Models for nonlinear charge transport in biomolecules

Radial and twist polarons in DNA with base-pair inhomogeneities

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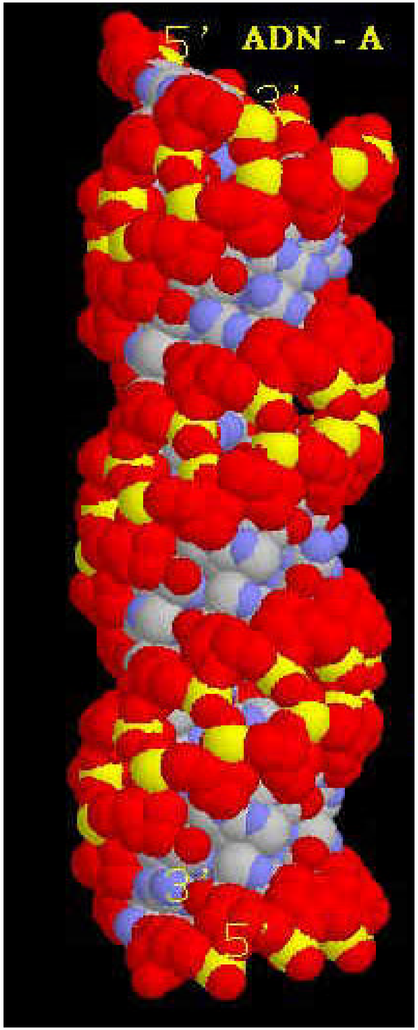
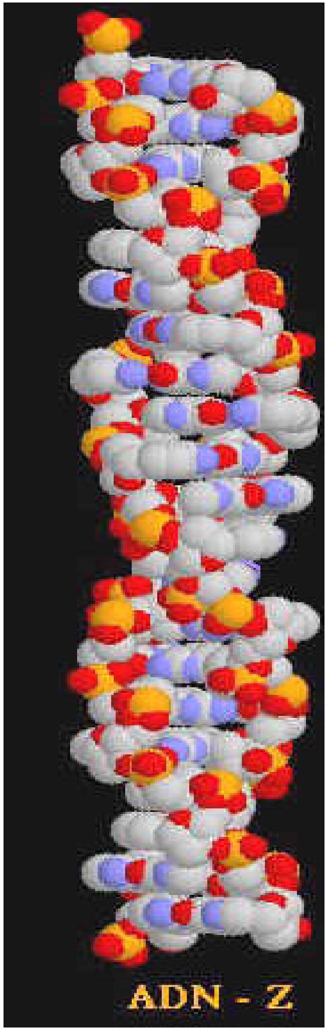
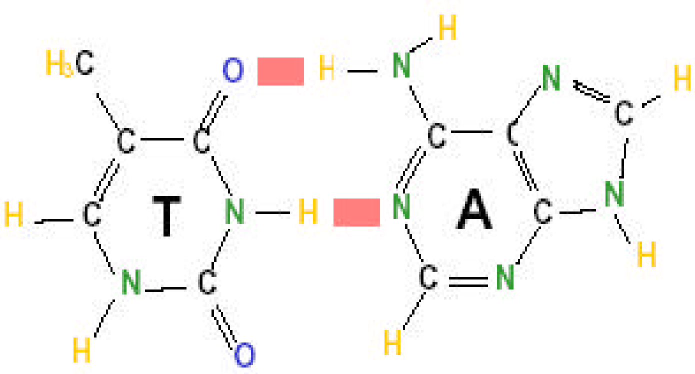
DNA types



