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THEORY OF LINEAR-CIRCULAR DICHROISM IN MONOATOMIC LAYERS OF TRANSITION METAL DICHALCOGENIDES TAKING INTO ACCOUNT THE RABI EFFECT

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We have developed a theory of dimensional quantization for nanostructures, both one-dimensional and zero-dimensional, constructed from monoatomic layers of transition metal dichalcogenides (TMDCs). This theory has enabled us to derive expressions for the energy spectra of charge carriers in both even and odd states (relative to coordinate inversion), as these states occur within quantum-confined lines and points of the TMDC monoatomic layers, dependent on their geometric dimensions. Our numerical analysis provides a detailed exploration of the quantum-confined energy states of electrons within these nanostructures, offering insights into their potential applications in advanced nanoelectronic devices. This work not only advances our understanding of the energy characteristics of TMDC monoatomic layers but also contributes to the broader field of material science by exploring the effects of dimensional quantization on electronic properties.
Keywords: *Dimensional quantization; One- and zero-dimensional nanostructure; Monatomic layer of transition metal dichalcogenides; Energy dispersion; Valence band; Conduction band*
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INTRODUCTION

A vital component of modern solid-state physics, both in experimental and theoretical aspects, is the rapidly developing field of low-dimensional systems of charge carriers. In particular, such low-dimensional systems include various nano-sized structures: superlattices, structures with quantum-confined wells, wires, and dots [1-3], the practical development of which is continually growing with the advancement of modern technologies for their fabrication [4]. Alongside, these low-dimensional structures serve as the elemental base of contemporary nanoelectronics and are the subjects of forward-looking research aimed at creating fundamentally new devices for spintronics, opto- and nanoelectronics with unique physical properties [5-8]. The main properties of these quantum-confined structures are determined by the spatial restriction of charge carriers' movement in one or several directions, which leads to the restructuring of the sample's band structure, i.e., the energy dispersion of carriers and other quasiparticles, such as phonons, excitons, and polaritons [9-11].

The unique properties of quantum-confined structures, which distinguish them from bulk samples on which they are based, allow for addressing the following tasks: a) new effects often arise in such structures related to dimensional quantization, and studying them is of undeniable interest from the physical point of view for analyzing the fundamental properties of low-dimensional objects. Secondly, nanostructures can be used to create solid-state devices of a new generation [12].

It should be noted that while many studies have been dedicated to dimensional quantization in crystals with tetrahedral symmetry and their multilayer structures [13-16], the question of quantizing the energy dispersion of charge carriers in monatomic layers of transition metal dichalcogenides (TMDCs) remains open, which is the focus of this work.

ENERGY DISPERSION IN A QUANTUM WIRE GROWN FROM MONATOMIC LAYERS OF TRANSITION METAL DICHALCOGENIDES

Fig. 1a, b schematically shows the crystal structure and arrangement of atoms in TMD monolayers. The monomolecular layer D_{3h} is characterized by a point group in which the horizontal plane of specular reflection passes through the layer of metal atoms. The elementary lattice contains a metal atom and two chalcogen atoms located in planes above and below the metal plane. The Brillouin zone is described as a regular hexagon (Fig. 1c). The correct exclusion zone is determined at the points $K_{\pm}$, which are determined by the time inversion operator with each other. $K_{\pm}$ electron dispersion in the valence band and conduction band near the points has a parabolic form (Fig. 1d). Note that the $K_{\pm}$ inclination of the bands relative to the spin at the points completely disappears. In this case, symmetry with respect to the time reversal operator makes it possible to associate states with opposite spins in different valleys.

