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Sensor Nanochips Based on Single Exciton Quasimolecules

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Abstract

A model of the sensor nanochip based on germanium/silicon heterostructures (Ge/Si-heterostructures) with Ge quantum dots is suggested. Two-Quasimolecule Spectrum: Infrared radiation can sense the formation of a single exciton quasimolecule. Optical transitions of this state to higher-lying SIE levels lead to the emission of radiation in the infrared part of the spectrum, with the energy of the emitting radiation being ~70 meV and its normalized intensity ~0.11. Such a unique type of infrared light can be considered as a strong signature in detection of single exciton quasimolecules in Ge/Si heterostructures. The sensor nanochip model to be proposed would should lay the foundation for both the fundamentals and applications, and the process could lead to the next generation of high-efficient sensor nanochip. Its successful fabrication would open a way for high performance exciton optoelectronic devices, highly sensitive, miniaturized, and CMOS-based infrared detectors for the biomedical, environmental and communication applications.

Article Info.

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Spatially Indirect Excitons, Double Quantum Dots, Single Exciton, Quasimolecules, Sensor Nanochips.

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1. Introduction

Currently, nanosystems with reduced dimensionality (in particular, quantum dots (QDs)) are used in nanotechnology for producing sensor nanochips [1–4]. If the dimensions of QDs does not exceed values of the order of 100 nm, then the physical (optical and electrical) properties of these nanosystems are mainly determined by the energy spectra of quantum-sized states of quasiparticles (electrons and holes) [5,6]. Depending on the size of the QDs, a finite number of quantum-sized energy levels of quasiparticles arose in single QDs [7,8]. The photoluminescence and absorption spectra in these nanosystems are formed by quantum transitions between quantum-sized states of quasiparticles. Moreover, the peaks of the photoluminescence and absorption spectra, as well as the emission intensity of the nanosystems, have a strong dependence on the size of the QDs [9]. Photoluminescence and absorption in these nanosystems were experimentally detected in the visible and infrared ranges [10–12]. Such nanosystems can be used in sensor nanochips. In these sensor nanochips, it is possible to observe dynamic processes caused by optical transitions between quantum-sized energy levels of quasiparticles in single QDs.

In the second type, Ge/Si heterostructures containing germanium QDs, spatial separation of electrons and holes was experimentally discovered. In this case, Spatially Indirect Excitons (SIE) appeared in the heterostructure [10–12]. In this case, the Coulomb interaction causes attraction between the electron and the hole. In the SIE, the electron is localized above the surface of the QD, and the hole is in the QD [5–13]. In these nanosystems, a gigantic increase in the binding energy of the SIE (by almost two orders of magnitude) compared to the binding energy of the exciton in silicon single crystals was

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theoretically shown [5–13]. From experimental studies [10–12], a photoluminescence signal was detected at room temperature in the infrared region of the spectrum

(0.20 to 1.14) eV **(1)**

In these nanosystems, the occurrence of SIEs was theoretically predicted [5], and single exciton quasimolecules were formed [7, 13]. The single exciton quasimolecule consisted of two interconnected SIEs. The binding energy of this single exciton quasimolecule was in the infrared region of the spectrum [7, 13].

In this paper, the possibility of detecting a single exciton quasimolecule in a sensor nanochip model is considered. The sensor nanochip model consists of two germanium QDs placed in a silicon matrix. In this sensor nanochip, a single exciton quasimolecule emits in the infrared range.

2. Formation of a Single Exciton and a Single Exciton Quasimolecule

In previous studies [5–7], a model of a nanosystem consisting of a spherical germanium QD with an average radius a and permittivity ε_2 = 16.3, placed in a silicon matrix with ε_1= 11.7, was considered. In germanium/silicon heterostructures with germanium QDs of the second type, the main electron level was in the silicon matrix, and the main level of holes was in the germanium QD. In this nanosystem, a hole with effective mass (($m_{h,2}/\ m_0$) = 0.39) moved in the valence band of the germanium QD, and an electron with effective mass (($m_{e,1}/\ m_0$) = 0.98) moved in the conduction band of the silicon matrix (m_0 is free electron mass) [10–12]. The localization of holes in the QD was due to a significant shift of the upper edge of the valence band ($\Delta E_{v(Ge)}$ = 700 meV) of germanium QD (relative to the upper edge of the valence band of the silicon matrix) [10–12]. In the heterostructure, the electrons moved in the silicon matrix, they did not penetrate the germanium QDs, since a substantial shift of the bottom of the conduction band ($\Delta E_{c(Si)}$= 340 meV) of the silicon matrix (relative to the bottom of the conduction band of germanium QDs) was observed on the interface (a QD – a silicon matrix). In this case, the electron-hole Coulomb interaction energy caused localization of the electron in a potential well above the QD surface. The appearance of SIE was found on the germanium/silicon heterostructure [10–12].

