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A study of acousto-electron effects in semiconductor quantum dots and bionanocomplexes based on them using deep machine learning

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An artificial neural network architecture has been developed that is capable of predicting changes in the energy spectrum of semiconductor quantum dots and their bionanocomplexes under the influence of an acoustic wave and interaction with human serum albumin molecules based on its specified geometric sizes, elastic constants and deformation potential constants of the allowed energy bands of quantum dot materials, as well as the frequency and amplitude of the ultrasonic wave and the surface concentration of human serum albumin. Two artificial neural network architectures have been implemented and optimized. Both models contain three hidden layers; however, the second architecture involves expanding the input space by adding harmonic functions. Both approaches to neural network modeling demonstrate good agreement with the results of mathematical modelling, provided that the input parameters lie within the training range. In the case when the input parameters lie outside the training sample, the model using additional harmonic functions at the input demonstrates a much better result. Within the framework of the developed model for the CdSe/ZnS/CdS/ZnS QD–human serum albumin bionanocomplex, the dependence of the energy shift of the radiation quantum on the frequency of the acoustic wave was investigated for different values of the surface concentration of albumin and different geometric sizes of the core and shell of the quantum dot.

Keywords: core-shell quantum dot, bionanocomplex, acousto-electron effect, deep machine learning.

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Introduction

Semiconductor nanoheterostructures containing quantum dots (QDs) exhibit high quantum efficiency of photoluminescence, which makes them promising materials for the creation of heterolasers [1], solar cells [2] and for biomedical applications [3, 4]. However, in practice, the realization of the potential of QDs is limited by a number of complex physical phenomena. In particular, the high surface area to volume ratio leads to an increased concentration of surface defects, which act as traps for charge carriers and cause non-radiative recombination [5]. In addition, charge transfer from QDs to organic molecules, such as proteins, reduces the radiation intensity [6].

Classical approaches to overcome these problems, such as surface passivation using shells (core-shell structures), do indeed allow to reduce the defect content and improve the stability of QDs [7, 8]. However, even

multilayer structures do not allow to completely eliminate interfacial defects or guarantee stable optical properties, especially in conditions of interaction with biological objects. In this regard, it is advisable to use additional external influences, in particular ultrasonic treatment, which, on the one hand, affects the formation of the surface structure of QDs, and on the other hand, activates acousto-electron effects.

Modern studies [8–11] indicate that acoustic deformations can cause periodic shifts of energy levels in QDs, changes in the band gap, modulation of radiation, and also affect charge transfer and interaction with biomolecules. Particular attention is drawn to ultrasonic treatment, which not only affects the morphological properties of QDs, but also initiates complex acousto-electron effects that are difficult to describe using classical analytical models [12, 13]. This necessitates the construction of self-consistent models that take into account not only the electron-deformation coupling, but

also the biochemical interaction, in particular with human serum albumin (HSA) [8]. To date, the possibility of conjugating semiconductor nanoparticles with protein molecules, for example, HSA molecules, has been experimentally tested [14].

Given the multifactorial character of these interactions and the complexity of their precise theoretical description, classical analytical or semi-empirical models often prove insufficient for an adequate analysis of the behavior of QDs under conditions of dynamic external influence. In this regard, the use of deep machine learning methods [15, 16], which allow to detect hidden nonlinear regularities in a large amount of experimental and simulation data, becomes relevant. Such approaches allow to take into account the influence of structural parameters, mechanical strains, acoustic perturbations and characteristics of the biomolecular medium on the energetic properties of QDs.

The aim of this work is to study acoustoelectronic effects in A^2B^6 core-shell semiconductor QDs and bionanocomplexes based on them using deep machine learning to model the effect of ultrasonic waves on the spectral properties of such structures. The proposed approach combines analytical modeling of multilayer QDs, consideration of elastic deformations induced by acoustic waves, and training of a neural network based on simulated and experimental data.

