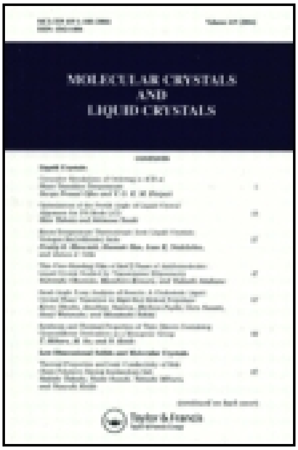

Molecular Crystals and Liquid Crystals

Synthetic and Biological Functional Compounds with Direct Excitons Conductivity for Nanoelectronic Devices

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Synthetic and Biological Functional Compounds with Direct Excitons Conductivity for Nanoelectronic Devices

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The design, synthesis, and investigations of the special functional compounds containing several (up to 12) π-electron systems are done. The results of spectral investigations of the synthetic nucleotides-containing compounds are compared with the results obtained on DNA. The predicted processes of transfer of direct triplet electronic excitations in all the investigated functional compounds are proved. This fact gives the possibility to propose these compounds to be used for the design of nanoelectronic devices. The computer simulations of electronic excitations passing through an oligomer functional macromolecule taking into account the reverse exciton currents and the multipath excitation energy migration show these macromolecular compounds are perspective to be applied in nanoelectronics.

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Keywords: DNA; exciton currents; nanoelectronics; triplets

1. INTRODUCTION

Recently in the world of modern nanoelectronics, the problem of development of the basic elementary functional units appears and requires for new technological solutions [1–14]. In this connection, organic π-electron-containing functional macromolecules with the known predetermined energy structure and, as a result, the properties of predicted one-way exciton conductivity have been proposed as the basic elements for nanoelectronics [14]. The combining of such elements in some logic structures is the way to the creation of advanced technology nanoelectronic devices. Here, we present our attempts to design, synthesize, and investigate some functional macromolecules for nanoelectronics or, to be more correct, for nanophotonics, as it has been pointed out by Prof. P. N. Prasad at the International Conference "Photon-Based Nanoscience and Technology" [14].

From our point of view, the non-conjugated π-electron-containing organic polymers are most perspective for nanoelectronic (and nanophotonic) applications. Contrary to conjugated polymers, in which π-electrons are delocalized along macromolecules (e.g., polyphenylvinylene), the electronic energy sites of non-conjugated polymers (e.g., polysterene, polyvinylnaphthalene, polyvinylcarbazole, RNA, DNA) or copolymers are determined mainly by peculiarities of the energy structure of the π-electron systems of individual polymer cells [15–22,26,27]. The weak interaction between neighbour π-electron systems in a macromolecule leads to the migration of electronic excitations (singlet and triplet excitons). These properties of macromolecules of such a type give the possibility to realize local changes in some macromolecular cells without essential perturbation of the energy structure of other cells (Fig. 1). On this way the design of functional macromolecules can be developed.

The excitonic processes in macromolecules have been studied by us [4,5,15–28] for a long time. The results of investigations of the electronic excitation energy transfer in macromolecules manifests that the speading of excitons in macromolecules is a random walk in character [15–18,24–26]. We also have evaluated some characteristics of singlet and triplet excitons in macromolecules (the spreading length, the number of jumps, the critical jump length [15,17,18,24–26]). The resulting displacement of excitons in a macromolecule is considerably less than their total way during the life-time. The question arises

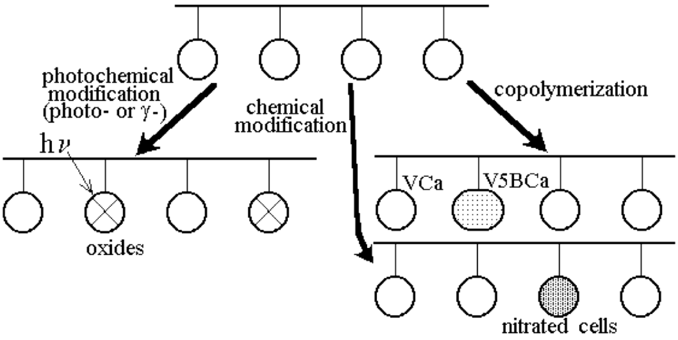


FIGURE 1 Different ways of modifications of the composition of macromolecules.

whether it is possible to create a macromolecule with direct one-way exciton motion, in which the energy transfer could be realized by following steps: 1) Absorption of a photon by the first link *only*; 2) one-way direct excitation energy transfer; 3) localization of an excitation in the end link *only*. Namely such a macromolecule could act as a one-way exciton conductor.