It has been shown that SIEs [5–7] and of exciton quasimolecules [7, 13] were formed in the Ge/Si heterostructure depending on the distance *D* between the surfaces of the QDs. If the average distance *D* between the surfaces of spherical germanium QDs significantly exceeded the Bohr radius of an electron (a_e = 0.63 nm) in a silicon matrix, i.e. at

$$D \gg a_e, \tag{2}$$

Then SIE-states with energies $E_{n=1,l}(a)$ (n and l are the main and azimuthal quantum numbers of the electron, respectively) are formed above the surface of a single QD [5–7]. The SIE Hamiltonian contained the electron-hole Coulomb interaction energy, the energy of the polarization interaction between the electron and the hole and the QD surface, and the centrifugal energy of the electron. The electron moves above the surface of the germanium quantum dot in an orbital described by a Coulomb variational wave function [5–7]

$$\varphi_{n=1,l}(r) = A exp\left(-\mu\frac{r}{a_{ex}}\right) \tag{3}$$

$$= \pi^{-1/2}\ a_{ex}{}^{-3/2}(2(\mu b)^2 + 2\mu b + 1)^{-1/2}\ \ exp(\mu b) \tag{4}$$

where *r* is the distance of the electron from the center of the QD, $\mu = \mu(a)$ is a variational parameter, and a_{ex} is the Bohr radius of 2D-SIE given as

$$a_{ex} = \tilde{\varepsilon}\,(m_0\,/\mu_{ex})\,(\hbar^2/m_0 e^2) = 2.6 \text{ nm} \quad \mathbf{(5)}$$

where $\mu_{ex} = m_{e,1}\, m_{h,2}/(m_{e,1} + m_{h,2})$ is the reduced SIE mass. $\tilde{\varepsilon} = 2\varepsilon_1\varepsilon_2/(\varepsilon_1 + \varepsilon_2)$ is the permittivity of the nanosystem, and $b = a/a_{ex}$.

Within the framework of the effective mass approximation, the energy spectrum $E_{1,l}(a)$ of quasistationary and stationary SIE-states (*n* =1, *l*), as a function of the average radius a of the germanium QD, is obtained by the variational method using the wave functions (Equations 3 and 4) [5–7]. The centrifugal energy of the electron in the SIE Hamiltonian causes the appearance of quasistationary SIE-states with energies $E_{1,l}(a) > 0$ in the nanosystem (the position of the bottom of the conduction band of the silicon matrix is taken as the zero energy level $E_{1,l}(a)$). Quasistationary and stationary states with energies $E_{1,l}(a) < 0$ forms a zone of surface SIE-states [5–7]. The band of stationary SIE-states $E_{1,l}(a)$ with a width

$$\Delta E_s = E_{ex}^{2D} = 2\hbar^2/\mu_{ex}^{2D}\,(a_{ex}^{2D})^2 = 82 \text{ meV} \quad \mathbf{(6)}$$

is located in the bandgap of the silicon matrix (where E_{ex}^{2D} is the binding energy of the two-dimensional SIE). The band of quasistationary SIE-states $E_{1,l}(a)$ with a width

$$\Delta E_{qs} = E_{n=1,l=3}^{max} \cong 248.4 \text{ meV} \quad \mathbf{(7)}$$

is located in the conduction band of the silicon matrix. In this case, the zone of surface SIE-states contains a finite number of levels (*n* = 1, *l* ≤ 3) [5–7]. For the occurrence of stationary and quasistationary SIE-states with energies $E_{1,l}(a)$ in a nanosystem, it is necessary to absorb a light quantum with an energy of [5–7]

$$\hbar\omega_{ex(1,l)}(a) = E_{g(Ge)} - \Delta E_{c(Si)} + E_{1,l}(a) \quad \mathbf{(8)}$$

In this case, a quasistationary SIE-state with energy $E_{1,l}(a) > 0$ appears in the conduction band and in the bandgap of the silicon matrix, a stationary SIE-state appears with energy $E_{1,l}(a) < 0$) [5– 7]. In Equation (8), the value $(E_{g(Ge)} - \Delta E_{c(Si)}) = 330$ meV (where $E_{g(Ge)}$ is the bandgap energy of the germanium QD, $\Delta E_{c(Si)}$ is the shift of the bottom of the conduction band of the silicon matrix relative to the bottom of the conduction band of germanium QD) [5–7].