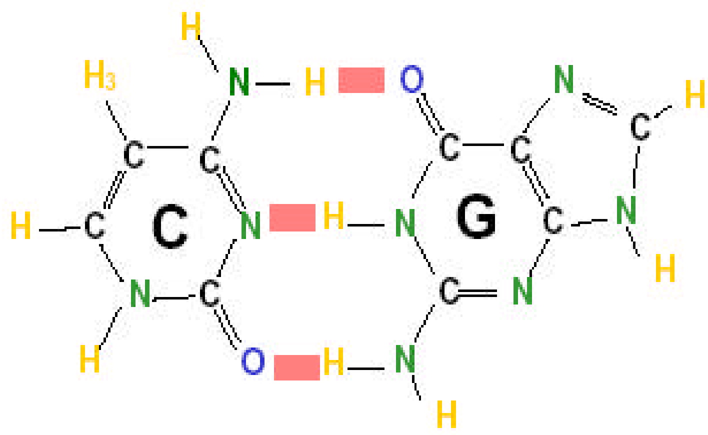


Base pairs

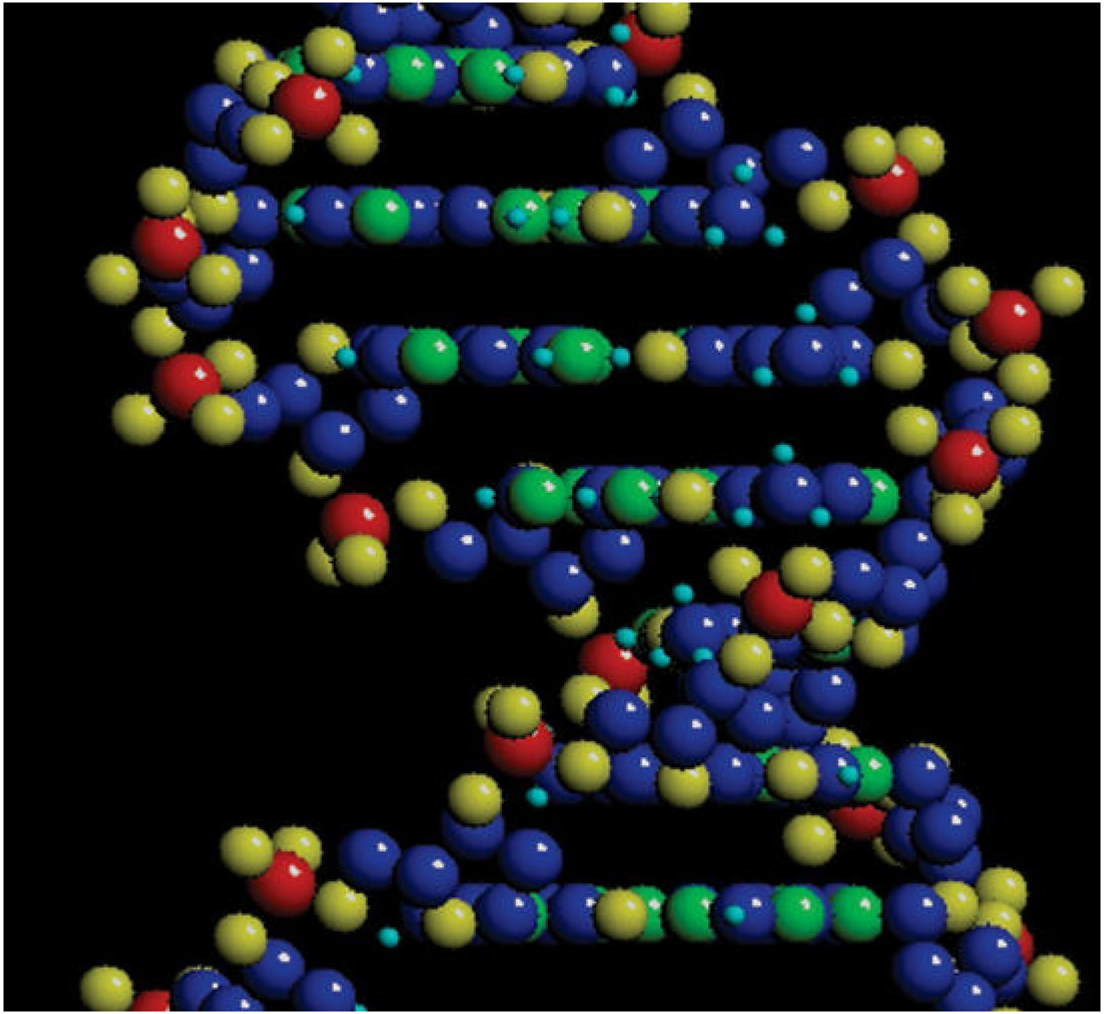
A pairs with T, with two H–bonds



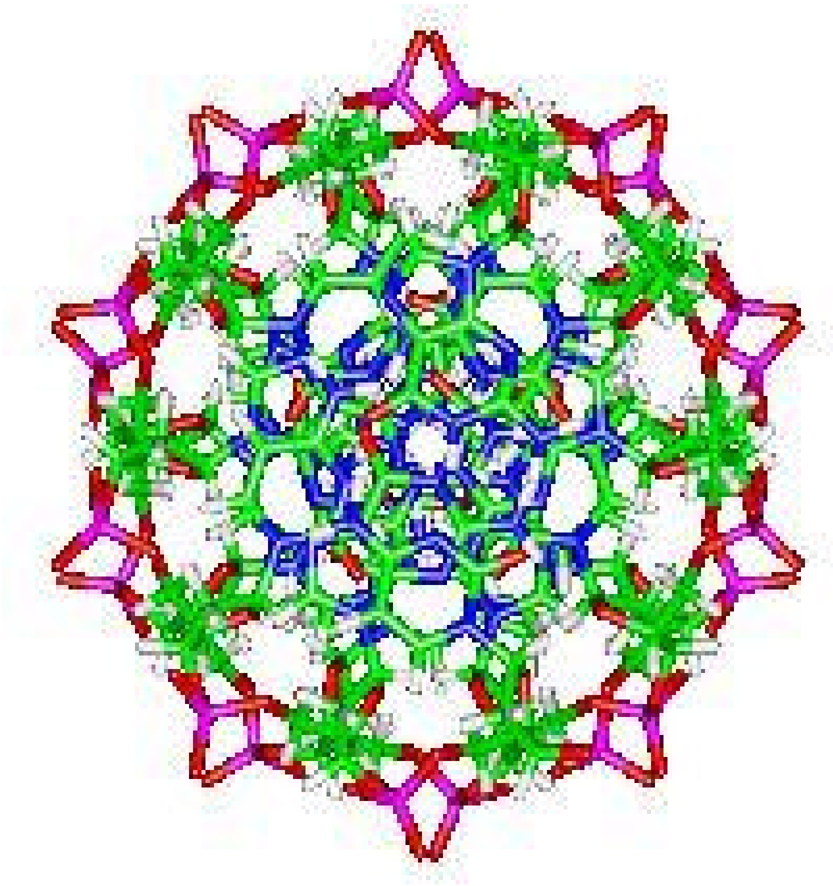
G pairs with C, with three H–bonds



B-DNA structure (1)



DNA structure (2)



Charge transport through DNA

- Role for biological functions:
  - Biosynthesis
  - DNA repair after radiation damage
- Technological interest
  - Molecular electronics
  - DNA wires
- Controversial experimental results
  - Well conducting one-dimensional wires: Giese et al., Nature 397 (1999)
  - Insulator: Braun et al., Nature 391 (1998)
  - Semiconductor: Porath et al., Nature 403 (2000)
- Charge transport can hop a few nanometres. Larger distances are unclear.