The effective Hamiltonian of electrons in bulk transition metal dichalcogenides is represented as [17]

$$H = \begin{pmatrix} E_g/2 & \gamma k_- \\ \gamma k_+ & -E_g/2 \end{pmatrix}, \quad (1)$$

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where $k_{\pm} = k_x \pm ik_y, \vec{k}_{\perp} = k_{\perp}(\sin\varphi,\ \cos\varphi)$ - is a two-dimensional wave vector directed along the interface, $\gamma = \hbar\left(\tilde{E}_g/m^*\right)^{1/2}$ is the bandgap width, m^* is the effective mass of the charge carriers. For simplicity, in subsequent calculations of wave functions and energy spectra of charge carriers in quantum-confined structures grown based on TMDCs, assume that the effective mass of the charge carriers does not depend on the quantum number of dimensional quantization.

First, consider dimensional quantization in a potential well with infinitely high walls, made from monatomic layers of TMDCs, where it is assumed that the direction of dimensional quantization corresponds to the Ox axis. Then, the Schrödinger equation with Hamiltonian (1) is written as

$$(H - E)\psi = \begin{bmatrix} (E_g/2) - E & \gamma(k_x - ik_y) \\ \gamma(k_x + ik_y) & (-E_g/2) - E \end{bmatrix}\psi = 0, \tag{2}$$

The solution of which is a column matrix with dimensions of 1×2.

Then, from the equality $det(H - E) = 0$, it is easy to obtain the Schrödinger equation, with which one can determine the wave function and energy dispersion of charge carriers

$$\frac{\partial^2\psi}{\partial x^2} + \aleph_y^2\psi = 0, \tag{3}$$

where $\aleph_y^2 = \frac{1}{\gamma^2}\left(E^2 - E_y^2\right)$, $E_y^2 = \left(E_g/2\right)^2 + \gamma^2 k_y^2$ and we assume $k_x = -i\frac{\partial}{\partial x}$, k_y –is the wave vector of the charge carriers directed along the Oy axis. Then, by representing solution (3) as a superposition of exponential functions describing de Broglie waves with the wave vector $\aleph_y$ propagating both along and against the Ox axis, and considering the continuity and uniqueness of the wave function, it is easy to obtain that:

a) The energy dispersion of dimensional quantization in even (with respect to coordinate inversion) states is determined from the condition $e^{i2\aleph_y a} = 1$ $\left(e^{i\aleph_y a} = \pm 1\right)$. Then, the condition $\cos\left(\aleph_y a\right) = +1$ yields the expression for the quantum-confined energy dispersion as

$$E_a^{(\pm)} = \pm\left[\left(E_g/2\right)^2 + \gamma^2 k_y^2 + \gamma^2\frac{\pi^2}{a^2}n^2\right]^{1/2}, \tag{4}$$

From the condition $\cos\left(\aleph_y a\right) = -1$, we obtain

$$E_b^{(\pm)} = \pm\left[\left(E_g/2\right)^2 + \gamma^2 k_y^2 + \gamma^2\frac{\pi^2}{4a^2}(2n+1)^2\right]^{1/2}, \tag{5}$$

where $n -$ is the number of quantum-confined states;

b) The quantum-confined energy dispersion in odd (with respect to coordinate inversion) states is determined from the condition $\sin\left(\aleph_y a\right) = 0$, from which we obtain the expression determined by equation (4).

