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Hierarchy of excitonic formations

Abstract

In the present paper we suggest the new fractal model for the description of energetic spectra of excitons and multi-excitons in amorphous, porous nanocluster semiconductors. Because of theirs chaotic structure the well-known analogy between excitons and hydrogen-like atoms is insufficient. Analysis of recent experimental data shows that nanostructured semiconductor films characterized by fractal self-affine and self-similar structure. These properties haven't been described by well-known theoretical models.

We derive the new equations for the description of energy of excitons and multi-excitons depending on energy of exciting photon. In the theory we have taken into account the nonlinear dependence of bandgap of a semiconductor on photon energy. On the base of the suggested theory we obtain spectra of excitons and multi-excitons in nanostructured films. Also we study influence of value of bandgap, binding energy of excitons and fractal dimension of a set of current carriers on spectra of excitons, biexcitons and trions. Comparison of the results of our theory to the recent experimental data devoted to study of spectra of excitons and biexcitons localized in quantum dots is also given in the paper. Theory shows the existence of most universal regularities of hierarchical systems.

Key words: Fractal measure, excitonic formations, hierarchical structures, photon energy, exciton-biexcitonspectra, coherent oscillators.

Introduction

Excitons and exciton formations such as biexcitons and trions can be used for distinguishing rapidly-changing information signals. So, excitons are the subject of many studies on nanoelectronics. Overlapping of wave functions of electrons and holes is probable in nanoclustered semiconductors. Therefore, exciton binding energy in such semiconductors is higher than in infinite homogeneous medium. These facts suggest a possibility of generation of excitonic quantum bits at sufficiently high, even at room temperatures.

Generally, due to the specific character of technological processes (implantation, diffusion-limited aggregation) nano-sized semiconductors have irregular, chaotic structure. Here, such semiconductors are characterized by fractal regularities on a small range of scale of measurements [1]. Therefore, properties of excitons cannot be described universally via smooth regularities which follow, for example, from differential equations. So, a common analogy between exciton and hydrogen-like atom is inapplicable for a full description of specifics of excitonic spectra in noncrystalline semiconductors with chaotic structure.

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Such conclusions follow also from experimental work [2-5] devoted to the description of certain self-similarity, quantum-mechanical coherence and decoherence between an exciton and biexciton. Therefore, we ask the natural question whether spectra of exciton energy possess properties of fractal curves.

The aim of this paper is to develop the new model for thedescription of nonlinear fractal evolution of excitons in dependence upon stimulating photon energy and comparison of results with available experimental data.

Nonlinear fractal measures

The basic properties of fractals are their self-similarity and dependence of measure on the scale of measurements. We mean that measure is a physical value characterized by an additive measurable set. For example, measures of a geometrical fractal are its length, square and volume. Nano-objects have surprising variety of physical properties because their measures depend on their values according to nonlinear laws. This fact brings out clearly the necessity of fractal analysis in nanoscience.

According to well-known fractal theories a minimal scale of measurement (size of cells covering an object) should be chosen independently on value of defining measure. For the description of evolution of measure in dependence upon given order parameter which is

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determining variable of a physical process, we choose the scale of measurement via this parameter and desired measure. Hence, fractal measure is a nonlinear function depending on the process.

The conventional definition of fractal measure M can be written as

$$M = M_0 \left(|\Delta M| / M_* \right)^{-\gamma}, \gamma = D - d, \gamma > 0, \quad (1)$$

where M_0 is a regular (non-fractal) measure, ΔM is a scale of measurements, M_* is norm of M, D is fractal dimension of the set of values of M, d is topological dimension of the measure carrier. ΔM is independent of M, therefore, measure defined by (1) can be tentatively called the linear value.

If λ is taken as order parameter, then we can choose ΔM as

$$\Delta M_M = \frac{|M-\lambda|}{M} = \left|1 - \frac{\lambda}{M}\right|,$$
$$\Delta M_\lambda = \frac{|M-\lambda|}{\lambda} = \left|1 - \frac{M}{\lambda}\right|, \quad (2)$$

where indices M and λ correspond to the norms ΔM. According to (2) we can rewrite the formula (1) as

$$M_M = M_0 \left(\left| 1 - \frac{\lambda}{M} \right| \right)^{-\gamma}, \; M_\lambda = M_0 \left(\left| 1 - \frac{M}{\lambda} \right| \right)^{-\gamma}. \quad (3)$$

At $\gamma \to 0$ we have $M_M = M_\lambda = M_0$, this corresponds to the meaning of M_0. At $\lambda = 0$ we have $M_M = M_0$, $M_\lambda = 0$. It means that the fractal measure defined by its own norm exists in the case when external influence characterized by parameter λ is absent.