Theoretical Models

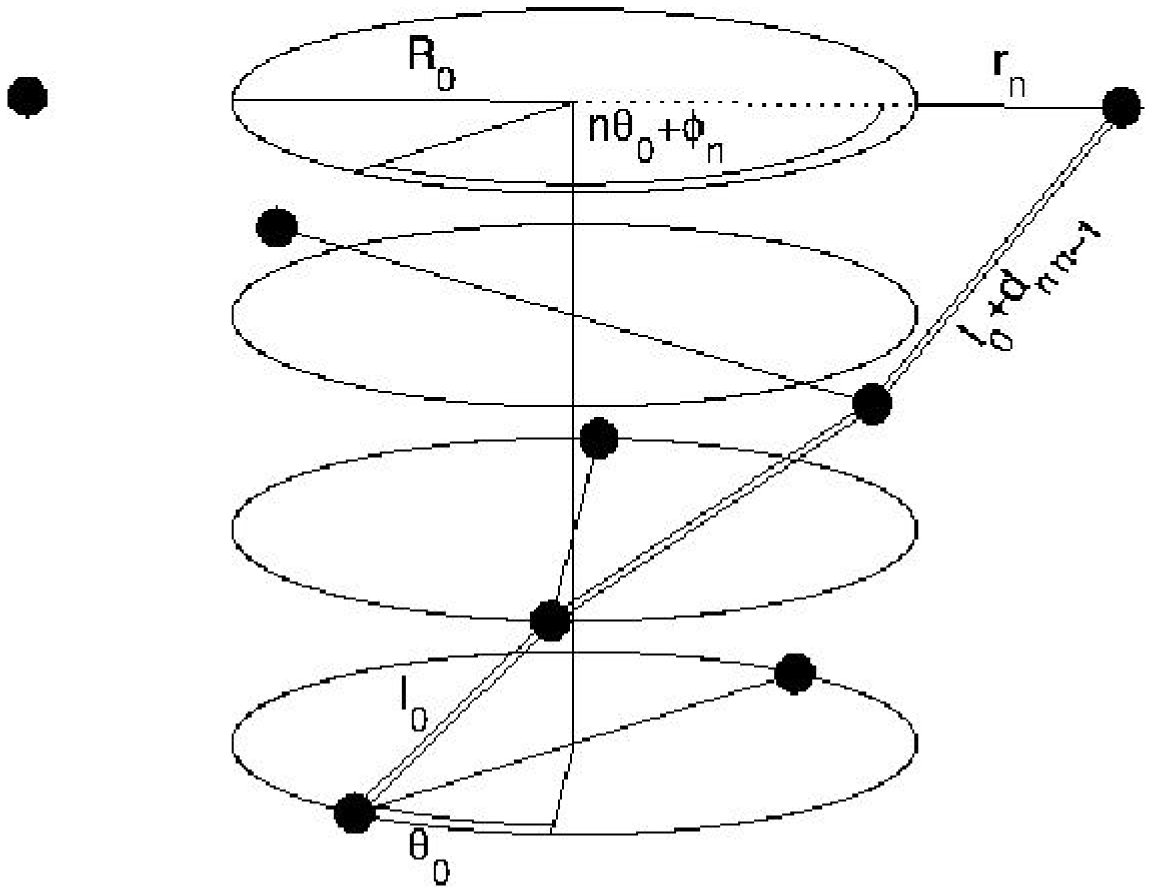
- Electron/hole transfer along π orbitals between base pairs.
- Conduction mechanisms:
  - Coherent tunneling
  - Incoherent phonon-assisted hopping
  - Classical diffusion under thermal fluctactions
  - Variable range hopping
  - Charge carriers assisted by solitons
  - Charge carriers assisted by polarons
- Our model: polarons in a 3D, semi-classical, tight-binding system

Polarons

- A charge in a lattice or biomolecule brings about a local deformation
- The local deformations and the charge can move together across the lattice

- Different cases:
  - Homogeneous molecule
  - Disordered molecule
  - A molecule with a local inhomogeneity: a different base pair.

3D DNA model



Hamiltonian

Hamiltonian of the system $\widehat{H} = \widehat{H}_{el} + \widehat{H}_{rad} + \widehat{H}_{twist}$

Particle charge transport Hamiltonian

$$\widehat{H}_{el} = \sum_n E_n |n\rangle\langle n| - V_{n-1,n}|n-1\rangle\langle n| - V_{n+1,n}|n+1\rangle\langle n|$$

$$E_n = E_n^0 + kr_n \qquad V_{n,n-1} = V_0(1-\alpha d_{n,n-1})$$

$$d_{n,n-1} = [a^2 + (R_0+r_n)^2 + (R_0+r_{n-1})^2 - 2(R_0+r_n)(R_0+r_{n-1})\cos(\theta_0+\theta_{n,n-1})]^{1/2} - l_0$$

$$l_0 = (a^2 + 4R_0^2\sin^2(\theta_0/2))^{1/2}$$

Lattice oscillations Hamiltonian (classical)

$$H_{rad} = \sum_n \left[\frac{1}{2M}(p_n^r)^2 + \frac{M\Omega_{r_n}^2}{2} r_n^2\right],$$

$$H_{twist} = \sum_n \left[\frac{1}{2J}(p_n^\phi)^2 + \frac{J\Omega_\phi^2}{2}(\phi_n - \phi_{n-1})^2\right]$$

Parameter values

Realistic parameter for DNA molecules. M. Barbi *et al.*, Phys. Lett. A **253**, 358 (1999); L. Stryer, *Biochemistry*, Freeman, New York, 1995.

- $a=3.4$ Å
- $m=300$ amu
- $R_0=10$ Å
- $\Omega_\phi=9\ 10^{11}\ s^{-1}$
- $V_0=0.1$ eV
- $M=2m$
- $J=MR_0^2$
- $\theta_0=36^o$
- No reliable data for parameters α and k. Adjustable parameters

$\Omega_r=8\ 10^{12}\ s^{-1}$.Radial frequency in a homogeneous model.

As Ω_r^2 is proportional to the strength of the hydrogen bond, we consider $\Omega_m^2 = b_n\ \Omega_r^2$

- Homogeneous case: $b_n=1.0$
- Inhomogeneous case:$b_n=0.8$ (A-T), $b_n=1.2$ (C-G). We introduce the inhomogeneity by distinguishing double and triple hydrogen bonds M. Salerno. Phys. Rev. A **44**, 5292 (1991). We study an homogeneous chain with a local inhomogeneity (all base pairs identical except one)

In the disordered case, we introduce parametric disorder in the on-site electronic energy by means of a random potential $E_n^0 \in [-\Delta E,\Delta E]$.

Dynamical equations (I)

General charge state: $|\Psi\rangle = \sum_n c_n(t)|n\rangle$

New time scale: $t \rightarrow \Omega_r t$ 1 t.u. ~ 0.1 ps

Separation between time scales: $\tau = \hbar\,\Omega_r / V_0$

Dimensionless quantities:

$$\tilde{r}_n = \sqrt{\frac{M\Omega_r^2}{V_0}}\, r_n\,, \qquad \tilde{k}_n = \frac{k_n}{\sqrt{M\Omega_r^2 V_0}}\,, \qquad \tilde{E}_n^0 = \frac{E_n^0}{V_0}$$

$$\tilde{\Omega} = \frac{\Omega_\theta}{\Omega_r}\,, \quad \tilde{V} = \frac{V_0}{J\,\Omega_r^2}\,, \quad \tilde{\alpha} = \sqrt{\frac{V_0}{M\,\Omega_r^2}}\,\alpha\,, \quad \tilde{R}_0 = \sqrt{\frac{M\,\Omega_r^2}{V_0}}\,R_0$$