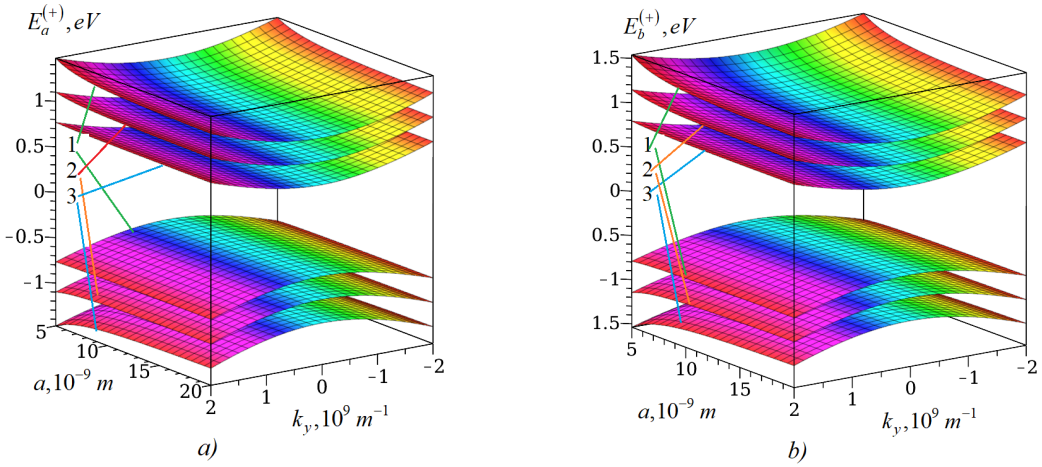

Figure 1. Graphs of the first quantum-confined energy dispersion of charge carriers as a function of well thickness and wave vector of charge carriers in a one-dimensional quantum well in a monatomic layer of various TMDCs, with bandgap widths $E_g = 1\ eV$ (graph 3), $E_g = 1.5\ eV$ (graph 2), and $E_g = 2\ eV$ (graph 3): a) energy dispersion of even states; b) energy dispersion of odd states.

On Fig. 1, the graphs of the first quantum-confined energy dispersion of charge carriers are presented as a function of well thickness and wave vector of charge carriers in a one-dimensional quantum well in a monatomic layer of TMDCs for bandgap widths $E_g = 1\ eV$ (graph 3), $E_g = 1.5\ eV$ (graph 2), and $E_g = 2\ eV$ (graph 3): a) energy dispersion of even states; b) energy dispersion of odd states, where the wave vector of the charge carriers is in units of $10^9\ m^{-1}$. Calculations were carried out according to formulas (5) and (6) with $m = 0.5\ m_0$. It can be seen from Fig. 1 that the energy dispersion

of charge carriers, both for even and odd states, consists of a set of quantum-confined subbands, the shape of which (in momentum space $\hbar k_y$) remains almost unchanged.

In Fig. 2, graphs of the quantum-confined energy dispersion of charge carriers in a one-dimensional quantum well in a monatomic layer of TMDCs are presented for a bandgap width of $E_g = 1\ eV$ (Figures a and b) and $E_g = 1.5\ eV$ (Figures c and d): a) and c) energy dispersion of even states; b) and d) energy dispersion of odd states, where the wave vector of the charge carriers is in units of $10^9 m^{-1}$. Calculations were carried out according to formulas (5) and (6) with $m = 0.5\ m_0$. It can be seen from Fig. 2 that for both even and odd states, the shape (steepness in momentum space k_y) of the quantum-confined subbands depends on both the subband number and the geometric size (a). In particular, the bandgap width in this quantum-confined structure increases with decreasing a, which is associated with the fact that the energy dispersion of charge carriers is inversely proportional to the magnitude of a.

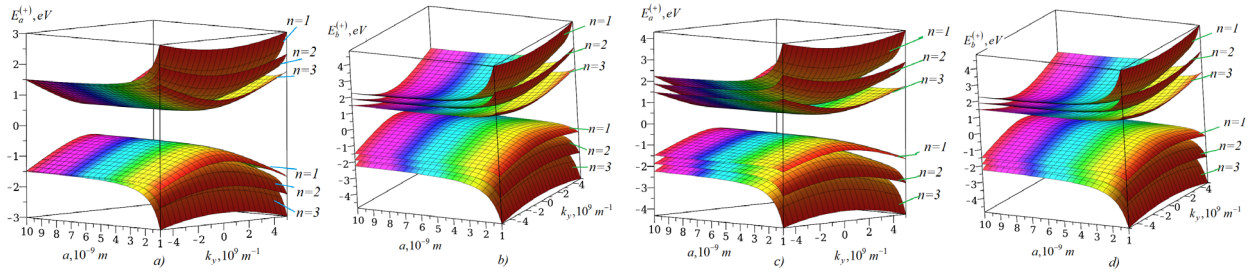

Figure 2. Graphs of the quantum-confined energy dispersion of charge carriers as a function of well thickness and wave vector of charge carriers in a one-dimensional quantum well in a monatomic layer of two TMDCs, with bandgap widths $E_g = 1\ eV$ (Figures a and b) and $E_g = 1.5\ eV$ (Figures c and d): a) and c) energy dispersion of even states; b) and d) energy dispersion of odd states.