The independence of the energy structure of elementary polymer cells allows one to modify the composition of macromolecules without essential influence on basic unit energy sites. There are different ways of modification: a) chemical or photochemical modification of elementary polymer cells; b) copolymerization of π-electron-containing monomers ("active units") with monomers which do not contain such a system ("neutral units") (Fig. 1). Such a modification gives a possibility to create model macromolecules for unique experiments.

2. MAIN IDEAS AND MODELS

The idea of unidirect excitation energy transfer was proposed by R. Kopelmann for *j*-aggregates [29] and was applied for dendrimers (see, e.g., [30,31]). In these works, the direct singlet excitation energy transfer could be realized due to a displacement of suitable singlet energy levels. In all the systems proposed, the singlet levels of macromolecule cells fall down along the macromolecule chain. The disadvantage of this model is in the fact that photons exciting the highest singlet level can excite all cells of a macromolecule.

Our model of a macromolecule with unidirect triplet excitation motion was proposed by the co-author of this article, Prof. V. M. Yashchuk in 1995 [5,32] and developed in details later on

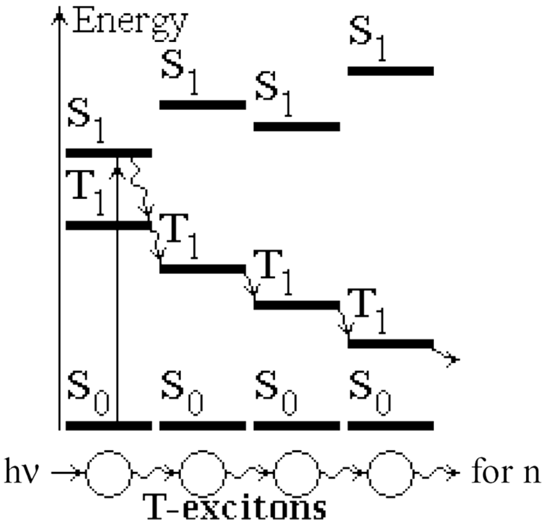


FIGURE 2 Best variant for the displacements of energy levels for a macromolecule with the direct triplet excitation energy transfer.

[4,20,21,33,34]. The team of chemists headed by Prof. V. G. Syromyatnikov has realized this model in a number of molecular compounds [4,19,23,32–34]. The proposed scheme of energy levels (Fig. 2) allowed one to excite the first macromolecular cell without excitation of other cells. Then the nonradiative transition $S_1 \rightarrow T_1$ in the first cell with the following unidirect triplet excitation energy transfer from the first cell to the n-th cell through the macromolecule has to be realized.

In [5,19], the definition of exciton current in a functional macromolecule was done.

Due to the possible deactivation processes running in each cell, the value of the exciton current has to depend on the number of cells in a macromolecule. According to [4,5,19], if one neglects the reverse current, the direct current between the n-th and $(n+1)$-th cells equals

$$I \approx I_0 \cdot (1-\alpha)^n,$$

where α is the probability of excitation deactivation in an elementary unit of the macromolecule. For triplet states of aromatic-containing macromolecules with effective intramolecular energy transfer, this probability is $\alpha \sim 10^{-5}$. Then, for $n = 10^4$, $I/I_0 \approx 0.9$. So, 90% of excitons initially generated in the first cell will reach the 10^4-th cell.

The problems of the composition of such functional macromolecules were discussed in [19–21,23,26,33,34]. The first cell selection, the optimal distance between neighbor cells, the connection between the π-electron-containing macromolecular cell structure and the singlet-triplet splitting were among the main problems on the way to the creation of macromolecules of the proposed type. In [4,5,19,21–23], the first steps to the construction of functional macromolecules possessing the proposed energy structure were done.