Numerical values (omitting tildes):

$$\tau = 0.053,\ \Omega^2 = 0.013,\ V = 2.5\times 10^{-4}$$

$$R_0 = 63.1 \quad l_0 = 44.5$$

k=1, a adjustable,

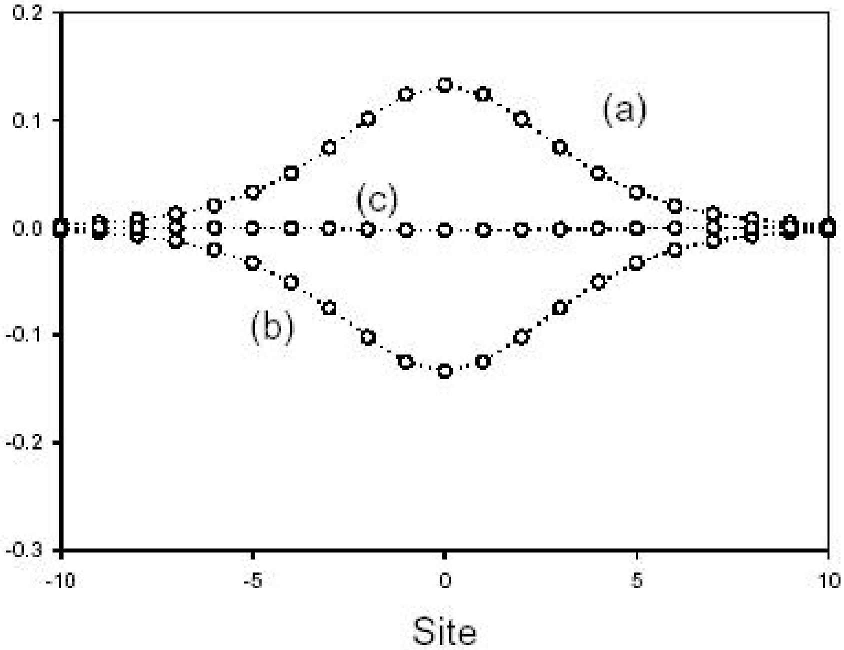
Dynamical equations (II)

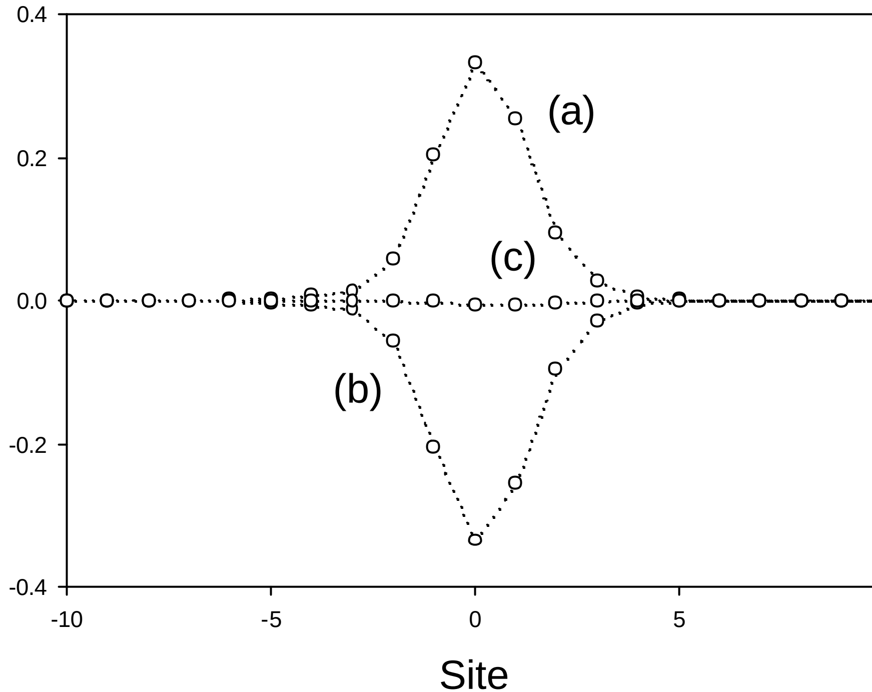
$$
\begin{aligned}
\mathrm{i}\,\tau\dot{c}_n &= (E_n + k\,r_n)\,c_n \\
&\quad - (1-\alpha\,d_{n+1,n})\,c_{n+1} - (1-\alpha\,d_{n\,n-1})\,c_{n-1}, \\
\ddot{r}_n &= -b_n r_n - k\,|c_n|^2 \\
&\quad - \alpha\,\left[\frac{\partial d_n}{\partial r_n}(c_n^* c_{n-1} + c_n c_{n-1}^*) + \frac{\partial d_{n+1}}{\partial r_n}(c_{n+1}^* c_n + c_{n+1} c_n^*)\right], \\
\ddot{\phi}_n &= -\Omega^2\,(2\phi_n - \phi_{n-1} - \phi_{n+1}) \\
&\quad - \alpha\,V\,\left[\frac{\partial d_n}{\partial \phi_n}(c_n^* c_{n-1} + c_n c_{n-1}^*) + \frac{\partial d_{n+1}}{\partial \phi_n}(c_{n+1}^* c_n + c_{n+1} c_n^*)\right]
\end{aligned}
$$

The case a=0 and $E_n^0=E_0$ corresponds to a Holstein system. T.D. Holstein, Ann. Phys. NY **8**, 325 (1959).

The case a=0 and k=0 and random E_n^0 corresponds to the Anderson model. P.W. Anderson, Phys. Rev. **109**, 1492 (1958).

Stationary polarons





Ordered chain.