It was found that the optical absorption in the nanosystem is caused by electron interband transitions between quasistationary and stationary states, as well as electron intraband transitions between stationary states [5–7]. Since the maximum value of the exciton energy is $E_{1,l}(a) \leq E_{n=1,l=3}^{max} \cong 248.4$ meV, then for the occurrence of SIE in a nanosystem, according to Equation (8), the absorption of a quantum of light with energy in the infrared range is necessary [5–7] such that

$$330 \text{ meV} \leq \hbar\omega_{1,l}(a) \leq 578.4 \text{ meV} \quad \mathbf{(9)}$$

Moreover, the energy range (9) is in the energy range (1), in which the SIEs were experimentally detected in the nanosystem [10–12].

A nanosystem model [7, 13] was studied in which double germanium QD(A) and QD(B) with radius a were placed in a silicon matrix. Holes h(A) and h(B) were located at the centers of QD(A) and QD(B). Electrons $e(1)$ and $e(2)$ were localized above the surfaces QD(A) and QD(B) in the silicon matrix, respectively (m_0 is the electron mass in free space). It was assumed that the electrons did not penetrate QDs.

With a decrease in the average distance D between the surfaces of germanium QDs to a value (of the order of $2a_e$), i.e. at

$$\mathrm{D} \leq 2\mathrm{a}_{\mathrm{e}}, \quad \textbf{(10)}$$

The distance between SIEs localized above the QD surfaces decreased to a size comparable to the Bohr electron radius. This caused a significant increase in the overlapping integral *S*(a, *D*) of the electron wave functions. A significant increase in the energy of the exchange interaction of electrons in this case caused the appearance of a coupled state of two SIEs in the nanosystem; i.e., an exciton quasimolecule appeared in the nanosystem.

Within the framework of the adiabatic approximation and the effective mass approximation, the Hamiltonian of the exciton quasimolecule containing Hamiltonians of the SIEs were localized above the surfaces QD(A) and QD(B), as well as the Hamiltonian, which took into account only the energies of the Coulomb interaction of the electron *e*(1) with hole *h*(B), and the electron *e*(2) with hole *h*(A), as well as than between electrons *e*(1) and *e*(2), and holes *h*(A) and *h*(B). The normalized wave function of the ground singlet state of the exciton quasimolecule was presented [7, 13] in the form of a symmetric linear combination of wave functions $\Psi_1\,(r_{A(1)}, r_{B(2)})$ and $\Psi_2\,(r_{A(2)}, r_{B(1)})$

$$\Psi_s(r_{A(1)}, r_{A(2)}, r_{B(1)}, r_{B(2)}) =$$

$$[2(1S^2(D,a))]^{-1/2}[\Psi_1(r_{A(1)}, r_{B(2)}) + \Psi_2(r_{A(2)}, r_{B(1)})] \quad (\mathbf{11})$$

where S(*D*, *a*) is the overlap integral of single-electron wave functions. The wave function of the ground singlet state of the exciton quasimolecule, Eq. (11) was written under the assumption that the spins of the electrons *e*(1) and *e*(2) were antiparallel. The wave functions $\Psi_1(r_{A(1)}, r_{B(2)})$ and $\Psi_2(r_{A(2)}, r_{B(1)})$ (Equation 7) were written as a product of single-electron wave functions $\varphi_{A(1)}(r_{A(1)})$ and $\varphi_{B(2)}(r_{B(2)})$, as well as $\varphi_{A(2)}(r_{A(2)})$ and $\varphi_{B(1)}(r_{B(1)})$, respectively. In equation 10, $r_{A(1)}$ is the distance of the electron *e*(1) from the QD(A) center, $r_{B(2)}$ is the distance of the electron *e*(2) from the QD(B) center, $r_{A(2)}$ is the distance of the electron *e*(2) from the QD(A) center, and $r_{B(1)}$ is the distance of the electron *e*(1) from the QD(B) center. In this case, the single-electron wave functions $\varphi_{A(1)}(r_{A(1)})$ and $\varphi_{B(2)}(r_{B(2)})$, as well as $\varphi_{A(2)}(r_{A(2)})$ and $\varphi_{B(1)}(r_{B(1)})$, were described by variational functions of the Coulomb type (Equation 3 and 4) [7, 13].