Values of γ are known for self-similar (similarity coefficients of respective variables are equal) and self-affine (similarity coefficients are different) sets. Normalized values of information entropy for self-similar and self-affine sets are equal $I_2 = 0.806$ and $I_1 = 0.567$ respectively [1]. Fractal dimension of secant of surface of a chaotic object embedded to space with topological dimension $\tilde{d}$ is equal to

$$D = \tilde{d} - I, \quad I = (I_1, I_2). \quad (4)$$

Thus,

$$\gamma = \tilde{d} - d - I, \; 1 - I \le \gamma \le \tilde{d} - I, \; \tilde{d} = 2,3. \quad (5)$$

We apply equations (3) to the description of excitonic spectrum. Suppose that we have an electron in conduction band and a hole in valence band in a semiconductor with bandgap E_g. Effective mass of electron is m_e, effective mass of hole is m_h. The rest particles produce a background with dielectric permittivity ε. We assume that interaction between these two quasi-particles occurs according to Coulomb's law. So, by using the Schrödinger equation for hydrogen atom, we obtain the equation for full energy of electron-hole pair:

$$E_n\left(\vec{k}\right) = E_g + \frac{h^2 k^2}{2\left(m_e + m_h\right)} - \frac{m_e m_h e^4}{2\varepsilon^2 \hbar^2 n^2 \left(m_e + m_h\right)},$$
$$n = 1,2,3 \ldots \quad (6)$$

We discard energy of motion of an exciton "as a whole". Hence, exciton quasi-momentum $\vec{k}$ is set to zero. Let us denote binding energy of electron and hole in exciton (the last term in equation (6)) as E. We consider the case of excitation of electron by photon with energy $\hbar w$ sufficient for electron transition to valence band. So, we can rewrite equation (6) as

$$\hbar w - E_g = E. \quad (7)$$

Using the new simplified notations $M_M = M = E_1$, $M_0 = E_0$, $\lambda = \hbar w - E_g$, $M_\lambda = E_{1,w}$, we obtain the from equations (3)equation for energy of single exciton E_1:

$$E_1 = E_0 \left(\left| 1 - \frac{\hbar w - E_g}{E_1} \right| \right)^{-\gamma} \equiv f\left(E_0, E_1\right),$$
$$E_{1,w} = E_{0,w} \left(\left| 1 - \frac{E_{1,w}}{\hbar w - E_g} \right| \right)^{-\gamma} \equiv f\left(E_{0,w}, E_{1,w}\right). \quad (8)$$

Here E_0 is exciton energy at excitation threshold when energy of exciting photon is $\hbar w = E_g$. In this case $E_{0,w} = 0$.

We can describe biexcitons, trions and other clusters as hierarchical structures:

$$E_n = f\left(\ldots f\left(\frac{E_0}{n}, E_n \right) \ldots \right), \quad (9a)$$
$$E_{n,w} = f\left(\ldots f\left(\frac{E_{0,w}}{n}, E_{n,w} \right) \ldots \right), n = 1,2,\ldots, \quad (9b)$$

where number of brackets is equal to n. Equation (9*a*) correspond to the choice of scale of measurements of fractal measure relative to the measure and describe evolution of excitons, biexcitons and other structures existing in the ground state, i.e. without external radiation. Equations (9*b*) correspond to the choice of scale of measurements of fractal measure relative to photon energy and describe excited states only. Equations (8), (9*a*), (9*b*) determine energy of the system consisting of excitonic formations:

$$E = \sum_{i=1}^{n} E_i, \; n = 1,2,\ldots \quad (10)$$

Here $n = 1$ describes an exciton, $n = 2$ describes a biexciton, $n = 3$ describes a trion and so on.

In the simplest case, at the absence of nonradiative transitions in a semiconductor with intrinsic conductivity

photoluminescence intensity and absorption coefficient $\alpha(w)$ are defined via density of number of states. So,

$$\alpha(w) = \alpha\left(E_g/\hbar\right)E(w)^{\frac{1+\gamma}{2}}. \qquad (11)$$

Formula (11) includes γ in order to take into account fractality of impulse space for which we determine density of states. In this way, we can take into account influence of external field P on relative photoluminescence intensity of biexciton $\alpha_2(P)$ and exciton $\alpha_1(P)$ as

$$\alpha_2(x,w) = \alpha_2(w)\left(\frac{\left\langle \alpha_1(w)^2 \right\rangle}{\left\langle \alpha_2(w)^2 \right\rangle}\right)^{1/2} th(x)^{\frac{1+\gamma}{2}}, \qquad (12)$$

$$x = \frac{P}{P_0}.$$

Here P_0 is relative minimal intensity for saturation of $\alpha_2(x,w)$. Function $th(x)$ is used for accounting of the well-known expression for variation of number of coherent oscillators; angle brackets denote frequency averaging.