I. Model

Let us consider a core-shell QD that interacts with HSA molecules and is exposed to an acoustic wave. A mathematical model (MM) of a QD with a multilayered shell and a QD–HSA bionanocomplex exposed to an acoustic wave is presented in [8]. This model is based on solving a self-consistent system of mechanical equilibrium equations, the Poisson equation, and the Schrödinger equation with appropriate boundary conditions that take into account the influence of HSA molecules. We consider a spherical QD with a core radius R_0 and thicknesses d_i of the corresponding i -th layers of the shell, where $i = 1, 2, \dots, n$. The internal mechanical strains resulting from the mismatch of the lattice parameters $a^{(i)}$ of the materials of the core and shells, as well as the external pressure caused by the interaction with the HSA protein and the ultrasound wave lead to volumetric deformation of the core and shell layers. This deformation, in turn, changes the redistribution of electric charge and energy levels of QD due to the modification of the potential profile, which is taken into account when solving the Schrödinger equation. This model is effective, but very complicated and has many input parameters. The energy spectrum of charge carriers in QD or bionanocomplex based on them depends significantly on the concentration of HSA, elastic constants, the core radius, the structure and thickness of the shells, the frequency and amplitudes of the acoustic wave, as well as the concentration of electrons in QD. Due to the complexity of this mathematical model, there is a need to use deep learning models based on artificial neural networks to effectively analyze and predict the behavior of such systems.

In this work, the results of the numerical solution within the mathematical model were used to construct a sample of training data, which subsequently serves as the basis for training an artificial neural network and validating the results obtained within the neural network model.

II. Research methods

To model the dependence of the amplitude of the modulation of the radiation energy of the core-shell QD, which corresponds to the recombination transition between the ground states of the electron and hole, on the frequency of the acoustic wave, two different deep machine learning approaches based on artificial neural networks were used. The main goal was to construct a generalized model that allows predicting the amplitude of the modulation of the radiation energy ΔE of the QD from known geometric sizes (core radius and shell layers thicknesses), elastic constants (C_{11}, C_{12}), constants of the deformation potential of the conduction and valence bands ($a_c^{(i)}, a_v^{(i)}$), frequency ω and amplitude A_0 of ultrasound, surface concentration of HSA n_s : $\Delta E = f(R_0, d_i, C_{11}, C_{12}, a_c^{(i)}, a_v^{(i)})$. The optimization method used was the ADAM (Adaptive Moment Estimation) algorithm, an adaptive modification of stochastic gradient descent, with a batch size of 64. Training was performed on the CPU. The input data was split into training and validation sets in an 80/20 ratio. To ensure efficient training of the neural network, the input parameters, in particular acoustic wave frequency and HSA concentration, were normalized. The mean squared error (MeanSquaredLossLayer) was used as the loss function during training, which is a typical choice for regression problems in Wolfram Mathematica. A total of 10,779 unique training examples were used in the training process of the neural network, through which the network passed 274 epochs (iterations), which ensured comprehensive coverage of the parameter space and contributed to the convergence of the optimization process.

The architecture of the first neural network model (NNM) involved the use of a multilayer NetChain-type structure implemented in the Wolfram Mathematica, with three hidden layers. The hyperbolic tangent activation function was used to provide smooth nonlinearity and symmetric saturation. The first hidden layer contained 50 neurons, the second layer contained 150 neurons, and the third layer contained 250 neurons, which increases the network's ability to approximate complex functional dependencies.