Disordered chain

- (a) Probability amplitudes
- (b) Radial displacements
- (c) Twist deformations

Archilla, Palmero, Hennig, Romero, 2003

Participation number

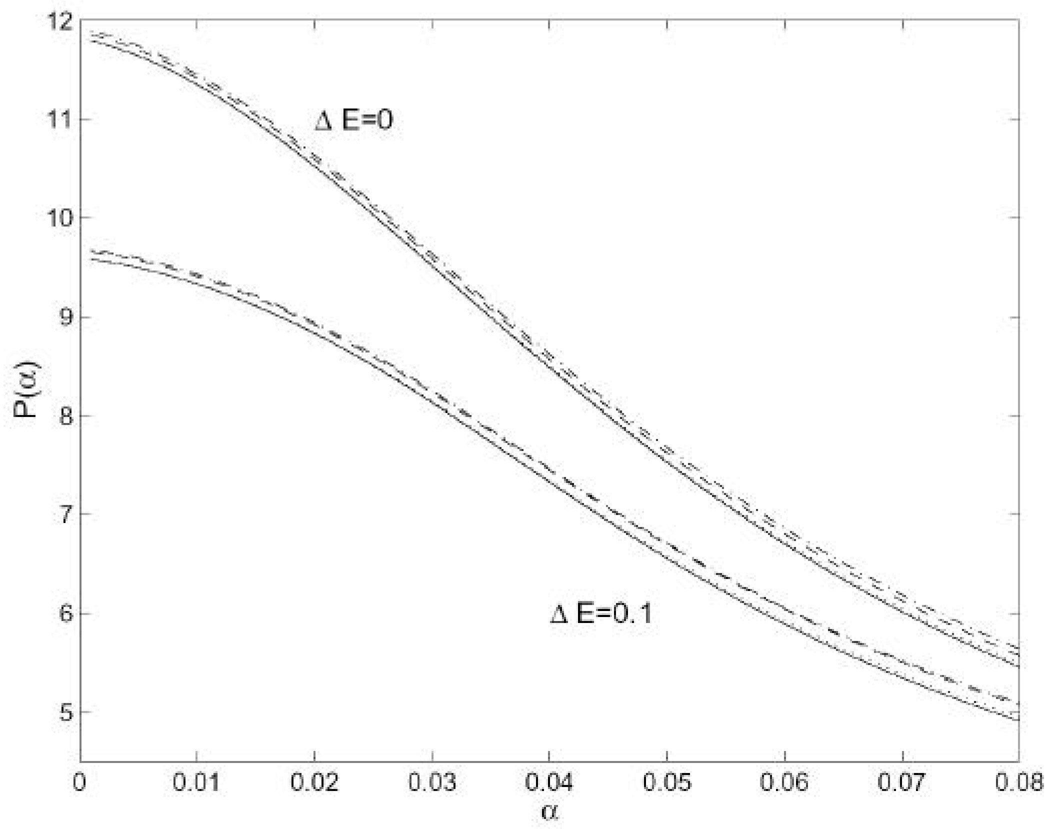
Degree of localization

$$L = \frac{\sum_n |u_n|^2}{(\sum_n |u_n|)^2}$$

Participation number

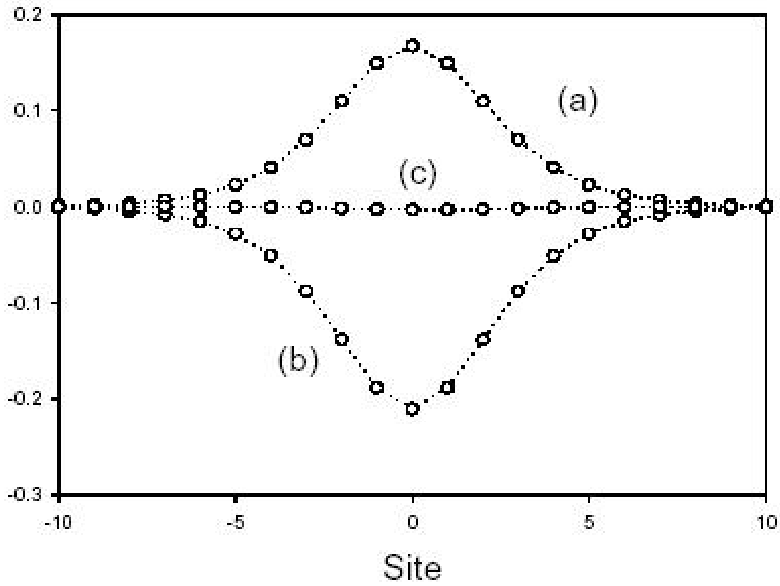
$$P = 1/L$$

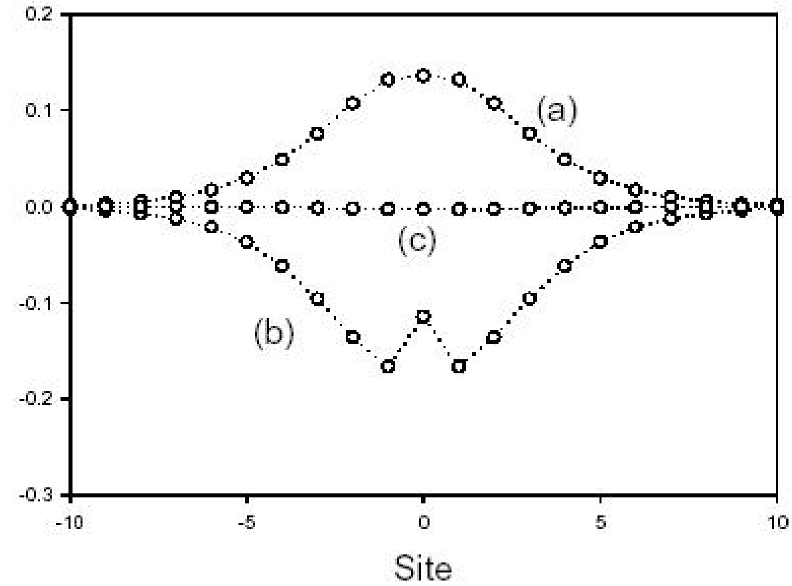
The value of the parameter a determines the degree of charge localization



Participation number for electronic probabilities, radial displacements, angular twists and density of energy for the homogeneous and the inhomogeneous case

