Long-Range Charge Transfer in DNA through Polaron Diffusion

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Recent experimental evidence shows that the \(\pi\) orbitals along the stacking of base pairs can facilitate the long-range charge transfer in DNA [5–7]. Proton motion in the base pair hydrogen bonds has also been found to affect the transfer rate. To explain this behavior we propose a model considering interactions of doped charges with hydrogen bonds and vibrations in DNA. The charge trapped by either protons or vibrations can cause structural distortions leading to polaron formation. By further considering polaron diffusion in DNA we find that the charge transfer rate derived from the diffusion coefficient is in agreement with the experimental results [33].

I. INTRODUCTION

The question on whether DNA is a conductor or an insulator is still controversial due to the different results obtained in the measurements of DNA conductivity\(^1\)\(^–\)\(^4\). The related issue of long-range charge transfer in DNA which is associated with the problem of charge mobility has also been discussed for many years with an equal amount of heated debate. It has been suggested that the overlap of the electronic \(\pi\) orbitals along the stacked base pairs provides a pathway for charge propagation over 50 Å and the reaction rate between electron donors and acceptors does not decay exponentially with the distance\(^5\)\(^–\)\(^7\).

A multiple-step hopping mechanism was recently proposed to explain the charge transfer behavior in DNA\(^8\). In this theory, the single G-C base pair is considered as a hole donor due to its low ionization potential if compared to the one on A-T base pairs. Long-range charge transfer is accomplished by a series of incoherent charge tunneling events between two nearest G-C base pairs separated by A-T pairs. The probability of the short-range tunneling (super-exchange) strongly depends on the distance between the two G-C base pairs. However, recent experimental results for the reaction rate only showed a very slight distance dependence for the charge hole transfer through more than three A-T base pairs\(^9\). It is now believed that a hole can be created in a A-T base pair by thermal activation from a G-C base pair when charge tunneling becomes unlikely over a long distance\(^10\)\(^–\)\(^12\). The long-range hopping can, therefore, be carried out through a long A-T bridge just as by a series of short-range tunnelings in the G-C pairs. Although this mechanism seems to give a good interpretation of the charge transfer in DNA, the cause of the incoherent charge hopping over localized states of the hole carriers with such low reaction rate \((10^9 \sim 10^6 s^{-1})\) has not yet been well understood.

Besides the multiple-step hopping mechanism, polaron motion has also been considered as a possible mechanism to explain the phenomena of DNA charge transfer\(^13\)\(^,\)\(^14\). The charge coupling with the DNA structural deformations can create a polaron and cause a localized state. From the study of the dynamical properties in one-dimensional systems, it is known that the polaron behaves as a Brownian particle that collides with the low energy excitations of its environment which acts as a heat bath\(^15\). This diffusive behavior is very similar to the multiple-step hopping mechanism based on a random walk model, although the former occurs in a continuous media while the latter is considered on the discrete sites of a lattice. Thus, the behavior of the incoherent charge hopping can be understood as polaron diffusion.

For most physical systems acoustical and optical phonons are the main lattice excitations. However, when a system contains hydrogen bonds, proton motions also need to be considered. Instead of oscillatory motions in a single-minimum potential, protons can tunnel from one side of a hydrogen bond to another in a double-minimum potential\(^16\). This proton tunneling causes interstrand charge hopping and is important in spontaneous mutations. Since genetic information can be preserved only when a nucleotide base is matched with its complementary one by hydrogen bonding, the positions of protons are the main identification for DNA polymerases to make replications with high fidelity. However, the reaction of proton transfer could generate tautomeric base pairs and destroy the fidelity\(^17\). The mechanism of the proton tunneling in the isolated base pairs and its possible biological implications were widely discussed by Löwdin many years ago\(^18\). It has also been suggested that proton transfer and charge conduction in stacked base pairs are affected by each other\(^19\)\(^–\)\(^21\). The recent experimental results on the influence of mismatched base pairs show that the proton transfer is required for the long-range charge transfer\(^22\). Thus the effect of proton tunneling will be discussed here.