The architecture of the second neural network model (NNM1) provided for the expansion of the input space. Namely, in order to increase the accuracy of approximation in areas of strong nonlinearity and take into account the possible harmonic structure of the function, the NNM1 model was expanded by adding sinusoidal dynamic harmonics to the input space. Due to the inclusion of harmonics, the model demonstrates improved ability to approximate periodic or quasi-periodic dependencies. The constructed models (NNM, NNM1)

were tested for configurations that were not included in the training set (with different shell thicknesses, different core radii, but with similar frequency ranges and HSA concentrations). The NNM1 model showed better generalization results and more accurate reproduction of the nature of the functional dependence in a wide range of input parameters. The study was conducted for CdSe QDs with core radii of 3, 5, 7 and 9 nm, and shell thicknesses $2a^{(\text{ZnS})}$, $4a^{(\text{ZnS})}$, $6a^{(\text{ZnS})}$ for a single-layer ZnS shell and with sets of thicknesses $(2a^{(\text{ZnS})}, 4a^{(\text{CdS})}, 2a^{(\text{ZnS})})$, $(2a^{(\text{ZnS})}, 6a^{(\text{CdS})}, 2a^{(\text{ZnS})})$, $(4a^{(\text{ZnS})}, 2a^{(\text{CdS})}, 5a^{(\text{ZnS})})$, $(5a^{(\text{ZnS})}, 4a^{(\text{CdS})}, 5a^{(\text{ZnS})})$ for a three-layer ZnS/CdS/ZnS shell in the ultrasonic wave frequency range $\omega = (0 - 0.3 \cdot 10^{12}) \text{ s}^{-1}$. For QD-HSA bionanocomplexes, a surface concentration of HSA molecules in the range of $n_s = (0 - 2 \cdot 10^{14}) \text{ cm}^{-2}$ was considered, using similar geometric size parameters for frequencies $\omega = 0.02 \cdot 10^{12} \text{ s}^{-1}$, $0.08 \cdot 10^{12} \text{ s}^{-1}$, $0.14 \cdot 10^{12} \text{ s}^{-1}$, $0.2 \cdot 10^{12} \text{ s}^{-1}$. All models showed errors within $(0.5 - 2)\%$ in the training range and $(0.8 - 7)\%$ outside it.

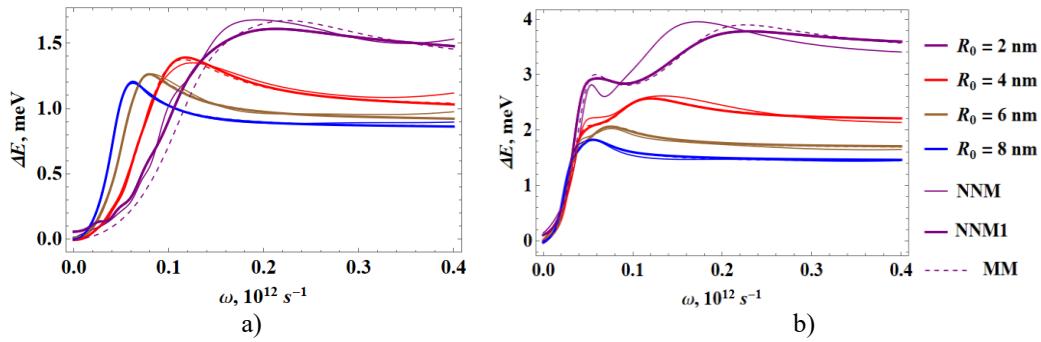


Fig. 1. Dependence of the modulation amplitude of the radiation quantum energy of a CdSe/ZnS QD, corresponding to the recombination transition between the ground states of the electron and the hole, on the acoustic wave frequency for different core radii and shell thicknesses: $d_1 = 3a^{(\text{ZnS})}$ (a); $d_1 = 15a^{(\text{ZnS})}$ (b).