Within the framework of the variational method, at the first approximation, the total energy (9) of the ground singlet state of the exciton quasimolecule was determined by the average value of the Hamiltonian of the exciton quasimolecule for states, that were described by wave functions of the zeroth approximation $\Psi_s(r_{A(1)}, r_{A(2)}, r_{B(1)}, r_{B(2)})$ (10) [7, 13]

$$E_0(\widetilde{D}, b) = 2\,E_{n=1,l=0}(a) + E_b(\widetilde{D}, b) \quad \textbf{(12)}$$

where $E_b(\widetilde{D}, b)$ is the binding energy of the ground singlet state of the exciton quasimolecule, and $E_{n=1,l=0}(a)$ is the binding energy of the ground state (*n*=1, *l*= 0) of the SIE localized above the surface of QD [5- 7] (parameter $\widetilde{D} = D/a_{ex}$).

3. Radiation Intensity of Single Exciton Quasimolecule

The radiation intensity $I(a)$ caused by the optical transition of electrons from the ground energy level of single exciton quasimolecule to the ground energy levels of the SIE is determined the square of the overlap integral of exciton quasimolecule wave function $\Psi_s(r_{A(1)}, r_{A(2)}, r_{B(1)}, r_{B(2)})$(11) and of SIE wave functions $\varphi_{1,l}(r_{A(1)})$ and $\varphi_{1,l}(r_{B(1)})$, Eq.(3)

$$I(a) \approx \left| \int_a^\infty \varphi_{1,l}(r_{A(1)})\ \varphi_{1,l}(r_{B(1)})\ \Psi_s(r_{A(1)}, r_{A(2)}, r_{B(1)}, r_{B(2)})\ d^2r_{A(1)} d^2r_{B(1)}\ dr_{A(2)}\ dr_{B(2)} \right|^2 \quad \textbf{(13)}$$

SIE wave functions $\varphi_{1,l}(r_{A(1)})$ describes the SIE localized above the surface QD(A), and SIE wave functions $\varphi_{1,l}(r_{B(1)})$ describe the SIE localized above the surface QD(B). In this case, the wave functions $\varphi_{1,l}(r_{A(1)})$ and $\varphi_{1,l}(r_{B(1)})$ are described by Equation (3). After integrating Equation 13, taking into account (3), (4) and (11), an expression was obtained that describes the radiation intensity

$$I(a) \approx \pi^{-4}\ \mu^{-4}\ exp(-4\mu b)/\ (1 + S^2(D, a))(2(\mu b)^2 + 2\mu b + 1)^2 \quad \textbf{(14)}$$

4. Numerical Results and Discussion

The binding energy $E_b(\widetilde{D}, b)$ of the of the ground singlet state of the single exciton quasimolecule was obtained [16, 17] in the Ge/Si heterostructure consisting of double germanium QDs with mean radius $\bar{a}_1 = 12.8$ nm under the condition

$$\left(E_b(\widetilde{D}, b)/\ E_{1,0}(a)\ \right) \ll 1 \quad \textbf{(15)}$$

Such Ge/Si heterostructures was investigated in experimental works [7, 13].

The single exciton quasimolecule appeared in the nanosystem at distances $D \geq D_c^{(1)} \cong 2.1$ nm between the surfaces of QD [7, 13]. The binding energy $E_b(\widetilde{D}, b)$ of the single exciton quasimolecule ground state in a nanosystem with QD germanium of the mean radius $\bar{a}_1 = 12.8$ nm had a minimum $E_b^{(1)}(D_1, \bar{a}_1) \cong -6.1$ meV (at the distance $D_1 \cong 3.1$ nm) [7, 13]. In this Ge/Si heterostructure, the SIE binding energy was $E_{1,0}(\bar{a}_1) \cong -64$ meV [5-7]. The ground state energy of the single exciton quasimolecule (Equation 12) in this case was $E_0(\widetilde{D}_1, \bar{a}_1) \cong -134.1$ meV [7, 13]. In this case, condition 15 was fulfilled ($\left(E_b^{(1)}(D_1, \bar{a}_1)/E_{1,0}(\bar{a}_1)\ \right) \cong 0.09$). A single exciton quasimolecule with an increase in the distance D between the surfaces of the QD, so that $D \geq D_c^{(2)} \cong 4.4$ nm, decayed into two SIE [7,13]. In this Ge/Si heterostructure, the binding energy $E_b^{(1)}(D_1, \bar{a}_1) \cong -6.1$ meV of the ground singlet state of the single exciton quasimolecule significantly exceeded the binding energy of biexciton in a silicon single crystal by almost two orders of magnitude [7, 13].

