Dissipative dynamics of a hole transfer in single- and double-strand electronic model of DNA

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I. Introduction

Formulation of the problem and theoretical approach

The transfer of a hole carrier in a fragment of artificial DNA (oligonucleotide) is investigated by considering the "hole carrier in DNA + dynamic environment" as an open quantum system. DNA structure is approximated either by (Mod1) a simplified single-strand model or (Mod2) the more realistic double-strand one. Time evolution of the system density operator $\rho(t)$ is described by the Lindblad equation that is solved numerically.

Two physical sources, and the corresponding operators $D_1[\rho]$ and $D_2[\rho]$ in the Lindblad equation, give rise to the carrier dissipative dynamics: (1) capture of the carrier by the environment, resulting in the escape of the hole from the DNA, (2) the loss of the carrier wavefunction coherence under the influence of stochastic fluctuations of the environment.

Our goal is to check whether both structurally distinct DNA models, discussed earlier in [1] and denoted here as Mod1 and Mod2, enable to reproduce accurately two different regimes of a hole motion of a hole carrier, namely, coherent tunneling in short DNA oligonucleotides and incoherent hopping in longer ones, in accordance with results of experimental measurements of the rate of charge transfer along a DNA oligonucleotide as a function of its length N.

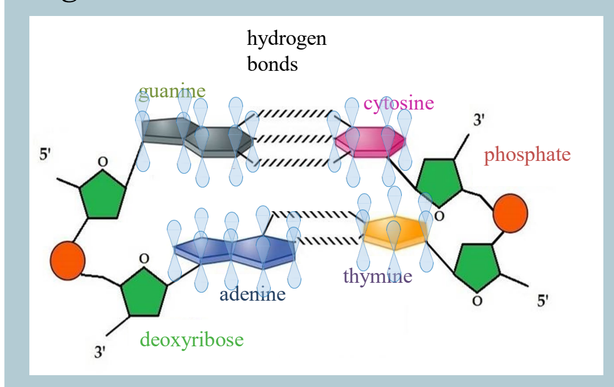

Fig. 1. Fragment of DNA with two pairs of nitrogenous bases: adenine - thymine (A-T) and guanine - cytosine (G-C). The overlapping π-orbitals of the carbon and nitrogen atoms of the nitrogenous bases form the "paths" for charge movement along DNA.

Giese et al. [2] laid the foundations for measuring a charge transfer in oligonucleotides - homogeneous and regular DNA sequences. Here, a quantum models, Mod1 and Mod2, for describing results obtained in these experiments are explore.

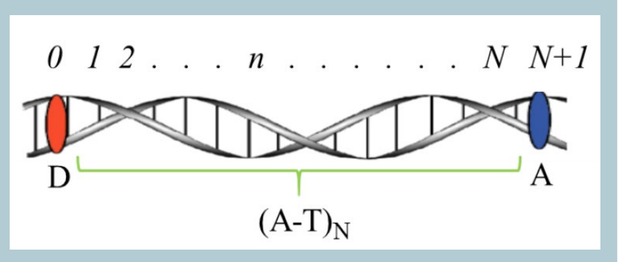

Fig. 2. Schematic view on DNA oligonucleotide in the Giese experiment [2, 3]. Donor and acceptor centers, each formed by (G-C) base pairs, are highlighted and joined by the $(A\text{-}T)_N$ bridge along which a hole carrier transfer.

II. Simplified single-strand electronic model (Mod1) of DNA oligonucleotide.

Each base pair is considered as a structureless site numbered as $n = 0, 1.., N, N+1$, in a single-stranded electron model with the Hamiltonian :

$$H = \sum_{n=0}^{N+1} \varepsilon_n a_n^+ a_n + \sum_{n=0}^{N} V_{n,n+1}\left(a_n^+ a_{n+1} + a_{n+1}^+ a_n\right)$$

where $a_n^+ \left(a_n\right)$ are the operators of creation (annihilation) of a spinless fermion, ε_n is the energy level of the hole carrier at site n, and $V_{n,n+1}$ are the amplitudes of a hole transitions between neighboring sites.