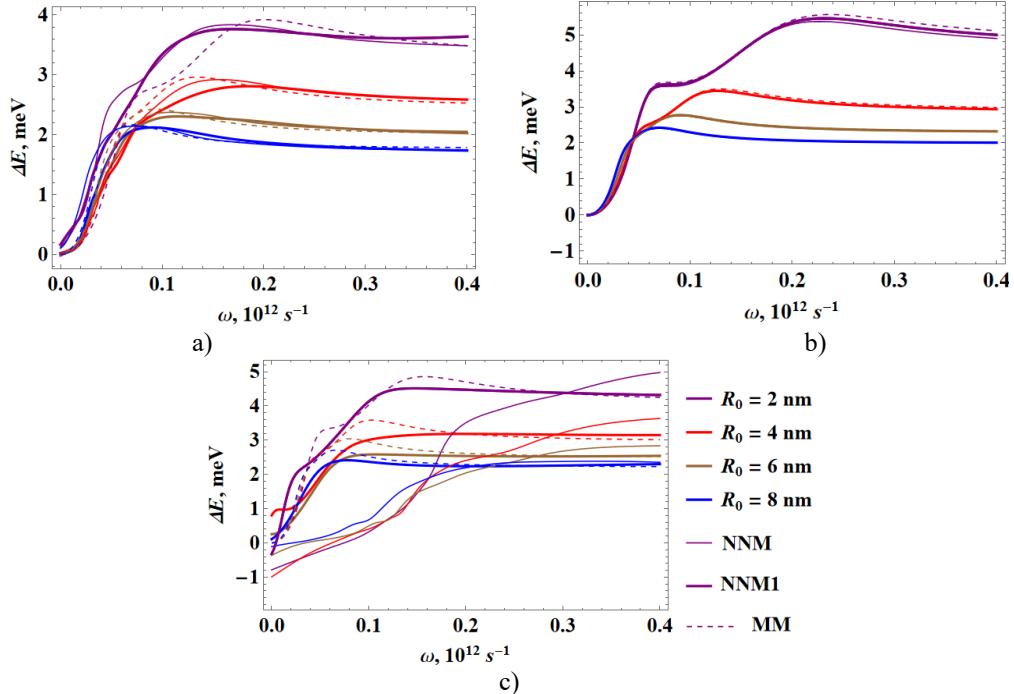


Fig. 2. Dependence of the modulation amplitude of the radiation quantum energy of a CdSe / ZnS/CdS/ZnS QD, corresponding to the recombination transition between the ground states of the electron and the hole, on the acoustic wave frequency for different core radii and shell thicknesses: $d_1 = d_3 = 3a^{(\text{ZnS})}$, $d_2 = 3a^{(\text{CdS})}$ (a); $d_1 = d_3 = 4a^{(\text{ZnS})}$, $d_2 = 4a^{(\text{CdS})}$ (b); $d_1 = d_3 = 6a^{(\text{ZnS})}$, $d_2 = 6a^{(\text{CdS})}$ (c).

III. Results and their discussion

During ultrasonic processing of core-shell QDs, periodic deformation caused by self-consistent electron-deformation coupling leads to local periodic shifts in the bottom of the conduction band and the top of the valence band, and, accordingly, to the modulation of the electron and hole energies [8]. Fig. 1, 2 show the results of calculations of the amplitude of the modulation of the radiation energy of CdSe QDs with a single-layer ZnS shell (Fig. 1) and a three-layer ZnS/CdS/ZnS shell (Fig. 2), which corresponds to the recombination transition between the ground states of the electron and hole, depending on the frequency of the acoustic wave at different core radii and shell thickness. The calculations were performed within the MM framework presented in [8] and using the NNM and NNM1 approaches. As we can see, both neural network modeling approaches demonstrate good agreement with the results of

mathematical modeling provided that the input parameters lie within the training range. In the case where both the core radius and the thicknesses of all shell layers lie outside the training sample, the NNM1 model demonstrates a much better result. This is especially evident for the three-layer shell with thick layers (Fig. 2c).

In the future, the NNM1 approach was used for neural network modeling. In particular, Fig. 3 shows the change in the radiation energy of the bionanocomplex depending on the concentration of HSA under the action of an

acoustic wave. Again, we observe good agreement between the results of neural network and mathematical modeling. The obtained results indicate that the constructed neural network has a high degree of generalization and can be used to study acousto-electron effects in QDs and bionanocomplexes based on them.

