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Nonlinear dynamics of excitations in DNA

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Abstract

The dynamics of a hole transfer in short fragments of a DNA molecule is considered. The conditions under which irreversible transition of a hole takes place are found. The transition time of a DNA fragment consisting of 6 nucleotides is calculated to be $\sim 10^{-11}$ s. © 2000 Elsevier Science B.V. All rights reserved.

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Recently, we have witnessed increased interest in the transfer of excitations in DNA [1–9]. Experiments do not give any unambiguous evidence in favour of one or other mechanism of this transfer. In particular, the possibilities are considered for it to proceed either by a hopping mechanism or by a superexchange mechanism or by a combination of both [4,5,10–13]. An alternative to all the above is a soliton [14,15] or a polaron mechanism of a charge transfer in proteins and DNA [16–18].

In this Letter we model the dynamics of a hole transfer examining a simple system consisting of a nucleotide sequence of the form GTTGGG, where G is guanine, T is timine. Migration of holes was studied experimentally in the sequences of the form $G_1TTG_2TTG_3 \ldots G_NTTGGG$, where guanine was the donor of a hole and the guanine triplet GGG was the acceptor [6,8,9].

In the case of linear chain under consideration the initial Hamiltonian $\mathcal{H}$ has the form [19–21]

$$\begin{aligned}
&\mathcal{H} = H + T + U,\\
&H = \sum_i \alpha_i a_i^+ a_i + \sum_{i,j} v_{ij}\left(a_i^+ a_j + a_j^+ a_i\right),\\
&T = \frac{1}{2}\sum_i M_i \left(\frac{du_i}{dt}\right)^2, \quad U = \sum_i \frac{k_i}{2} u_i^2,\\
&\alpha_i = \alpha_i^0 + \alpha_i' u_i, \qquad (1)
\end{aligned}$$

where i runs the values from 1 to N, N is the number of sites in the chain; a_i^+, a_i are the operators of creation and annihilation of the excitation at the ith site; H is the operator of the excitation energy; v_{ij} are matrix elements of the transition; α_i^0 is the excitation energy at the ith site; T is the kinetic energy of the sites; M_i is the mass of the ith site; u_i is the displacement of the ith site from the equilibrium position; U is the potential energy of the sites; k_i are elastic constants. It is believed that excitation energy α_i depends linearly on the site displacement,

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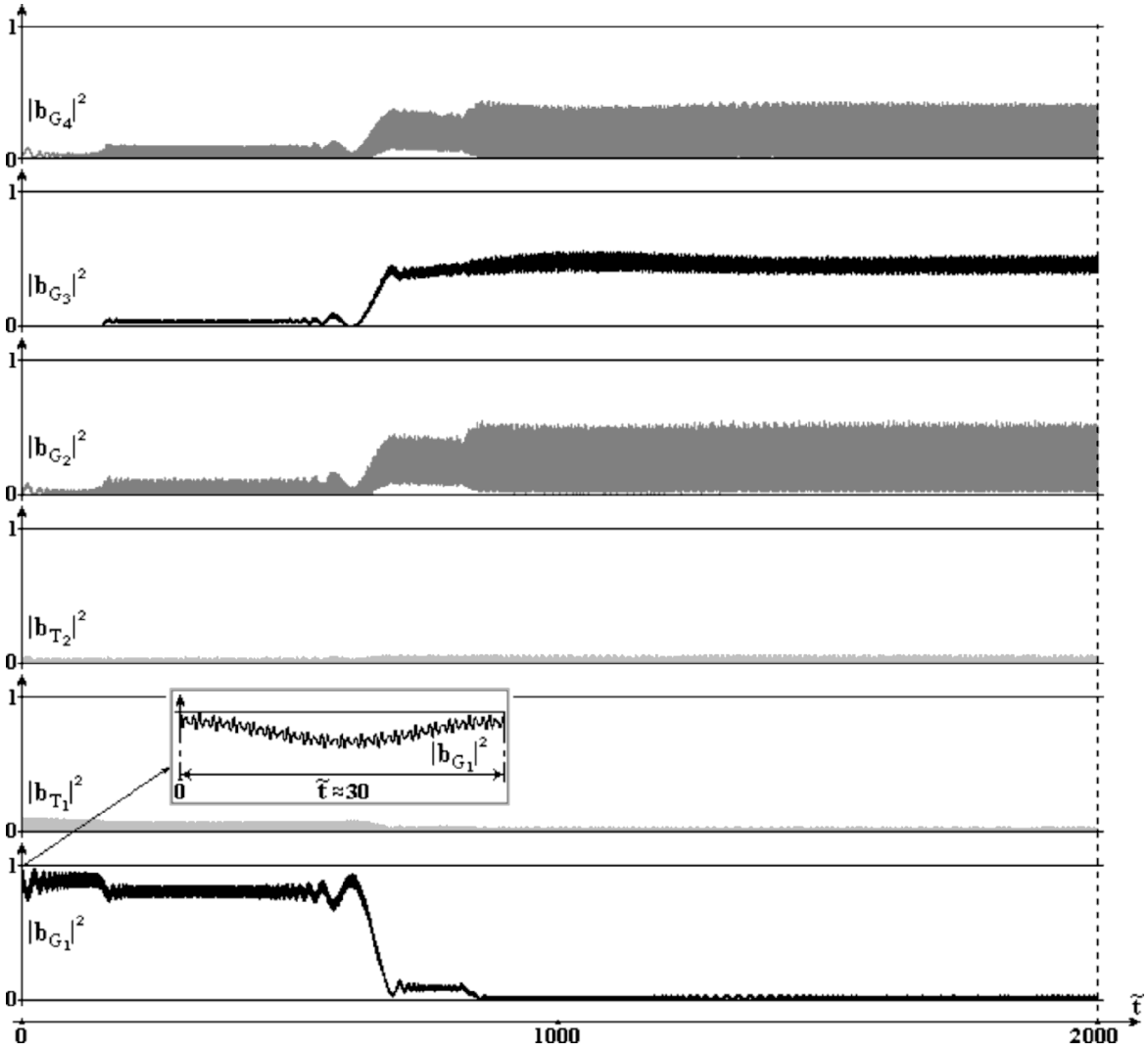