Another model proposed is a nonlinear branched macromolecule, each branch of which has the same energy structure as that in the model described above. On the first sight, such a macromolecule looks like a dendrimer [31], but there is the essential difference between the dendrimer branching and the branching of our model which is related to a special hierarchy of energy levels in our model that causes the absorption of a photon only in the first link and the long-distance triplet exciton transport to the end cell [19].

In macromolecules of the third model, the system of link energy levels is similar to those of models 1 and 2. However, the electron affinity increases from cell to cell along a macromolecule [19]. This leads to that not only the excitation energy transfer but the photoinduced charge transfer complex transport along a macromolecule from the first to the end link can take place.

In this article, the results of the design and investigations of the functional compounds that are the short synthetic DNA fragments and possess the properties of one-way direct triplet excitation conductivity, as well as the results of measurements of the electroconductivity of some compounds possessing the properties mentioned above, are presented.

3. DESIGN, SPECTRAL INVESTIGATION, AND TESTING OF THE π-ELECTRON-CONTAINING FUNCTIONAL COMPOUNDS

3.1. The Design of Functional Compounds on the Base of the π-Electron Systems of Nucleotides

A number of the sequences of compounds with a hierarchy of energy levels that is suitable for the design of functional macromolecules was selected [19–22]. The scheme of energy levels of one of these sequences (that contains nucleotides: deocytidine (dCMP), deoguanosine (dGMP), thymidine (dTMP), and deoxyadenosine (dAMP)) is given in Figure 3. It is worth to stress that these π-electron systems are contained in such important macromolecule as DNA.

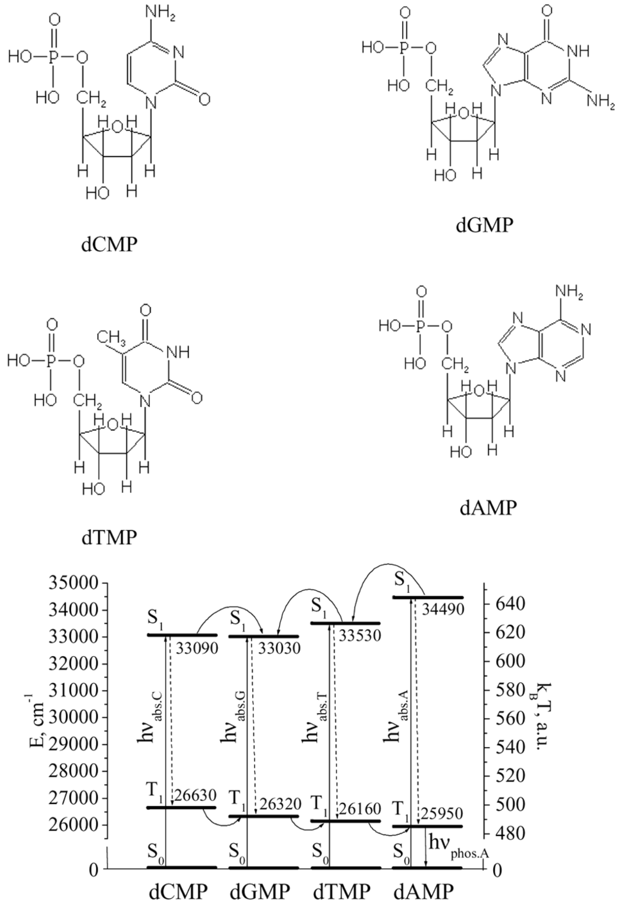


FIGURE 3 Chemical formulae and the scheme of energy levels of the sequences that contain nucleotides.

The test for functionality: the phosphorescence of dAMP must be observed ($\lambda \approx 385\,\text{nm}$) only under the excitation of dCMP ($\lambda \approx 300\,\text{nm}$).

3.2. The Main Model Realization and Investigations of the Electronic Excitation Energy Transfer

Some functional molecular systems with direct energy transfer which already have been synthesized [4,19–21,27] are presented below.