The energy level diagram with the model parameters used is shown in Figure 3.

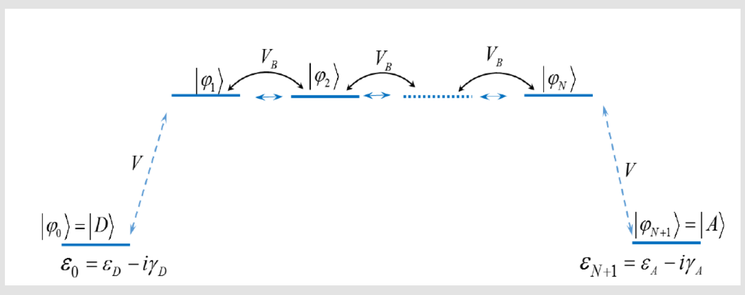

Fig. 3. Energy level diagram. $|D\rangle$ and $|A\rangle$ – states of a hole carrier on the donor (n =0) and acceptor (n =N+1), $|\varphi_n\rangle (n = 1 \div N)$ – state of the charge carrier on the n-th (A-T)-pair of the bridge.

III. Method of calculating the charge transfer rate.

a) From Fermi to Pauli operators:

$$a_n \to \left(\prod_{j=0}^{n-1} \sigma_j^z\right)\sigma_n^+, \qquad a_n^+ \to \left(\prod_{j=0}^{n-1} \sigma_j^z\right)\sigma_n^-$$

The Jordan-Wigner transformation defines the Hamiltonian in the qubit representation:

$$H = \frac{\Delta}{2}\left(\sigma_0^z + \sigma_{N+1}^z\right) + \sum_{n=0}^{N} V_{n,n+1}\left(\sigma_n^+\sigma_{n+1}^- + \sigma_{n+1}^+\sigma_n^-\right)$$

b) The charge transfer evolution is described by the Lindblad equation:

$$\frac{\partial\rho}{\partial t} = -\frac{i}{\hbar}[H,\rho] + D_1[\rho] + D_2[\rho],$$

$$D_1[\rho] = \sum_{n=0,1,N+1} \gamma_n\left(\sigma_n^+\rho\sigma_n^- - \frac{1}{2}\left\{\sigma_n^+\sigma_n^-,\rho\right\}\right), \qquad D_2[\rho] = g\sum_{n=1}^{N}\left(\sigma_n^z\rho\sigma_n^z - \rho\right)$$

c) Selection of Mod1 parameters:

$$\Delta = \varepsilon_{D/A} = -150\text{meV}; \qquad V_{01} = V_{N,N+1} = V = 90\text{meV};$$

$$V_{n,n+1} = V_B = 30\text{meV}; \qquad n = 1,\ldots N-1$$

$$\gamma_0 = 0.1\text{meV}, \qquad \gamma_1 = 10\text{meV}, \qquad \gamma_{N+1} = 40\text{meV}, \qquad g = 0.3\text{meV}$$

d) Numerical solutions of the Lindblad equation are obtained using the Lindblad MPO Solver program [3].

IV. Results

Numerical solutions of the Lindblad equation in the qubit representation are found using the open source LindbladMPO code, which simulates elements of a quantum algorithm on a classical computer [3]. The calculated results are compared to available experimental data [2].