Fig. 1. Time-dependencies of the probabilities $|b_i|^2$ of the occurrence of a hole for 6-site model. At $\tilde{t}=0$ the hole localized at G_1 site. At $\tilde{t}\approx 900$ it passes to sites $G_2G_3G_4$. The final probabilities at G_2 and G_4 are approximately equal ($|b_{G_2}|^2\approx 0.27$, $|b_{G_4}|^2\approx 0.22$), and $|b_{G_3}|^2\approx 0.47$. The probabilities at T_1, T_2 are small during the transition. The insert shows the detailed behaviour of the probability at G_1 at the initial time moments.

α_i' has the meaning of the coupling constant between quantum and classical subsystems.

The equations of motion resulting from Hamiltonian (1) in the neighbourhood approximation have the form

$$i\frac{db_i}{d\tilde{t}} = \eta_i b_i + \eta_{i,i+1}b_{i+1} + \eta_{i,i-1}b_{i-1} + \kappa_i\omega_i^2\tilde{u}_i b_i, \tag{2}$$

$$\frac{d^2\tilde{u}_i}{d\tilde{t}^2} = -\omega_i'\frac{d\tilde{u}_i}{d\tilde{t}} - \omega_i^2\tilde{u}_i - |b_i|^2. \tag{3}$$

Here Eqs. (2) present Shrödinger equations for the probability amplitudes b_i describing the excitation evolution in the deformed chain, and (3) are the classical motion equations.

The quantities involved in (2), (3) relate to the parameters of Hamiltonian (1) as

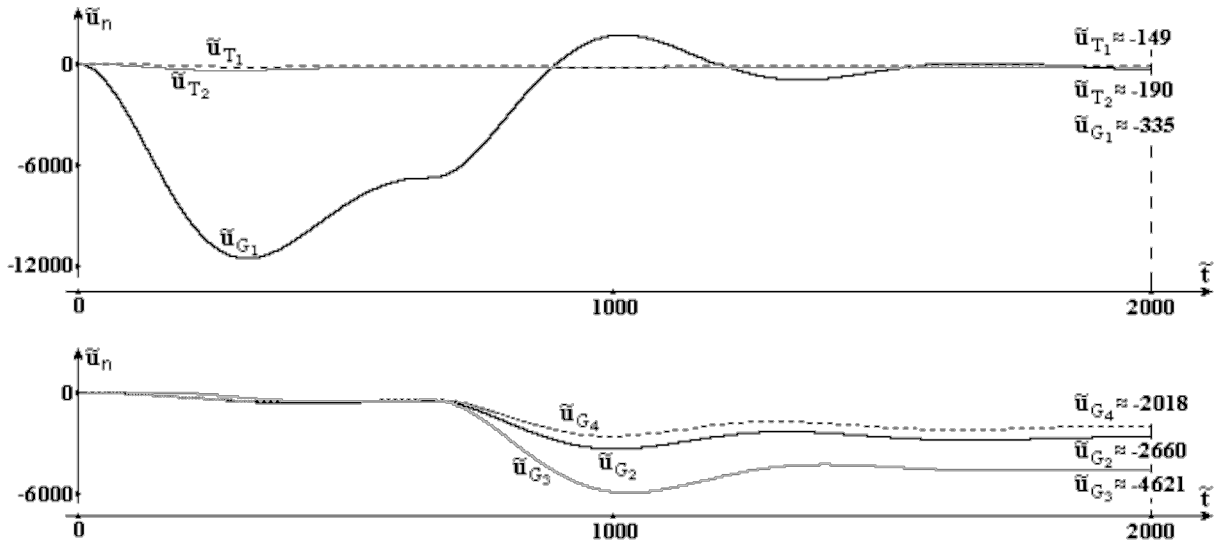

Fig. 2. Time-dependencies of displacements for the 6-site model. Top: displacements of G_1 (black line), T_1 (gray line) and T_2 (dotted line); bottom: G_2 (black line), G_3 (gray line), G_4 (dotted line). Displacements at T_1 and T_2 are small during the transition. Sites gradually tend to new equilibrium states.