System 1. Basic units – naphthalimide (NAPHMA) and aminophenole (APH), their mutual system – dimer – oxyphenylnaphthalimide,

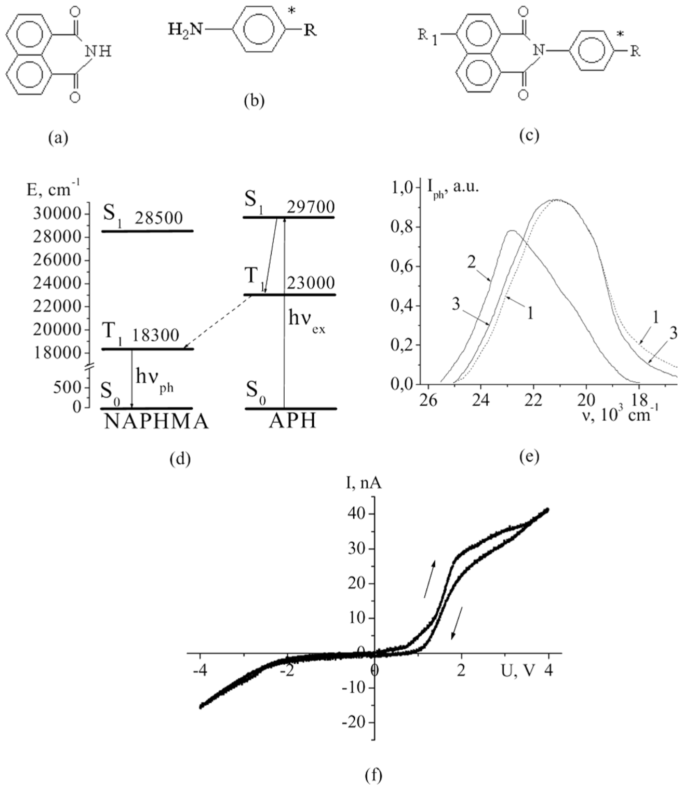


FIGURE 4 Structure formulae of naphthalimide (NAPHMA) (a), aminophenole (APH) (b), dimer – oxyphenylnaphthalimide (c) (when R–H or OH; R_1–NO_2, H, NH_2; *-π-, m-isomer); the scheme of energy levels and transitions between links (d); phosphorescence spectra (e) of dimer (1), NAPHMA (2), APH (3) under excitations by $\lambda = 337\,\text{nm}$ (dioxane solutions, $C = 10^{-4}\,\text{M/l}$) the $I(U)$ characteristic of an oxyphenylnaphthalimide polymer-gilded film (f).

the corresponding energy levels and the spectral proofs of the processes occurring in the system are presented in Figure 4. We note that the existence of the lowering of energy levels from chromophore to chromophore along the molecular system gives the ground to predict not only the unidirect neutral electronic excitation transfer but the unidirect charge carrier current. Really, the “diode” $I(U)$ characteristic for a metal-organic system of gold islands connected by oxyphenylnaphthalimide molecules was observed (Fig. 4f).