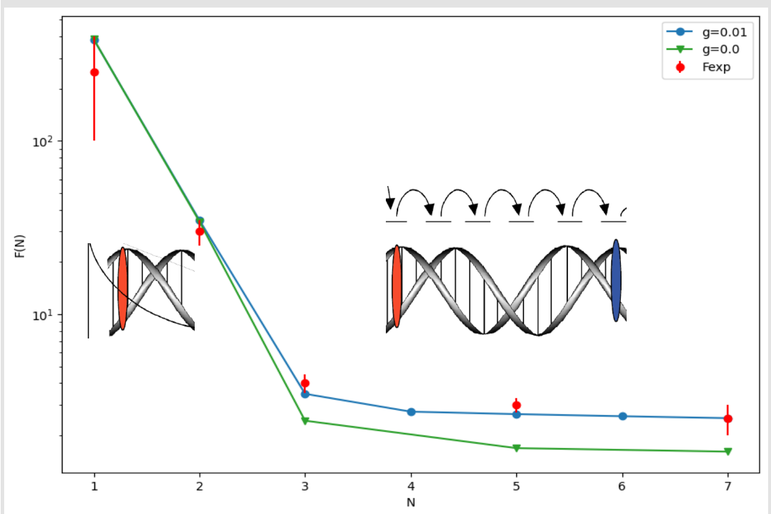

Fig. 4. Obtained from Mod1 the dimensionless hole transfer velocity $F(N)$ as a function of the length of the bridge $(A\text{-}T)_N$ is presented without dephasing, i.e. g=0 in $D_2[\rho]$ (marked in green) and including dephasing, g=0.3 meV (marked in blue).

In the framework of a more realistic double-stranded electronic model Mod2 (not specified here for brevity) the hole transfer velocity $F(N)$ have been also obtained withing the same approach based on the Lindblad equation and compared to experimental data [2] (see Fig.5).

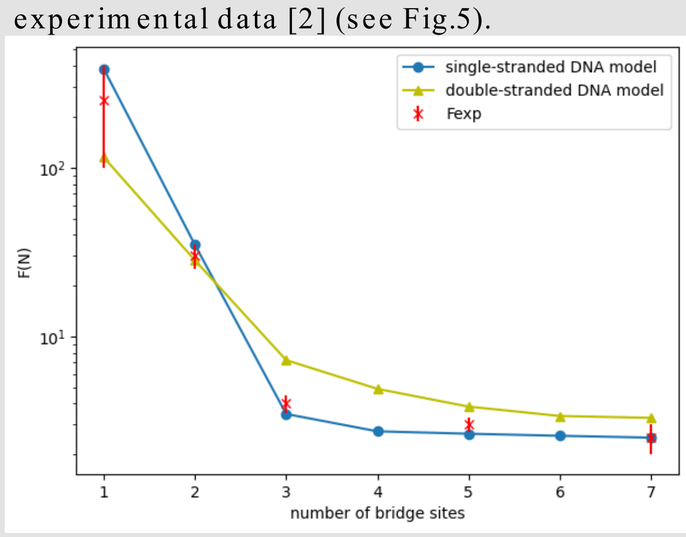

Fig. 5. Obtained from Mod 2 (marked in yellow) the dimensionless hole transfer velocity $F(N)$ as a function of the length of the bridge $(A\text{-}T)_N$ is presented without dephasing, i.e. g=0 in $D_2[\rho]$. The curve marked in blue is the same as in Fig. 4

V. Conclusion

We conclude that a drastic change in the hole transfer mechanism from tunneling (fast decrease of F at $N \le 3$), to hopping (nearly constant F at N >3) is accurately reproduced by Mod 1, (see Fig. 4). This behavior is maintained with the capture of the carrier by the environment as described by $D_1[\rho]$. Effects of the carrier wavefunction dephasing caused by thermal noise and described by $D_2[\rho]$, are small for tunneling ($N \le 3$), but introduce significant increase of F by factor 2 in the hopping transfer regime at N > 3.

As seen from Fig.5, the double-strand Mod 2 reproduces only a general trend of the N-dependence of the measured hole transfer velocity $F(N)$. The matter is that when solving the Lindblad equation for Mod 2 only one particular set of the model parameter values was used by us. We are planning to test other parameter sets available from the literature and check their better relevance for reproducing data of Giese experiment .

References

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2. Giese B., Amaudrut J., Köhler A.K., Spormann M., Wessely S. Direct observation of hole transfer through DNA by hopping between adenine bases and by tunneling. Nature. 2001. V. 412. P. 318-320.

3. Landa H. and Misguich G. Nonlocal correlations in noisy multiqubit systems simulated using matrix product operators. SciPost Phys. Core. 2023. V. 6. P. 037-38.