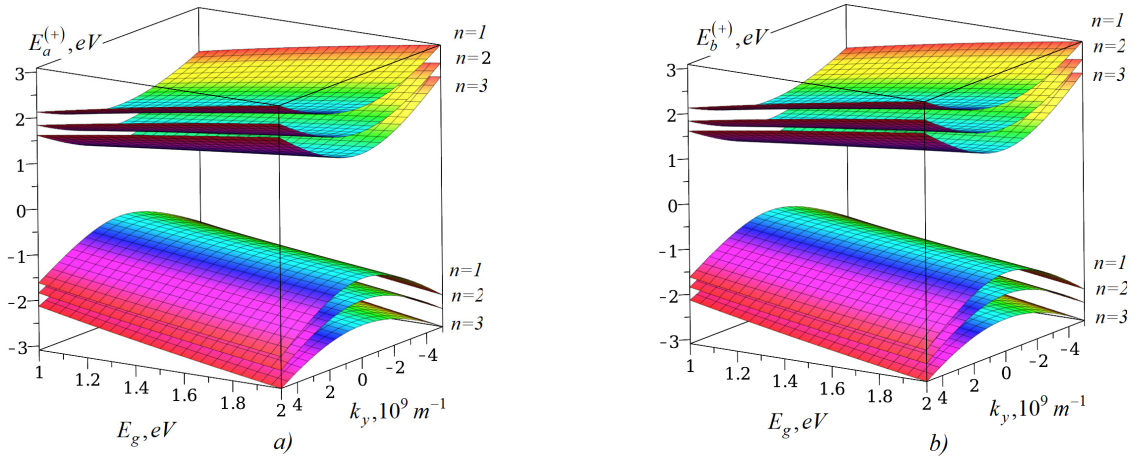

Figure 3. Graphs of the quantum-confined energy dispersion of charge carriers as a function of bandgap width and wave vector of charge carriers in a one-dimensional quantum wire of a monatomic layer of TMDCs, depending on the bandgap width E_g: a) energy dispersion of even states; b) energy dispersion of odd states.

$$E_b^{(\pm)} = \pm\left[\left(E_g/2\right)^2 + \gamma^2 k_y^2 + \gamma^2 \frac{\pi^2}{4a^2}(2n+1)^2\right]^{1/2}. \tag{6}$$

On Fig. 3, graphs of the quantum-confined energy dispersion of charge carriers in a one-dimensional structure - a quantum wire grown from a monatomic layer of TMDCs are presented as a function of the bandgap width E_g: a) energy dispersion of even states; b) energy dispersion of odd states, where the well thickness is in units of $20 \cdot 10^{-10}\ m$, and the wave vector of the charge carriers is in units of $10^9\ m^{-1}$. Calculations were carried out according to formulas (5) and (6) with $m = 0.5\ m_0 eV$. It can be seen from Fig. 3 that for both even and odd states of the quantum-confined energy dispersion of charge carriers, the narrower the bandgap width of the two-dimensional TMDC layer, the more noticeably the bandgap width of the one-dimensional structure decreases. This is associated with the dimensional quantization of the energy dispersions of charge carriers of the valence band and conduction band separately.

THE ENERGY DISPERSION IN A QUANTUM DOT GROWN FROM A MONATOMIC LAYER OF TMDCS

To calculate the energy spectrum in a quantum dot grown from a monatomic layer of TMDCs, let's choose the potential to be zero.