Results of numerical analysis

Let us describe exciton-biexciton spectra in nanostructured semiconductor films by use of equations (8)-(10). By varying the parameters E_g, E_0, γ, it is possible to obtain different types of excitonic spectra.

Fractality leads to increase of amplitude of oscillations and extension of spectra (Figure 1). We use self-similar $(1-I_2)$ and self-affine $(1-I_1)$ values of fractal dimensions. Value of energy gap defines location of oscillations area, i.e. range of exciton, biexciton and trion energies. Line 1 in Figure 1 corresponds to exciton spectrum, lines 2 and 3 correspond to biexciton and trion spectra respectively. Lower boundary of oscillations area is exceeds E_g by the value which has the order of E_0.

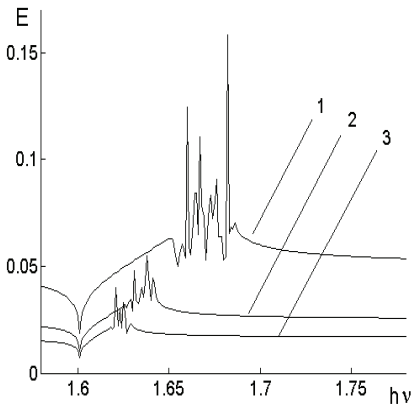


Figure 1 - Spectra of excitons, biexcitons and trions with

$$E_g = 1.6eV,\ E_0 = 0.05eV,\ \gamma = 1 - I_2$$

Figure 2 shows models of multiexciton spectra calculated as summation of spectra of excitons, biexcitons and trions by use of the formula (9(a)). Comparison of the figures 2(a) and 2(b) shows that multiexciton spectra strongly depend on fractal dimension γ. Increase of E_0 leads to broadening of oscillation area towards greater photon energy and increase of the amplitude of oscillations. Increasing of energy gap width E_g leads to broadening of oscillation area and increase of their amplitude.