The developed NNM1 architecture was used to establish the regularities of the shift in the radiation quantum energy of the CdSe / ZnS/CdS/ZnS QD–HSA bionanocomplex, which corresponds to the recombination

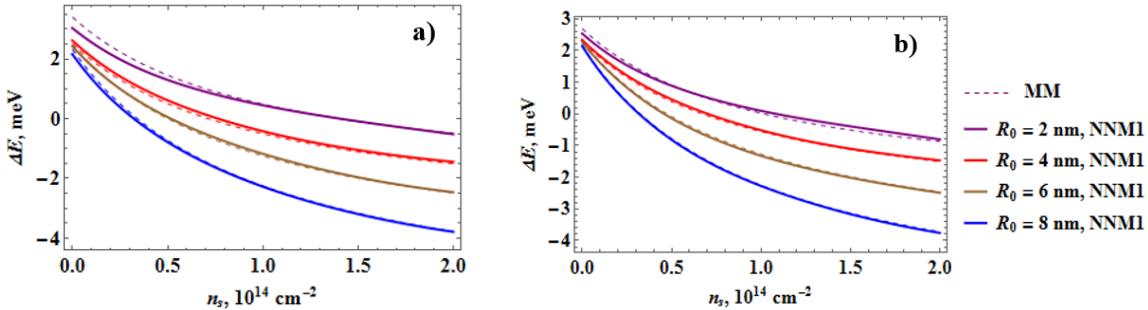


Fig. 3. Dependence of the shift in the radiation quantum energy of the CdSe / ZnS/CdS/ZnS QD–HSA bionanocomplex, corresponding to the recombination transition between the ground states of the electron and the hole under the influence of ultrasound ($\omega = 7 \cdot 10^{12} \text{ s}^{-1}$), on the surface concentration of albumin for different core radii and shell thicknesses: $d_1 = d_3 = 3a^{(\text{ZnS})}$, $d_2 = 5a^{(\text{CdS})}$ (a); $d_1 = d_3 = 3a^{(\text{ZnS})}$, $d_2 = 3a^{(\text{CdS})}$ (b)