Where a and b are the thicknesses of the potential well along the Ox and Oy axes, respectively.

Then the Schrödinger equation takes the form:

$$\begin{bmatrix} (E_g/2)-E & \gamma(k_x-ik_y) \\ \gamma(k_x+ik_y) & (-E_g/2)-E \end{bmatrix}\begin{bmatrix}\psi_1 \\ \psi_2\end{bmatrix}=0, \tag{7}$$

where $k_x=\frac{1}{i}\frac{\partial}{\partial x}$, $k_y=\frac{1}{i}\frac{\partial}{\partial y}$. Considering the operators of wave vectors, it is easy to

$$\begin{cases}\left[(E_g/2)-E\right]\psi_1+\gamma\left(\frac{1}{i}\frac{\partial\psi_2}{\partial x}-i\frac{1}{i}\frac{\partial\psi_2}{\partial x}\right)=0, \\ \gamma\left(\frac{1}{i}\frac{\partial\psi_1}{\partial x}+i\frac{1}{i}\frac{\partial\psi_1}{\partial x}\right)+\left[(-E_g/2)-E\right]\psi_2=0,\end{cases} \tag{8}$$

where ψ_1 and ψ_2. Considering the operators of wave vectors, it is easy to

$$\psi_1=-\frac{\gamma}{(E_g/2)-E}\left(\frac{1}{i}\frac{\partial\psi_2}{\partial x}-\frac{\partial\psi_2}{\partial y}\right),\ \psi_2=-\frac{\gamma}{(-E_g/2)-E}\left(\frac{1}{i}\frac{\partial\psi_1}{\partial x}+\frac{\partial\psi_1}{\partial y}\right).$$

Then it is easy to obtain the Schrödinger equation for ψ_1

$$\frac{\partial^2\psi_1}{\partial x^2}+\frac{\partial^2\psi_1}{\partial y^2}+\frac{E^2-\left(E_g/2\right)^2}{\gamma^2}\psi_1=0, \tag{9}$$

assuming $\psi_1(x,y)=X(x)\times Y(y)$, we obtain equations for the functions $X(x)$ and $Y(y)$

$$\frac{\partial^2X(x)}{\partial x^2}+\kappa^2X(x)=0,\ \ \frac{\partial^2Y(y)}{\partial y^2}+\chi^2Y(y)=0, \tag{10}$$

where

$$\chi^2=\frac{E^2-\left(E_g/2\right)^2}{\gamma^2}-\kappa^2, \tag{11}$$

where κ^2 is an unknown quantity determined depending on the boundary conditions of the problem. We seek solutions to the latter equations in the form of

$$X(x)=\mathrm{C}_+e^{i\kappa x}+C_-e^{-i\kappa x},\ Y(y)=D_+e^{i\chi y}+D_-e^{-i\chi y}. \tag{12}$$

From the continuity condition of wave functions for even states with respect to coordinate inversion, we obtain the following relationships:

$$X_+(x)=(2\kappa)^{1/2}e^{-i\kappa a}\frac{\cos(\kappa(x-a))}{[\cos(\kappa a)\cdot\sin(\kappa a)+\kappa a]^{1/2}}, \tag{13}$$

$$Y_+(y)=(2\chi)^{1/2}e^{-i\chi b}\frac{\cos(\chi(x-b))}{[\cos(\chi\mathrm{b})\cdot\sin(\chi\mathrm{b})+\chi\cdot\mathrm{b}]^{1/2}}, \tag{14}$$

and for odd states

$$X_-(x)=(2\kappa)^{1/2}e^{-i\kappa a}\frac{\sin(\kappa(x-a))}{[-\cos(\kappa a)\cdot\sin(\kappa a)+\kappa a]^{1/2}}, \tag{15}$$

$$Y_-(y)=(2\chi)^{1/2}e^{-i\kappa b}\frac{\sin(\chi(y-b))}{[-\cos(\chi b)\cdot\sin(\chi b)+\chi b]^{1/2}}, \tag{16}$$