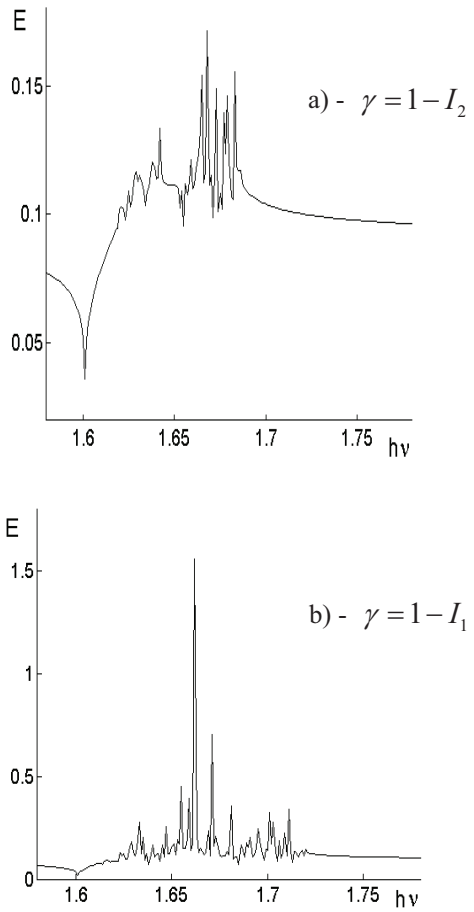


Figure 2 - Multiexciton spectra with $E_g = 1.6eV$, $E_0 = 0.05eV$

Comparison of theoretical results to experimental data

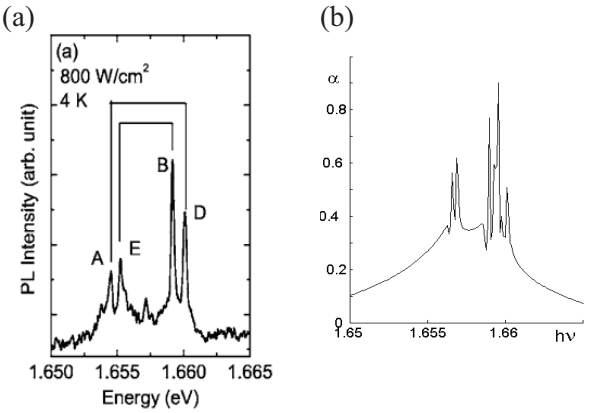
Figure 3(a) shows experimental exciton photoluminescence (PL) spectrum from InGaAs quantum dots fabricated by metalorganic vapor epitaxy [4]. The sample has two quantum dot layers. Separation between layers is about 5 nm. Figure 4(b) shows results of simulation by equations 9(a), (10) and (11).

Figure 4(a) shows exciton-biexciton spectrum of a quantum dot recorded at 4 K [5]. The quantum dot was fabricated on a GaAs(311) substrate by metalorganic vapor epitaxy. The quantum dot were formed by self-

assembly process. Each peak of PL spectrum corresponds to an excited state of exciton or biexciton. Figure 4(b) illustrates the exciton-biexciton spectrum, which was calculated by use of equations 9(a), (10) and (11). Curves in Figures 3(b) and 4(b) are plotted in steps of $\Delta\hbar w = 10^{-4}$ eV.

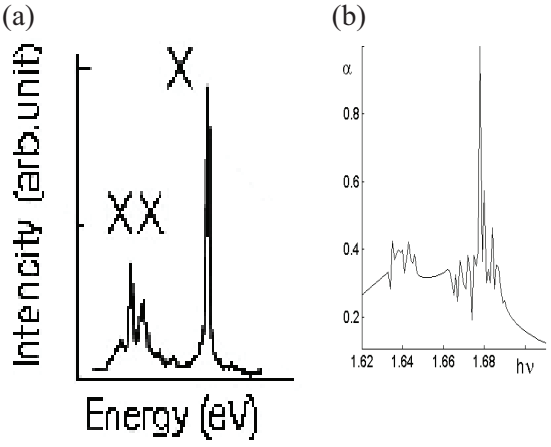
Discussion

Shift of exciton and biexciton spectra by photon frequency is observed in theoretical figures at increasing of E_0, E_g. Exciton peaks in theoretical curves are seen in ranges about 10^{-3} eV. These facts correspond to



(a) - Experimental data [4]
(b) - numerical results
$E_g = 1.65\ eV, \gamma = 1 - I_2,\ E_0 = 4 \cdot 10^{-3}\ eV$

Figure 3 - PL spectrum from bilayer InGaAs quantum dots



(a) - Experimental data [5]
(b) - numerical results
$E_g = 1.60\ eV, \gamma = 0.244,\ E_0 = 5 \cdot 10^{-3}\ eV$

Figure 4 - Exciton-biexciton PL spectrum from the isolated InGaAs quantum dot

experimental data. If E_0, E_g, γ are chosen according to the conditions of experiment, there is good agreement between theoretical and experimental results. Amplitudes of exciton spectra and spectra of excitonic formations correspond to experimental data.

It is worthy of notice that in experiment [5] biexciton forms around a quantum dot. Region of its localization it not limited artificially. Biexciton can be formed between two quantum dots [4]. Distance between quantum dots is about 5 nm, i.e. strong localization stimulates formation of exciton at small E_0. These facts also correspond to the theory.

Our theory can be applied for the description of influence of external field on structure of an excitonic formation. Amplitude of an exciton stronger depends on external field than amplitude of a biexciton. Also from our theory follows that decoherence between excitons and biexcitons is stronger at linear polarization of external radiation than at circular polarization.

So, fractal model of the dependence of electron-hole pair energy on energy of exciting photon can be used for the description of coherence between exciton formations. The terms “quantum-mechanical coherence” and “coherence of wave processes of different nature” have diverse meanings. Quantum-mechanical coherence means the time interference between wave functions only. It appears as energy oscillations in a system. Generally, coherence means that phase difference is constant by some determining variables at subsystem evolution. In excitonic formations coherence can be observed in these two types.

Comparison of the theoretical results with experimental data confirms that basic regularities of exciton and biexciton spectra can be described within a framework of our theory.

Conclusion

In the present work we suggest the new equation for fractal measure depending on itself as scale of measurements. This measure correctly describes a hierarchy of excitonic formations. The theory describes conditions for initiation of oscillations and for energy peaks coherence of exciton formations. These new ideas can find wide applications in modern nanoelectronics.

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