Stationary states: A-T chain with a C-G impurity



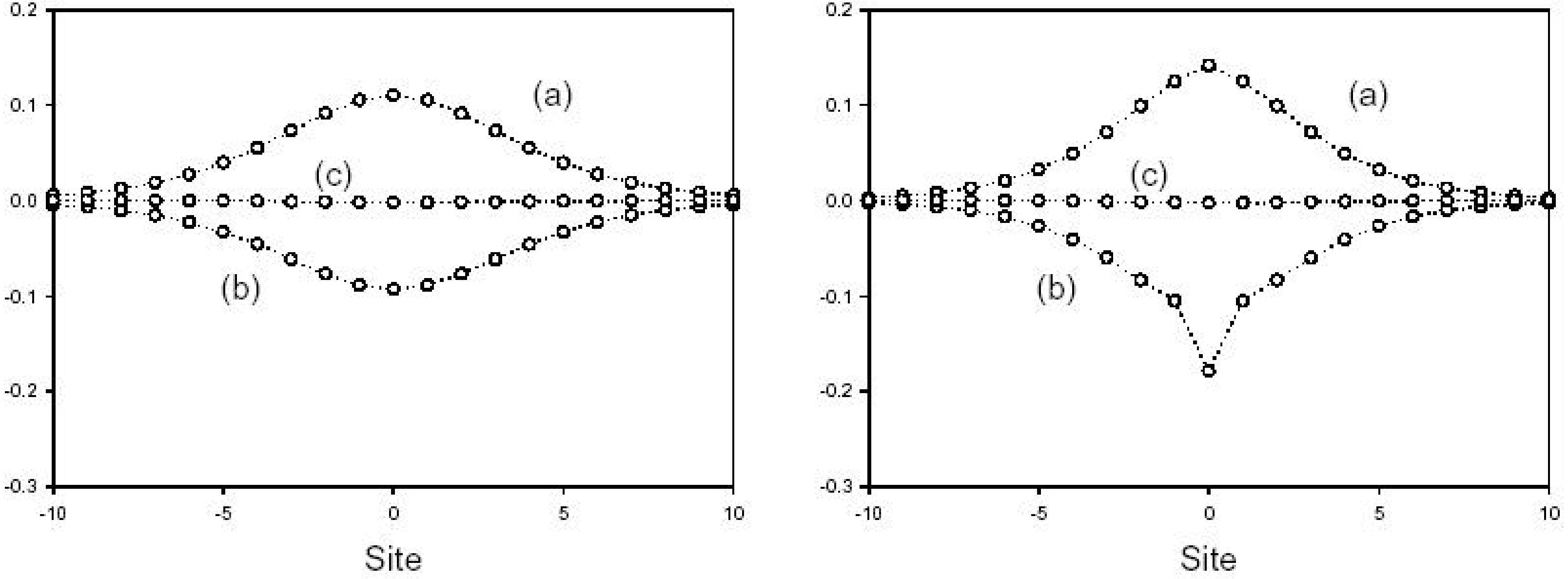


Homogeneous chain

A-T chain with a C-G

- (a) Probability amplitudes
- (b) Radial displacements
- (c) Twist deformations

Stationary states: C-G chain with an A-T impurity



Homogeneous chain C-G chain with an A-T

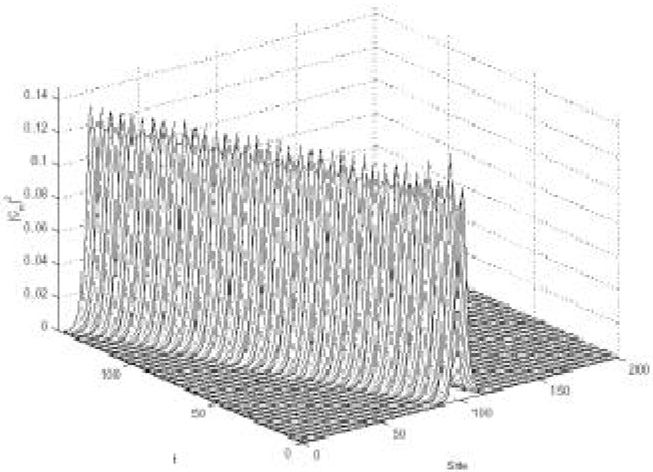
- (a) Probability amplitudes
- (b) Radial displacements
- (c) Twist deformations

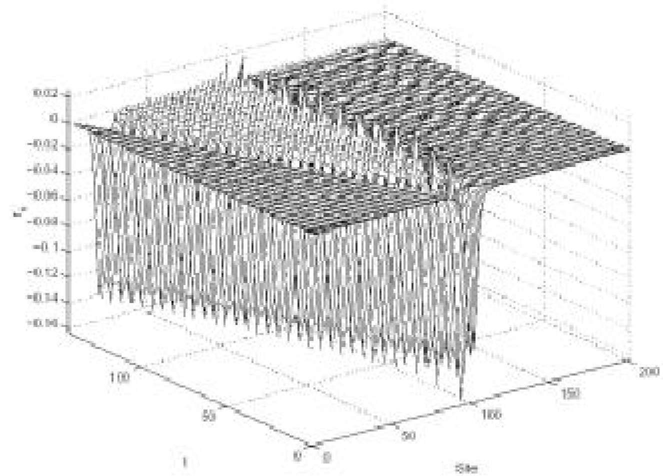
Archilla, Palmero, Hennig, Romero, 2003

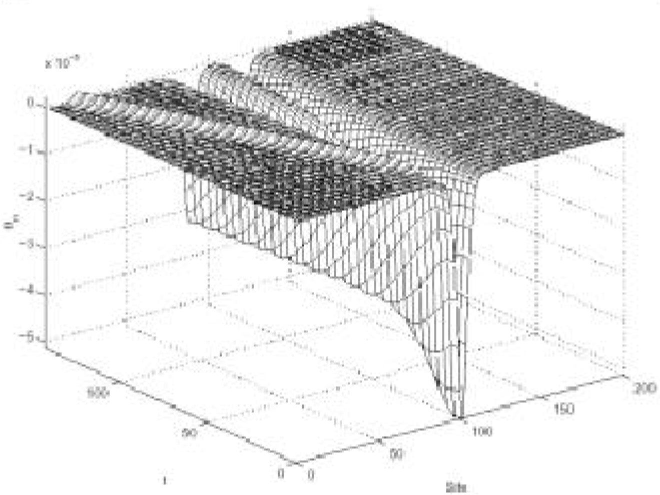
Movability regimes

1. Radial movability regime
   - α small ($\alpha \leq 0.0005$)
   - Only radial movability
2. Mixed regime
   - Intermediate α ($0.0005 \leq \alpha \leq 0.05$)
   - Both types of movability
3. Twist movability regime
   - Larger α ($0.05 \leq \alpha$)
   - Only twist movability