From the condition of uniqueness of the wave functions $X(x)$ and $Y(y)$, we obtain expressions for the unknown quantity κ^2 as follows:

a) for even states: $\kappa=\frac{\pi}{2a}(2n_x+1)$, and for odd states: $\kappa^2=\frac{\pi^2}{a^2}n_x^2$;

b) for the quantity χ, we have the following relationships: for even states: $\chi=\frac{\pi}{2a}\left(2n_y+1\right)$, and for odd states: $\chi=\frac{\pi}{b}n_y$, where n_x and n_y are integers numbering the quantum-confined states of charge carriers moving in the directions of Ox and Oy.

Taking into account the previous results, we obtain expressions for the quantum-confined energy dispersion of charge carriers in the quantum dot grown from a monatomic layer of TMDCs as follows:

a) for even states

$$E_{simm}^{(\pm)}(n_x,n_y)=\pm\left\{\left(E_g/2\right)^2+\gamma^2\left[\frac{\pi^2}{4a^2}(2n_x+1)^2+\frac{\pi^2}{4a^2}\left(2n_y+1\right)^2\right]\right\}^{1/2}; \tag{17}$$

b) for odd states

$$E_{asimm}^{(\pm)}(n_x,n_y)=\pm\left\{\left(E_g/2\right)^2+\gamma^2\left[\frac{\pi^2}{a^2}n_x^2+\frac{\pi^2}{b^2}n_y^2\right]\right\}^{1/2}. \tag{18}$$

From the last relationships, it is evident that if the quantum dot has symmetric shape (cube), i.e., equal dimensions $a=b$, then both $E_{simm}^{(\pm)}(n_x,n_y)$ and $E_{asimm}^{(\pm)}(n_x,n_y)$ are degenerate for certain values of the quantum number. This means

that the energy dispersion of charge carriers is doubly degenerate. Specifically, we have $E^{(\pm)}_{asimm(simm)}(1,2) = E^{(\pm)}_{asimm(simm)}(2,1), E^{(\pm)}_{asimm(simm)}(3,2) = E^{(\pm)}_{asimm(simm)}(3,2)$, and so on. A similar situation occurs in an asymmetric quantum dot (when $a \neq b$)) (see Fig. 4).

On Fig. 4, graphs of the quantum-confined energy dispersion of charge carriers in the quantum dot grown from a monatomic layer of TMDCs are presented for band gap widths $E_g = 1\ eV$ (Figs. a and b) and $E_g = 1.5\ eV$ (Figs. 4 c and d) as a function of geometric dimensions: a) and c) energy dispersion of even states; b) and d) energy dispersion of odd states. Here, the well thickness is in units of $10^{-9}\ m$ m, and the wave vector of the charge carriers is in units of $10^{9}\ m^{-1}$. The calculations are performed according to formulas (18) and (19) with $m = 0.5\ m_0$. The quantum numbers are chosen as (n_x, n_y). From Fig. 4, it can be observed that: a) the energy dispersion of charge carriers consists of a set of quantum-confined subbands, the values of which depend on both the band parameters and the geometric dimensions of the sample, as well as on the quantum number of the subbands; b) the energy difference between closely spaced quantum-confined subbands increases as the geometric dimensions of the structure decrease, which is related to the fact that the energy dispersion is inversely proportional to the size of the structure; c) some quantum-confined subbands intersect, resulting in a twofold degeneracy, and the regions of intersection depend on both the quantum numbers of the quantum-confined subbands and the aspect ratio (a/b).