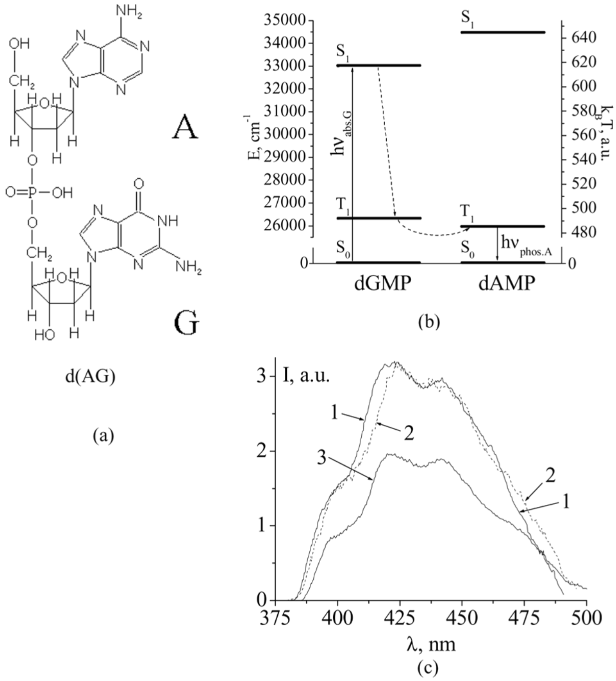


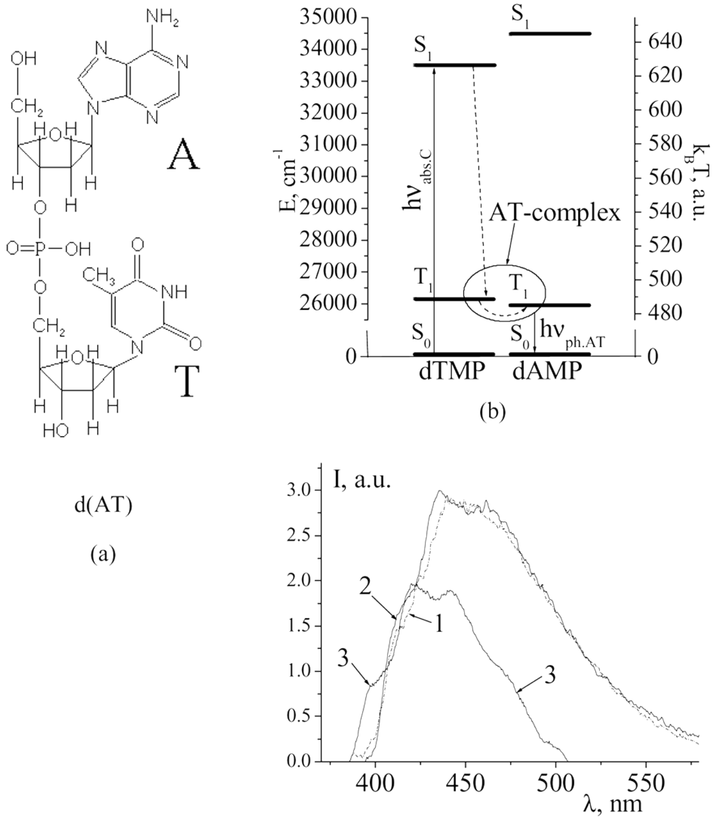
FIGURE 5 Structure formulae of d(AG) (a); the scheme of energy levels and transitions between links (b); phosphorescence spectrum of d(AG) under excitation by $\lambda = 294\,\text{nm}$ (1), the linear combination of dGMP and dAMP phosphorescence spectra (2), phosphorescence spectra of dAMP (3) at $T = 77\,\text{K}$ (c). Water solutions, $C = 10^{-4}\,\text{M}$ b.p.

System 2. Basic units – dGMP, dAMP, their mutual system – deoxyadenylyl-deoxyguanosine d(AG), the corresponding energy levels and the spectral proofs of the processes occurring in the system are presented in Figure 5.

System 3. Basic units – dTMP, dAMP, their mutual system – deoxyadenylyl-thymidine d(AT), the corresponding energy levels are presented in Figure 6. The phosphorescence spectra manifest that some complexes are created between A- and T-cells in this system.

System 4. Basic units – dCMP, dTMP, dAMP, their mutual system – deoxyadenylyl-thymidylyl-deoxycytidine d(ATC), the corresponding energy levels and the spectral proofs of the processes occurring in the system are presented in Figure 7.

System 5. Basic units – dCMP, dGMP, dTMP, dAMP, their mutual system – block-copolymer oligonucleotide d(CCCGGGTTTAAA)



(a)

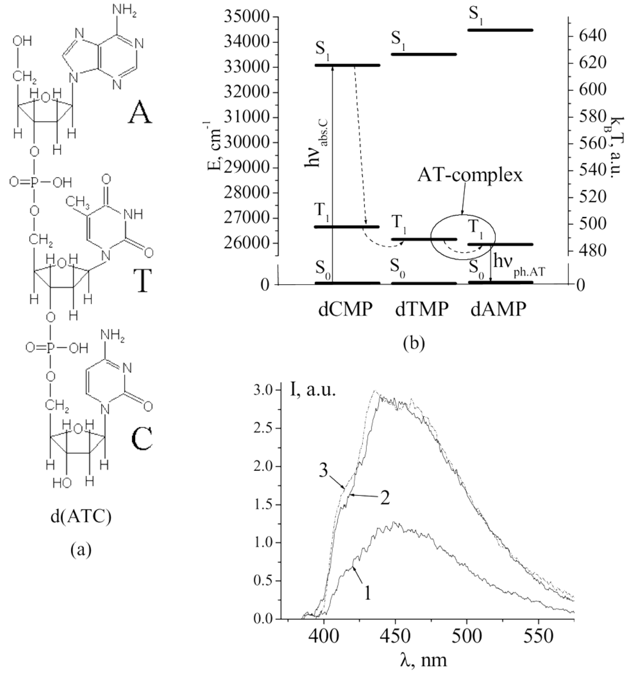
(b)