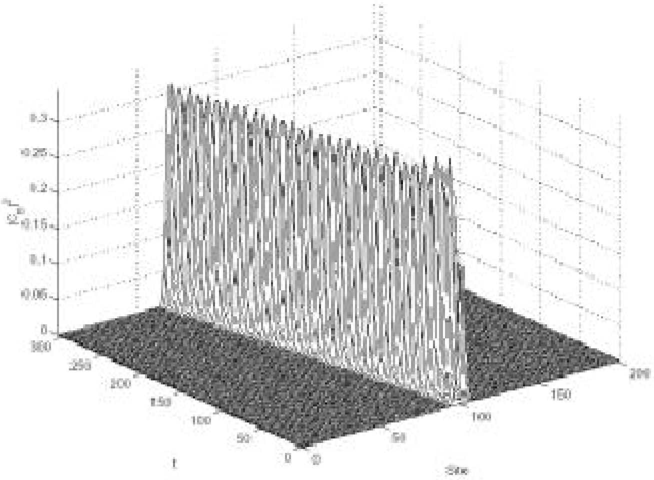
Homogeneous chain. Radial movability regime

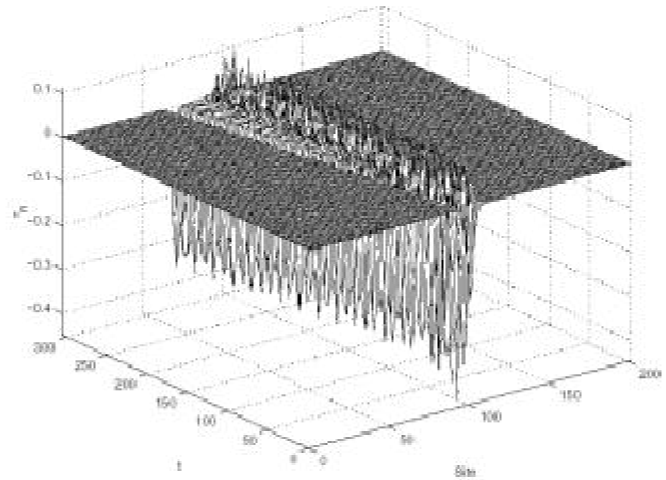


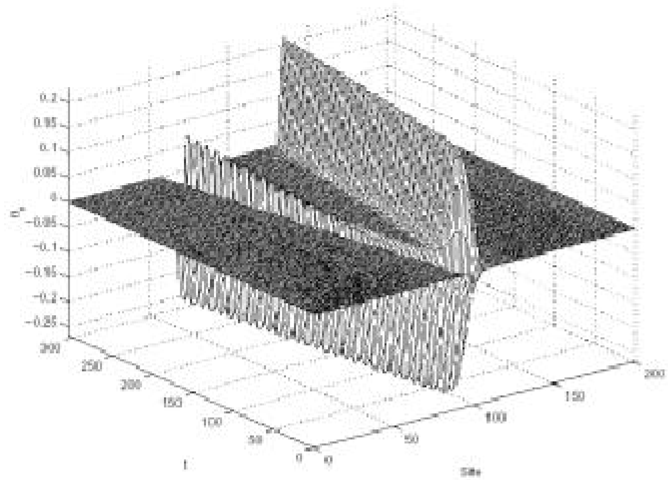




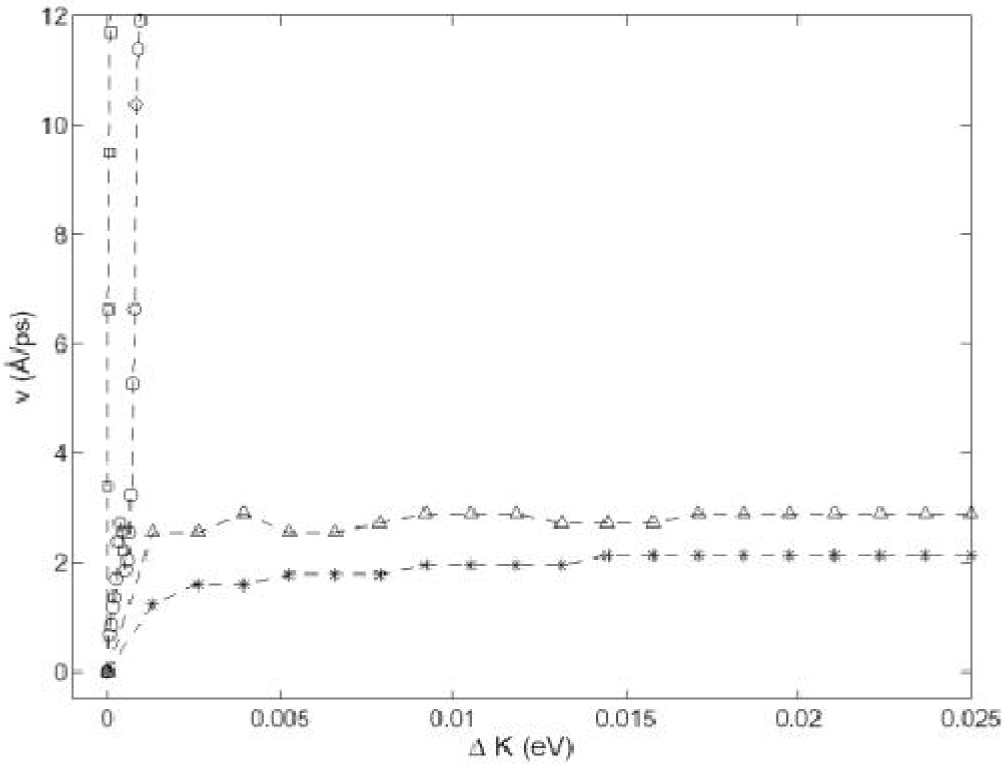
Homogeneous chain. Twist movability regime







Comparison between different regimes



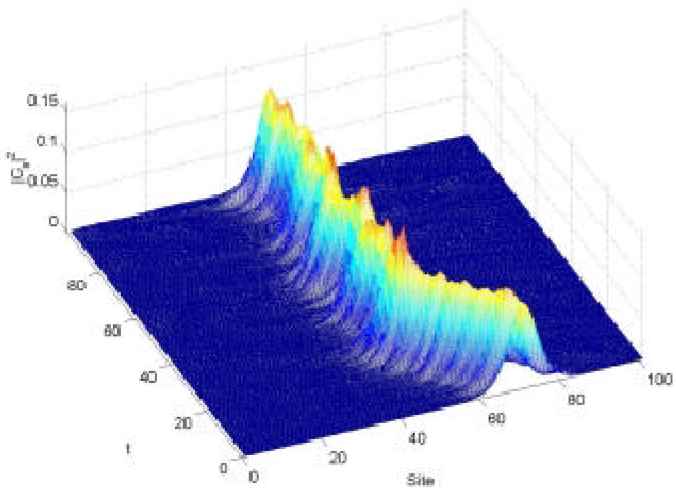
Squares: Radial regime; Circles: Mixed regime, radial activation; Triangles: Mixed regime, angular activation; Starts: Twist regime