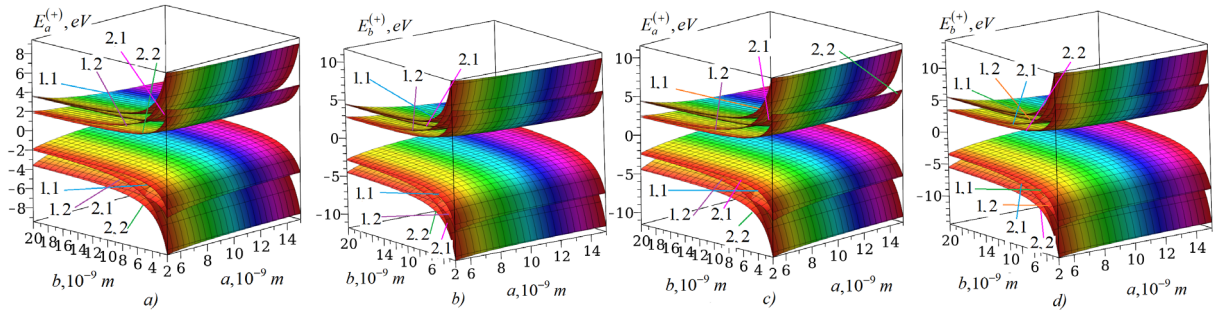

Figure 4. Graphs of the quantum-confined energy dispersion of charge carriers in the quantum dot of a monatomic layer of TMDCs for band gap widths $E_g = 1\ eV$ (Figs. a and b) and $E_g = 1.5\ eV$ (Figs. c and d): a) and c) energy dispersion of even states; b) and d) energy dispersion of odd states. The quantum numbers are represented on the figure as (n_x, n_y).

CONCLUSIONS

A theory of dimensional quantization of energy dispersions of charge carriers has been developed both in the valence band and in the conduction band in zero and one-dimensional nanostructures grown from monolayers of transition metal dichalcogenides (TMDs). Expressions for the energy spectra of charge carriers in one- and zero-dimensional structures have been obtained for both even and odd (with respect to spatial inversion) states, showing that the energy dispersion of charge carriers consists of dimensionally quantized subbands. The energy gap between them increases with decreasing geometric dimensions of the structure, both in quantum wires and quantum dots made from monolayers of TMDs

It should be noted here that the energy dispersion of charge carriers in dimensionally quantized structures grown based on monolayers of transition metal dichalcogenides (TMDs) should depend on temperature and mechanical deformation due to changes in the width of the bandgap, both in bulk [18, 19, 20] and in low-dimensional [21] semiconductors. However, this case requires separate consideration.

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ТЕОРІЯ ЛІНІЙНО-КРУГОВОГО ДИХРОЇЗМУ В ОДНОАТОМНИХ ШАРАХ ДИХАЛЬКОГЕНІДІВ ПЕРЕХІДНИХ МЕТАЛІВ З УРАХУВАННЯМ ЕФЕКТУ РАБІ

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Ми розробили теорію розмірного квантування для наноструктур, як одновимірних, так і нульвимірних, побудованих з одноатомних шарів дихалькогенідів перехідних металів (ДПМ). Ця теорія дозволила нам отримати вирази для енергетичних спектрів носіїв заряду як у парних, так і в непарних станах (щодо інверсії координат), оскільки ці стани відбуваються в межах квантово обмежених ліній і точок моноатомних шарів ДПМ, залежно від їхніх геометричних розмірів. . Наш чисельний аналіз забезпечує детальне дослідження квантово-обмежених енергетичних станів електронів у цих наноструктурах, пропонуючи розуміння їхнього потенційного застосування в передових наноелектронних пристроях. Ця робота не тільки покращує наше розуміння енергетичних характеристик моноатомних шарів ДПМ, але й робить внесок у ширшу сферу матеріалознавства, досліджуючи вплив розмірного квантування на електронні властивості.

Ключові слова: *розмірне квантування; одно- та нульвимірна наноструктура; одноатомний шар дихалькогенідів перехідних металів; енергетична дисперсія; валентна зона; зона провідності*