$$\psi_{\perp}\left(z_e, z_h\right) = \psi_{\perp}\left(z_e\right) \psi_{\perp}\left(z_h\right) \tag{7}$$

After this, we average the electron-hole interaction potential $V\left(z_e, z_h, \vec{\rho}\right)$ over single-particle states (7) [2,13,19]:

$$V_{e-h}^{2D}\left(\varepsilon_w, \varepsilon_b, \rho_e, \rho_h\right) = -e^2 \sum_{n=-\infty}^{\infty} \frac{q_n}{\varepsilon_w} \int_0^{L_z} \int_0^{L_z} \frac{\left|\psi_{\perp,e}\left(z_e\right)\right|^2 \left|\psi_{\perp,h}\left(z_h\right)\right|^2 dz_e dz_h}{\sqrt{\left(\rho_e - \rho_h\right)^2 + \left[z_e - (-1)^n z_h - nL_z\right]^2}}; \; q_n = \left(\frac{\varepsilon_w - \varepsilon_b}{\varepsilon_w + \varepsilon_b}\right)^{|n|} \tag{8}$$

To determine in-plane excitonic states in NPL, we now obtain the following Hamiltonian:

$$\hat{H}_{ex,F}^{2D} = -\frac{\hbar^2 \Delta_{\vec{\rho}_e}}{2\mu_{\square,e}} - \frac{\hbar^2 \Delta_{\vec{\rho}_h}}{2\mu_{\square,h}} + V_{e-h}^{2D}\left(\varepsilon_w, \varepsilon_b, \vec{\rho}_e, \vec{\rho}_h,\right) + \left|e\right|\left(\vec{F} \cdot (\vec{\rho}_e - \vec{\rho}_h)\right) \tag{9}$$

For typical NPLs, the following conditions between the longitudinal L_z and lateral L_x, L_y dimensions are always met [20]:

$$L_z^2 \square \left(a_{ex}\right)^2; \quad L_z^2, \left(a_{ex}\right)^2 \square L_x^2, L_y^2 \tag{10}$$

Here a_{ex} is the Bohr radius of exciton. The condition (10) allows us to separate the relative motion of electron and hole and the motion of the center of mass of *2D* exciton in *XY* plane. Correspondingly, for definition of full wave functions and energy of in-plane interacting electron – hole pare we can write the following two equations (11) and (12) - for center mass and relative motion respectively:

$$-\frac{\hbar^2}{2M}\left(\frac{\partial}{\partial X}+\frac{\partial}{\partial Y}\right)\psi_{N_x,N_y}(X,Y)=E^{c.m}_{N_x,N_y}\psi_{N_x,N_y}(X,Y);\ M=\mu_{\square,e}+\mu_{\square,h};\ N_x,N_y=1,2,3,...$$
$$X=\frac{\mu_{\square,e}x_e+\mu_{\square,h}x_h}{\mu_{\square,e}+\mu_{\square,h}};\ Y=\frac{\mu_{\square,e}y_e+\mu_{\square,h}y_h}{\mu_{\square,e}+\mu_{\square,h}};\tag{11}$$

$$\left[-\frac{\hbar^2}{2\mu_{ex}}\left(\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial y^2}\right)+V^{2D}_{e-h}(\varepsilon_w,\varepsilon_b,x,y,)+|e|Fy\right]\psi_{ex}(x,y)=E^b_{ex,F}\psi_{ex}(x,y)$$
$$\mu_{ex}=\frac{\mu_{\square,e}\mu_{\square,h}}{\mu_{\square,e}+\mu_{\square,h}}\tag{12}$$

Here $(\mu_{\square})_{e,h}$ - effective electron and hole masses in the NPL plane, $E^{c.m}_{N_x,N_y}$ - energy of motion of the center of mass of a *2D* exciton in the *XY* plane, $E^b_{ex,F}$ – energy of the bound excitonic state of an electron and a hole in the NPL plane in the presence of an external field.

$$\vec{F}=\vec{F}(0,F,0);(\vec{F}\cdot\vec{\rho})=Fy\tag{13}$$

For definiteness, we chose the external field to be directed along the y axis. The above reasoning and the corresponding expressions obtained on their basis are of a general nature for NPLs that satisfy the conditions presented in the problem. Let us now turn to consideration of the action of an in-plane uniform electric field on specific structures of CdSe NPL. We will consider NPLs with a thickness of n= 3,5;4,5; 5,5;7,5 atomic monolayers (ML).