Disordered chain

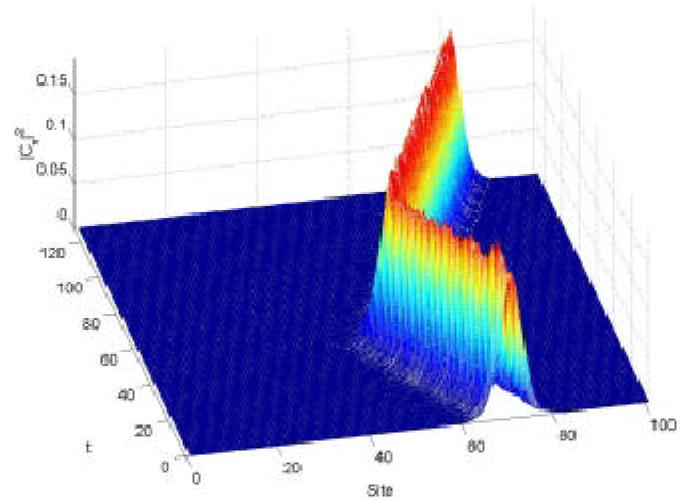
Mobile polarons survive to a certain degree of random disorder, $\Delta E \leq \Delta E_c$

1. Radial movability: Very sensitive to disorder ($\Delta E_c \sim 0.05$)
2. Twist movability: Very robust with respect to disorder ($\Delta E_c \sim 0.5$)
3. Mixed regime:
   - Twist polarons: robust and slower polarons
   - Radial polarons: fast polarons but sensitive to disorder

DNA with a base pair inhomogeneity. Radial movability regime



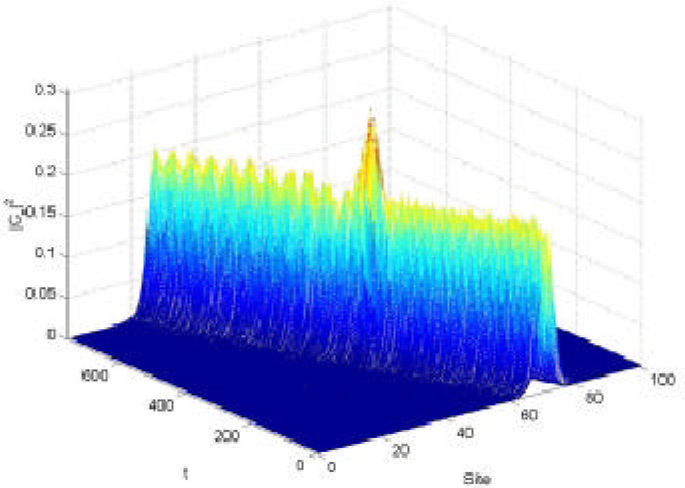
Trapping phenomenon due to the interaction between a moving polaron in a G-C chain with a A-T base pair

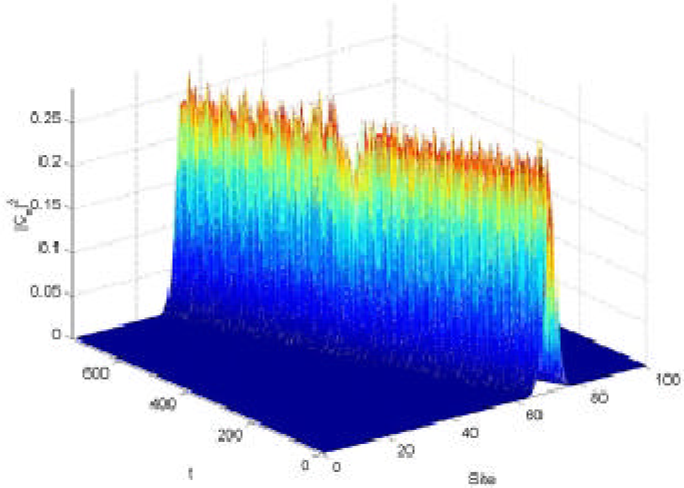


Reflection phenomenon due to the interaction between a moving polaron in a A-T chain with a G-C base pair

Radial polarons are either trapped or reflected by the inhomogeneity

DNA with a base pair inhomogeneity. Twist movability regime





Transmission phenomenon due to the interaction between a moving polaron in a G-C chain with a A-T base pair. The inhomogeneity acts as a potential well.

Transmission phenomenon due to the interaction between a moving polaron in a A-T chain with a G-C base pair. The inhomogeneity acts as a potential barrier.

Twist polarons travel across the base pair inhomogeneity

Inhomogeneous chain. Movability

	Radial movability regime	**Twist movability regime**	**Mixed regime Radial activation**	**Mixed regime angular (and radial) activation**
A-T chain	Reflection	Transmission	Reflection	Transmission
G-C chain	Trapping	Transmission	Trapping	Transmission

Conclusions

- Radial and twist polarons in a 3D, semi-classical, tight-binding DNA model.
- Caracteristics of the movement of the different polarons
  - Velocity
  - Sensitivity to random disorder
- Interaction with a base-pair inhomogeneity
  - Twist polarons travel across the impurity
  - Radial polarons are either refracted or trapped.

References (http://www.us.es/gfnl)

1. D Hennig, JFR Archilla and J Agarwal, *Nonlinear charge transport mechanism in periodic and disordered DNA,* Physica D 180(3-4):256-272, 2003.
2. JFR Archilla, D Hennig and J Agarwal. *Charge transport in a nonlinear, three-dimensional DNA model with disorder*, in Localization and Energy Transfer in Nonlinear Systems, World Scientific, 2003
3. F Palmero, JFR Archilla, D Hennig and FR Romero, *Effect of base-pair inhomogeneities on charge transport along DNA mediated by twist and radial polarons*, New Journal of Physics, to appear
4. D Hennig, E Starikov, JFR Archilla and F Palmero, *Charge transport in poly(dG)-poly(dC) and poly(dA)-poly(dT) DNA polymers.* Journal of Biological Physics, 2003. To appear.
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