(c)

FIGURE 6 Structure formulae of d(AT) (a); the scheme of energy levels and transitions between links (b); phosphorescence of d(AT) under excitations by $\lambda = 260\,\text{nm}$ (1) and $\lambda = 300\,\text{nm}$ (2), dAMP (3) at $T = 77\,\text{K}$ (c). Water solutions, $C = 10^{-4}$ M b.p.

(nucleotides in this compound are linked by PO_4H-groups), double-stranded polymer poly$(dAdT)_2$ (it is taken to study the d(CCCGGGTTTAAA) phosphorescence nature), the corresponding energy levels and the spectral proofs of the processes occurring in the system are presented in Figure 8.

It was shown that absorbing centers in all the compounds investigated are its corresponding basic groups; the fluorescence of the compounds investigated is related to a mixture of the corresponding emissions of basic groups. The phosphorescence of d(AG) is mainly due to the emission of the deoxyadenosine π-electron system. This fact



d(ATC)

(a)

(b)

(c)

FIGURE 7 Structure formulae of d(ATC) (a); the scheme of energy levels and transitions between links (b); phosphorescence of d(ATC) under excitations by $\lambda = 260\,\text{nm}$ (1) and $\lambda = 300\,\text{nm}$ (2), d(AT) by $\lambda = 300\,\text{nm}$ (3) at $T = 77\,\text{K}$ (c). Water solutions, $C = 10^{-4}\,\text{M}$ b.p.

indicates that the triplet electronic excitation energy transfer from deoxyguanosine residue to deoxyadenosine residue takes place. In contrast, for d(AT), d(ATC), and d(CCCGGGTTTAAA), the phosphorescence is not the emission of any individual nucleotide system. The obtained data suggest that the observed emissions can belong to AT-complexes. The additional investigations of DNA and poly(dAdT)$_2$ show that this complex is formed by deoxyadenosine and thymidine residues. So, the experimental data demonstrate the direct energy transfer to the AT-complex [21,22]. Thus, the existence of the dominant unidirect triplet electronic excitation energy transfer in the specially designed and synthesized nucleotides-containing functional compounds under selective photon excitation gives the possibility to propose these compounds to be used in the construction of nanobiodevices.

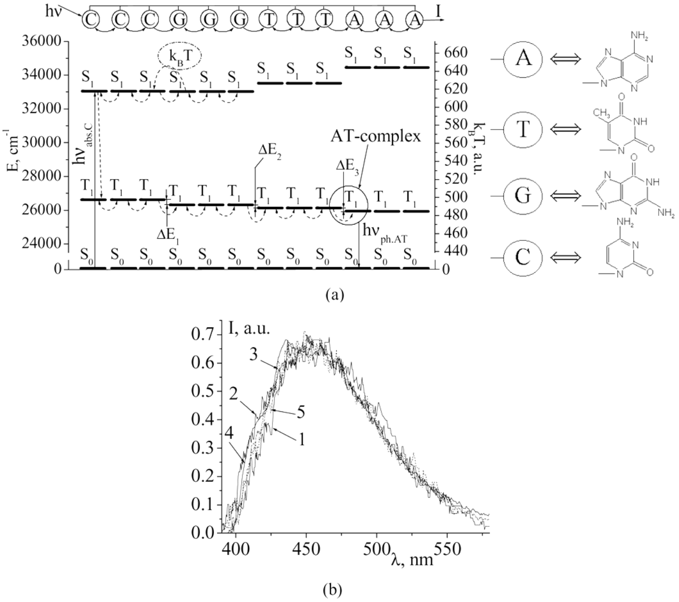


FIGURE 8 Scheme of energy levels and transitions between links in d(CCCGGGTTTAAA) (a); phosphorescence spectra (b) of poly(dAdT)$_2$ (1), d(CCCGGGTTTAA) (2), DNA (3), d(AT) (4), and d(ATC) (5) under excitations by photons at $\lambda = 300$ nm at T = 77 K. Water solutions, $C = 10^{-4}$ M b.p.