$$\eta_i = \tau\alpha_i^0/\hbar, \qquad \eta_{ij} = \tau\nu_{ij}/\hbar,$$
$$\omega_i^2 = \tau^2 k_i/M_i, \qquad \kappa_i\omega_i^2 = \tau^3(\alpha_i')^2/M_i\hbar,$$
$$u_i = \beta_i\tilde{u}_i, \qquad \beta_i = \tau^2\alpha_i'/M_i, \qquad t = \tau\tilde{t},$$

where $\hbar$ is the Plank constant; τ is the arbitrary time scale relating the time t and the dimensionless variable $\tilde{t}$ in terms of which the differentiation in (2), (3) is performed.

According to (2), (3) each site is determined by four real Coushi equations. The numerical integration of these equations was carried out by the Runge–Kutta method with initial values of velocities and site displacements equal to zero. At the initial moment the hole was assumed to be localized at G_1: $|b_{G_1}(0)|^2 = 1$, and the normalization condition $\sum_i |b_i|^2 = 1$ was used to make sure that the calculations are accurate.

Figs. 1 and 2 show the solutions of system (2), (3) for a nucleotide sequence $G_1T_1T_2G_2G_3G_4$ for $\tau = 10^{-14}$ s and the values of the parameters $\alpha_{G_i}^0 = 0$, $\alpha_{T_i}^0 = 0.7$ eV, $\nu_{i,i+1} = 0.12$ eV, $\nu_{ij} = \nu_{ji}$, which is in agreement with experimental results [6,8,9] and theoretical estimates [13]. These parameter values correspond to $\eta_{G_i} = 0$, $\eta_{T_i} = 11.2$, $\eta_{i,i+1} = 1.92$. The other parameters were taken to be $\kappa_i = 4$, $\omega_i = 0.01$, $\omega_i' = 0.006$, which correspond to picosecond characteristic scales of the DNA motion [22].

According to Fig. 1 the hole which at the moment $t = 0$ was at G_1 passes to GGG during the time $\Delta t \approx 9 \times 10^{-12}$ s.

The only parameters which cannot be estimated are the coupling constants κ_i. The numerical study of system (2), (3) shows that the transition takes place only if κ_i does not exceed a certain critical value. If so, in some range of κ_i values, the hole irreversibly transfers from G_1 to GGG, as is seen from the figure. At $\kappa \ll 1$ the excitation may occur with relevant probability either at G_1 or at GGG.

Fig. 2 which shows the time dependencies of the site displacements $\tilde{u}_i$ gives an illustration of how an irreversible transfer proceeds. At the initial moment the displacement at the first site G_1 grows thus decreasing the energy of the hole at G_1. After that, however, the displacement at the sites G_2, G_3 and G_4 (exhibiting complicated evolution) increases in modulus making energetically advantageous localization of the hole at the sites GGG. The displacement values $u_i(\infty)$ upon completion of the transfer are $u_{G_1} = -0.0462$ Å, $u_{T_1} = -0.0296$ Å, $u_{T_2} = -0.0384$ Å, $u_{G_2} = -0.542$ Å, $u_{G_3} = -0.926$ Å,

$u_{G_4} = -0.417$ Å. They correspond to polaron potential wells $\Delta E_i = \hbar\kappa_i\omega_i^2\tilde{u}_i/\tau$: $\Delta E_{G_1} = -0.0058$ eV, $\Delta E_{T_1} = -0.0037$ eV, $\Delta E_{T_2} = -0.0048$ eV, $\Delta E_{G_2} = -0.068$ eV, $\Delta E_{G_3} = -0.12$ eV, $\Delta E_{G_4} = -0.052$ eV. The large value of displacement $\tilde{u}_{G_1}$ at time $\tilde{t} \approx 300$ ($\max|u_{G_1}| \approx 2.3$ Å) demonstrates the so-called "open state" of DNA which can play an important role in the processes of DNA damage, denaturation and transcription [22–24]. The joint evolution of the excitation and deformation illustrates the polaron mechanism of the excitation transfer in DNA fragments.

The results obtained provide support for the idea [25] of a possibility of fast ($\leqslant$ ns) long-range transitions of excitations in polynucleotide chains.

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