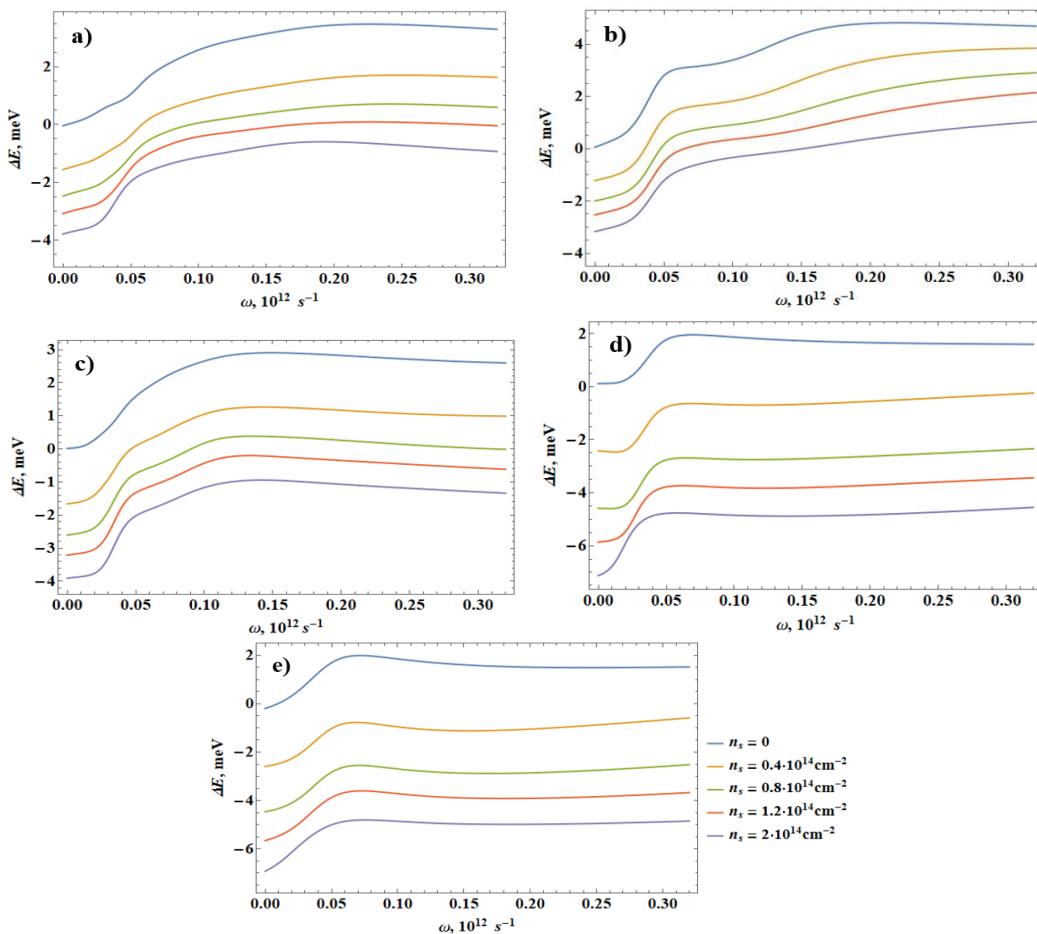


Fig. 4. Dependence of the shift in the radiation quantum energy of the CdSe / ZnS/CdS/ZnS QD–HSA bionanocomplex, corresponding to the recombination transition between the ground states of the electron and the hole under the influence of ultrasound ($\omega = 7 \cdot 10^{12} \text{ s}^{-1}$), on the acoustic wave frequency for different concentrations of HSA, core radii and shell thicknesses: $R_0 = 2 \text{ nm}$, $d_1 = d_3 = 2a^{(\text{ZnS})}$, $d_2 = 2a^{(\text{CdS})}$ (a); $R_0 = 2 \text{ nm}$, $d_1 = d_3 = 3a^{(\text{ZnS})}$, $d_2 = 6a^{(\text{CdS})}$ (b); $R_0 = 4 \text{ nm}$, $d_1 = d_3 = 3a^{(\text{ZnS})}$, $d_2 = 3a^{(\text{CdS})}$ (c); $R_0 = 8 \text{ nm}$, $d_1 = d_3 = 8a^{(\text{ZnS})}$, $d_2 = 8a^{(\text{CdS})}$ (d); $R_0 = 10 \text{ nm}$, $d_1 = d_3 = 3a^{(\text{ZnS})}$, $d_2 = 3a^{(\text{CdS})}$ (e).

transition between the ground states of the electron and hole, on the acoustic wave frequency at different concentrations of HSA, core radii and shell thicknesses (Fig. 4). The results of the research show that the influence of the interaction between QDs and HSA molecules, and the influence of the acoustic wave on the energy spectrum, are of opposite (mutually compensating) character. This leads to the fact that it is possible to select the albumin concentration at which the energy shift caused by acoustic oscillations is compensated. This fact is important, since both acoustic oscillations and HSA lead to a blurring of the energy spectrum.

The obtained results have potentially wide application in medicine. It is known that a number of diseases, in particular oncological, are accompanied by significant changes in the elasticity of cells (i.e., their elastic constants) [17]. In [18] it was shown that the presence of a QD shell significantly increases the deformation of the CdSe / ZnS/CdS/ZnS QD–HSA bionanocomplex. Accordingly, a change in the elastic constants of cell can affect the optical characteristics of QDs. In addition, in medical diagnostics, ultrasound-based research methods or irradiation with electromagnetic waves, which cause acoustic oscillations in QDs, are widely used. Therefore, the developed technology for determining changes in radiation energy due to deformation effects using artificial neural networks can be used as the basis for creating a new method for diagnosing cancer diseases based on deep machine learning. In addition, it is possible to formulate an inverse problem, namely, determining the concentration of HSA (human serum albumin) in order to quickly assess whether a bionanocomplex has been formed or not.