2.1. Influence of Uniform In- Plane External Field on Exciton States in CdSe NPL

When the external field is oriented according to (13), the field does not have any effect on the states of charge carriers as they move along the quantization axis (*z*). Accordingly, using the methodology of [2], we obtain a numerical solution for the wave functions and energies of the lowest states of size quantization of charge carriers along the z axis, which are of real physical interest. Table 1 presents the values of the first two energy levels of size quantization of electrons ($E^e_{n_z=1}, E^e_{n_z=2}$) and holes ($E^h_{n_z=1}, E^h_{n_z=2}$) for different numbers of monolayers (ML) along the quantization axis and the corresponding wave functions of. ground states charge carriers are shown in Fig1.

Table 1. Values of the first two levels of size quantization of electrons ($E^e_{n_z=1}, E^e_{n_z=2}$) and holes ($E^h_{n_z=1}, E^h_{n_z=2}$) in a CdSe nanoplate for different numbers of monolayers along the quantization axis (*z*). Data taken from [2].

ML	$\mu_{z,e}/m_0$	$\mu_{z,h}/m_0$	$E^e_{n_z=1}$ (meV)	$E^e_{n_z=2}$ (meV)	$E^h_{n_z=1}$ (meV)	$E^h_{n_z=2}$ (meV)
3,5	0.157	0.96	439	---	280	937
4,5	0.144	0.92	341	1700	197	665
5,5	0.138	0.90	277	1330	147	483
7,5	0.130	0.88	197	887	92	294

Here m_0 is the mass of free electron. The depths of the confinement potential for electron and hole are taken $U_0^e = 2eV$, $U_0^h = 2.5eV$ respectively [2].

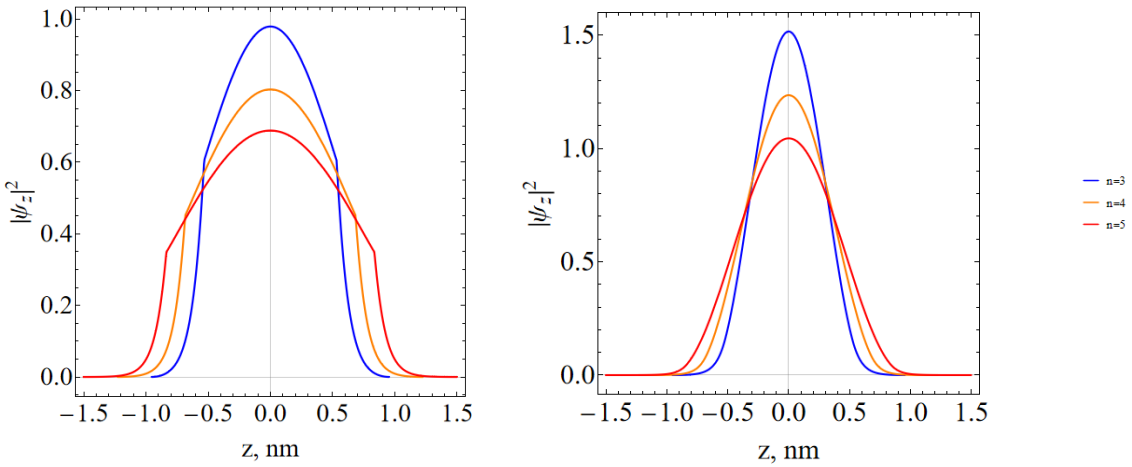

Fig 1. The electron (a) and hole (b) ground state density probability distribution in axial direction for different NPL thicknesses.