The energy of the exchange interaction of electrons and holes was the major contribution to the binding energy of a single exciton quasimolecule in this Ge/Si heterostructure. This energy significantly exceeded the value of the Coulomb interaction between electrons and holes (i.e. the ratio ≤ 0.08) [7, 13]. In experimental works [10-12], in Ge/Si heterostructure, a blurring of the absorption edge was observed in the infrared wavelength range (1) up to room temperatures. Such blurring of the absorption edge was caused by interband electron transition in SIE [5-7]. The radiation intensity $I(a)$ (Equation

14) caused by the optical transition of electrons from the ground energy level $E_0(\tilde{D}_1, \bar{a}_1) \cong -134.1$ meV of single exciton quasimolecule to the ground energy levels $E_{1,0}(\bar{a}_1) \cong -64$ meV of the SIE in a nanosystem with QDs germanium of the mean radius $\bar{a}_1 = 12.8$ nm was ofthe value $I(\bar{a}_1) \approx 0.11$. The energy of this transition is (≅70 meV) in the infrared range of the spectrum.

5. Conclusions

Germanium /silicon heterostructure containing germanium QDs as a model of a sensor nanochip was proposed. In this model, containing germanium QDs with an average radius $\bar{a}_1 = 12.8$ nm and a distance $D \cong 3.1$ nm between the QDs surfaces, there is a single exciton quasimolecule. The ground state energy of the single exciton quasimolecule was $E_0(\tilde{D}_1, \bar{a}_1) \cong -134.1$ meV. As a result of the optical transition of electrons from the ground energy level of this single exciton quasimolecule to the energy levels of the SIE, radiation with an intensity of $I(\bar{a}_1) \approx 0.11$ in the infrared range of the spectrum (≅70 meV) is observed. This radiation allows the detection of single exciton quasimolecules in sensor nanochips. The author hopes that the development of this model by experimenters will make it possible to create a new generation of efficient sensor nanochips based on germanium/silicon heterostructures with germanium QDs.

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Conflict of Interest

Authors declare that they have no conflict of interest.

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رقائق استشعار نانوية مبنية على جزيئات إكسيتونية أحادية

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الخلاصة

يُقترح نموذجٌ لرقاقة استشعار نانوية مبنية على بنى غير متجانسة من الجرمانيوم والسيليكون (بنى غير متجانسة من الجرمانيوم والسيليكون) مع نقاط كمية من الجرمانيوم. طيف شبه الجزيء ثنائي الجزيء: يمكن للأشعة تحت الحمراء استشعار تكوين شبه جزيء إكسيتون واحد. تؤدي الانتقالات الضوئية لهذه الحالة إلى مستويات SIE أعلى إلى انبعاث إشعاع في الجزء تحت الأحمر من الطيف، بطاقة الإشعاع المنبعث حوالي 70 ميجا فولت وشدته المعيارية حوالي 0.11. يمكن اعتبار هذا النوع الفريد من ضوء الأشعة تحت الحمراء بمثابة بصمة قوية في الكشف عن شبه جزيء إكسيتون واحد في بنى غير متجانسة من الجرمانيوم والسيليكون. من المفترض أن يُرسي نموذج رقاقة الاستشعار النانوية المقترح الأساس لكل من الأساسيات والتطبيقات، وقد تؤدي هذه العملية إلى الجيل التالي من رقائق الاستشعار النانوية عالية الكفاءة. إن نجاح تصنيعها من شأنه أن يفتح الطريق أمام إنتاج أجهزة بصرية إلكترونية عالية الأداء وأجهزة كشف الأشعة تحت الحمراء شديدة الحساسية والمصغرة والمعتمدة على CMOS للتطبيقات الطبية الحيوية والبيئية والاتصالات.

الكلمات المفتاحيه: الإكسيتونات غير المباشرة مكانيًا، النقاط الكمومية المزدوجة، الإكسيتون المفرد، الجزيئات شبه الذرية، رقائق الاستشعار النانوية.