Conclusions

Acousto-electron effects in QDs with a multilayer shell and bionanocomplexes based on them play an important role in shaping their optical properties and in their interaction with biological objects. Given the high complexity of the mathematical model, as well as the multifactorial dependence of the energy spectrum of charge carriers in bionanocomplexes based on semiconductor QDs on the input parameters, this work implements an approach to studying acousto-electron effects based on deep machine learning. An artificial

neural network architecture has been developed that is capable of predicting the QD radiation energy based on their geometric sizes, elastic constants and deformation potential constants of the allowed bands of QD materials, surface concentration of HSA, and the frequency and amplitude of the acoustic wave. This approach enables rapid and accurate estimation of the energy spectrum of QDs interacting with biological objects, without the need to solve a complex system of equations for each new set of parameters.

Based on the developed model, two architectures of artificial neural networks of the NetChain type were implemented and optimized using the Wolfram Mathematica software product. Both models have three hidden layers, but the second architecture involves expanding the input space by adding harmonic functions. This made it possible to improve the accuracy of approximation of quasi-periodic dependencies. The obtained models demonstrate high accuracy (with an error not exceeding 7%) and good generalization ability, especially the model based on harmonic expansion.

Within the framework of the developed model of the CdSe / ZnS/CdS/ZnS QD–HSA bionanocomplex, the dependence of the shift in the radiation quantum energy on the acoustic wave frequency for different values of the surface concentration of HSA was investigated. It was found that the effects of the ultrasonic wave and the HSA protein on the energy spectrum are opposite in nature, which helps reduce the blurring of the energy spectrum caused by external factors (the acoustic wave, the interaction with HSA). During the study, it was confirmed that a QD with a three-layer shell is more sensitive to deformation than a QD with a single-layer shell.

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Дослідження акустоелектронних ефектів у напівпровідникових квантових точках та біонанокомплексах на їх основі з використанням глибинного машинного навчання

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Розроблено архітектуру штучних нейронних мереж, яка здатна прогнозувати зміну енергетичного спектра напівпровідникових квантових точок та біонанокомплексів на їх основі під впливом акустичної хвилі та взаємодії з молекулами сироваткового альбуміну людини на основі заданих її геометричних розмірів, пружних стальних та констант деформаційного потенціалу дозволених енергетичних зон матеріалів квантової точки, частоти та амплітуди ультразвукової хвилі, поверхневої концентрації сироваткового альбуміну крові людини. Реалізовано та оптимізовано дві архітектури штучних нейронних мереж. Обидві моделі мають три приховані шари, проте друга архітектура передбачає розширення входного простору внаслідок додавання гармонічних функцій. Обидва підходи нейромережевого моделювання демонструють добре узгодження з результатами математичного моделювання за умови, що входні параметри лежать в межах навчального діапазону. У випадку, коли входні параметри лежать за межами навчальної вибірки, то істотно кращий результат демонструє модель з використанням додаткових гармонічних функцій на вході. У межах розробленої моделі для біонанокомплексу квантова точка CdSe / ZnS/CdS/ZnS – сироватковий альбумін крові людини досліджено залежність зсуву енергії кванта випромінювання від частоти акустичної хвилі при різних значеннях поверхневої концентрації альбуміну та різних геометричних розмірів ядра й оболонки квантової точки.

Ключові слова: квантова точка виду ядро-оболонка, біонанокомплекс, акустоелектронний ефект, глибинне машинне навчання.