Let us now turn to the consideration of electron-hole states in the plane in the presence of an external field (13). The external field does not affect the movement of the *2D* exciton center of mass. Solving equation (11) for the energy and wave functions of motion of the center of mass of an electron-hole pair in the *XY* plane leads to the following results:

$$E^{c.m}_{N_x,N_y} = \frac{\pi^2\hbar^2}{2M}\left(\frac{N_x^2}{L_x^2}+\frac{N_y^2}{L_y^2}\right);\ \psi_{N_x,N_y}(X,Y) = \sqrt{\frac{2}{L_x}}\sqrt{\frac{2}{L_y}}\sin\frac{\pi N_x X}{L_x}\sin\frac{\pi N_y Y}{L_y}; \qquad (14)$$

Equation (12) is solved numerically. In the case of *F=0*, the energy values of ground exciton states and Bohr radii coincide with the results of the works [2,10]. In the absence of a field at a given energy value of the lowest exciton level $\left(E^b_{ex}(F=0)\right)_1$ the corresponding value of the intraexciton field strength F_{cr}, holding the exciton in a bound state can be estimated from the relation

$$\left|\left(E^b_{ex,F=0}\right)_1\right| \sim 2|e|F_{cr}a^{NPL}_{ex} \qquad (15)$$

In Table 2 the values of the lowest exciton level at *F=0* $\left(E^b_{ex}(F=0)\right)_1$, exciton Bohr radius $a^{NPL}_{ex}(F=0)$, and F_{cr} in CdSe NPL at the thickness of the quantizing layer n=3,5; 4,5; 5,5; 7,5 atomic ML are presented.

Table 2: The values of the lowest exciton levels at *F=0* and exciton Bohr radii in CdSe NPL at the thickness of the quantizing layer n=3,5; 4,5; 5,5; 7,5 atomic ML.

n(ML)	3.5	4.5	5.5	7.5
$\left(E_{ex}(F=0)\right)_1$ meV	312	257	220	180
$a_{ex}^{NPL}(F=0)$ nm	1.493	1.775	2.031	2.380
F_{cr} kV/cm	1044	725	541	378

From the form of equation (12) it is clear that the presence of an external field changes the shape and behavior of the electron-hole interaction potential. Accordingly, as the numerical solution of equation (12) shows, this necessarily leads to a change in the position of the energy levels (Stark shift) and a change in the behavior of the wave function of the pair.

The change in the shape of the electron-hole interaction potential and the wave function of the pair as a result of the numerical solution of equation (12) is shown in Fig. 2