3.3. Simulations of the Excitonic Current Through a Functional Oligomer Macromolecule

The results of computer simulations of the electronic excitation transfer along an oligomer macromolecule (such as d(CCCGGGTTTAAA)) [21] are presented below. This model takes into account the reverse exciton currents that can be significant under increasing the temperature (Fig. 9).

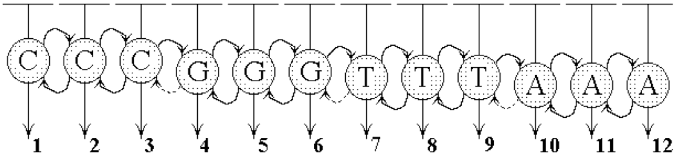


FIGURE 9 Model of electronic excitation transfer along a 12-link macromolecule.

The energy structure of this model system is such that one can excite the first (initial) link (by a photon with certain wavelength) without exciting the other links. This link can be deactivated by spontaneous radiative or irradiative transitions to the ground state with the general probability α, or pass its excitation to the neighbor right-hand link with the probability $(1 - \alpha)$. The excitation of the second link can be deactivated with the probability α too or be passed to the first and the third links with the probability $(1 - \alpha)/2$. The same processes with links 3, 5, 6, 8, 9, 11, and 12 take place. The 4, 7, and 10 links differ from others: they pass their excitations to the neighbor right-hand links with the probability $(1 - \alpha)/2$ and to the neighbor left-hand links with the probability $(1 - \alpha)/(1 + e^d)$, where $d = \Delta E/kT$. If the number of excitations generated in the first link is equal 1, then one has to calculate the number (C) of excitations that are localized in the last (12-th) link.

The results of these calculations are presented in Table 1. The recurrence formula and the value of C were calculated using MAPLE7 software.

For the cases $d = 17.353$ for $T = 4.2\,K$, $d = 7.843$ for $T = 77\,K$, and $d = 2.057$ for $T = 293\,K$ at $\alpha \leq 0.001$, the value is $C = I_0/I > 0.9$; $\alpha \leq 0.00001$ is the typical value for π-electron-containing molecular systems with $d \geq 1$.

The macromolecule investigated is the block-copolymer oligonucleotide d(CCCGGGTTTAAA). Photons with the wavelength $\lambda = 300\,nm$ excite the first singlet levels S_1 of π-electron systems of the first triad CCC. As a result of the singlet-triplet conversion, the triplet excitations in the first triad appears. Then the following events takes place: random walks of triplet excitations in the first triad → the jump to the

TABLE 1 The Values of C for Various Values of α and d

α d	0.00001	0.0001	0.001	0.01
0.0	0.99856	0.9857	0.8714	0.3522
0.5	0.99914	0.9914	0.9191	0.4819
1.0	0.99938	0.9938	0.9408	0.5697
1.25	0.99945	0.9983	0.9470	0.6000
1.5	0.99950	0.9950	0.9515	0.6235
2.0	0.99956	0.9956	0.9571	0.6554
2.5	0.99959	0.9959	0.9602	0.6744
3.0	0.99961	0.9961	0.9621	0.6857
3.5	0.99962	0.9962	0.9631	0.6925
4.0	0.99963	0.9963	0.9637	0.6966
20.0	0.99964	0.9964	0.9640	0.7029

second triad and the realization of the random walks here → jump to the third triad . . . , and so on. Though a part of triplet excitations disappears due to the spontaneous deactivation; another part overcomes the energy barrier (ΔE) between triads giving the contribution to the reverse current. All these steps were took into account in our computer simulations. The spectral investigations (see, e.g., [21,22,27]) lead to conclusion that AT-sequences are the triplet traps in our functional macromolecule, as well as DNA. So, the final point for triplet excitons in the 12-sequence oligomer compound is the boundary between TTT and AAA triads. The computer simulation and spectral results prove that the majority of triplet excitations generated in the first CCC-triad reaches this trap.

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