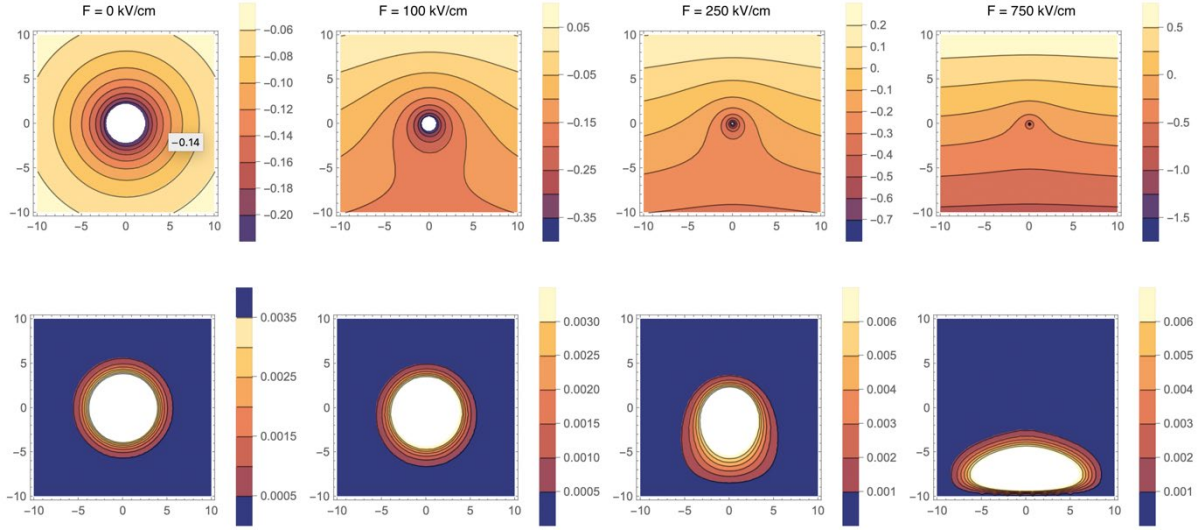

Fig 2. Exciton density distribution (upper row) and potential including electric filed (lower row). n=5,5.

From general quantum mechanical considerations, it is clear that in an asymmetric potential well from equation (12) with an increase in the field at a certain value, there comes a moment when the existence of bound states in this potential simply becomes impossible. This corresponds to a physical situation where the external field becomes so strong that it completely destroys the exciton as a bound state. That is, exciton dissociation occurs, induced by an external field.

Figure 3 graphically shows the Stark shift of the main exciton level under the influence of an external field.

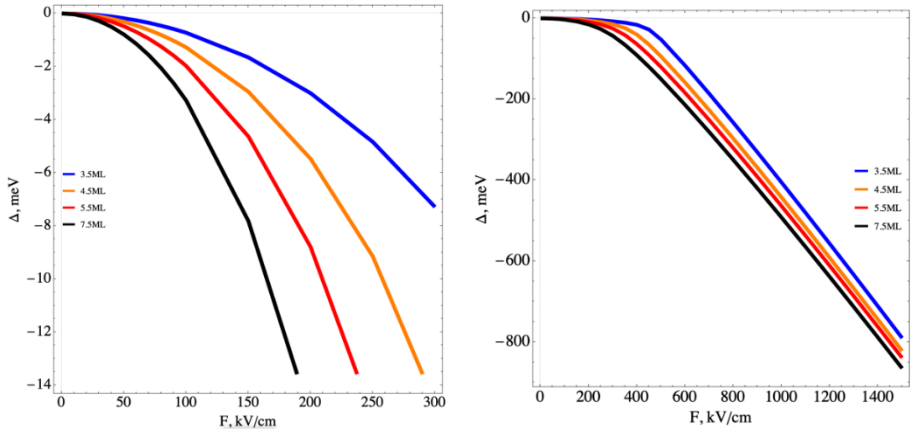

Fig 4. Stark shift dependence on external electric filed.

The left figure shows the offset for an area of relatively small values of external fields, when the following condition takes place: $F \ll F_{cr}$. The right figure shows the continuation of the exciton energy displacement curves already in the region of strong fields.

3. Conclusions

The binding energies of excitons in the system we are considering and in the structures considered, for example, in [14-16,21-23] are practically of the same order and amount to hundreds of meV. Accordingly, the calculated curve we obtained for the same range of external field values repeats the general behavior of the Stark shift curves for a 2D exciton in the mentioned works. For weak fields, this behavior corresponds to the quadratic dependence of the Stark shift on the field strength known from quantum mechanics [14-16,21-23]: $\Delta E\left(F\right) = \left(E_{ex}^{b}(F)\right)_1 - \left(E_{ex}^{b}(F=0)\right)_1 \sim F^2$. For strong fields, the dependence of the displacement on the external field strength becomes linear. Moreover, in both cases, both the magnitude of the displacement itself and the growth rate of the curve increase with increasing number of atomic monolayers of NPL in the quantization direction (z). This is explained by the weakening of the 2D exciton binding energy with increasing NPL thickness and the corresponding increase in the influence of the external field on the exciton state that follows from this fact. In this case, this manifests itself in an increase in the value of the Stark shift.

Summarizing the results of the work, it can be argued that by varying the number of layers of NPL and the intensity of the external field, it is possible to achieve a controlled change in the density of charge carriers and, accordingly, a number of characteristics of the sample.

Acknowledgements

I express my gratitude to the 4th year student in the field of "electronics and nanoelectronics" M. Manvelyan for his assistance in preparing this article

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