The paper is organized as follows. The model for charge motion in DNA will be introduced in the next Section, considering charge hopping, possible fluctuations in structure and their interactions. We will present the
formation of a polaron in Section III, together with the
study of polaron stability and proton delocalization in the
hydrogen bonds. In Section IV, we calculate the polaron
diffusion coefficient and the results will be compared with
the experimental reaction rate of charge transfer in DNA.
Section V contains our conclusions.

II. THEORETICAL MODEL

Although biologically functional DNA is an aperiodic
system, we will focus here on a periodic DNA structure.
Since the polaron state discussed here is highly localized
(see below), we believed that the periodicity condition is
not very constraining when we consider charge transfer
over intermediate distance. In fact, in some experimen-
tal setups one looks for charge transfer between two G-C
pairs which are separated by many (periodic) A-T pairs.
As we can see in Fig. 1, following hole injection into the
system, the charge localizes in the G-C base pair but can
migrate from one unit cell to another. The essential in-
redients in our model are (i) the electronic system, that
realizes the charge conduction; (ii) the possible structural
fluctuations, which include phonons and proton motion
in hydrogen bonds; and (iii) the coupling between the
charges and the structure.

\[ H_{\text{ph}} = \sum_i \left[ \frac{P_i^2}{2M} + \frac{1}{2} M \omega_0^2 (u_{i+1} - u_i)^2 \right] + \sum_i \frac{P_i^2}{2M} + \frac{1}{2} M \omega_0^2 v_i^2 , \]

where \( u_i \) and \( v_i \) are the lattice displacement and the in-
ternal vibration coordinate of the \( i \)-th unit cell, \( p_i \) and \( P_i \)
are their conjugated momentum, respectively, \( M \) is the
mass of a unit cell, \( \omega_0 \) the oscillation frequency of the
optical phonon and the dispersion relation of the acoustical
motion is: \( \omega_s (k) = c_s k \), where \( c_s \) is the sound velocity
along the chain.

The charge coupling to acoustical phonons is given by
the Su-Schrieffer-Heeger model and the interaction
with optical phonons is described via the molecular crys-
tal model of Holstein. Thus the total electron-phonon
interactions of DNA system is

\[ H_{\text{e-ph}} = \sum_i \frac{\gamma_s}{a} (u_{i+1} - u_i) (C_i^\dagger C_{i+1} + C_{i+1}^\dagger C_i) + \sum_i \gamma_v v_i C_i \] (3)

where \( \gamma_s \) and \( \gamma_v \) are the coupling constants. In sys-
tems with half-filled conduction band, such as one-
dimensional \( \pi \)-conjugated polymers, the SSH term gener-
ates dimerization in the ground state (Peierls instability)
and forms solitons in the excited states. However, for
DNA, considered here as a band insulator, both interac-
tions can generate lattice distortions and lead to polaron
formation when a charge is doped into the molecule. The
second term in (3) usually induces small polarons in ionic
crystals.

For the proton motion in the hydrogen bonds we use a
two-level system to describe the tunneling behavior (see
Fig. 2): 

\[ H_{\text{\sigma}} = \frac{1}{2} (-\varepsilon \sigma^z + t \sigma^x) , \]

where \( \sigma^z \) and \( \sigma^x \) are Pauli matrices, \( \varepsilon \) is the energy bias
between the two localized proton states and \( t \) is the tun-
neling matrix element. The ratio \( t/\varepsilon \) provides information
of the proton motion in the system. When the ratio

\[ \frac{1}{a} \]

the lattice spacing and the wave vectors \( k \) is \( 2\pi n/L \) with \( n \) as an integer and the system size \( L \). In
the continuum limit (\( a \rightarrow 0 \)), the Hamiltonian describes
the kinetic energy of charge particles with a Bloch wave-
function and effective mass \( m \), where \( h^2/2ma^2 \) is used
in replacement of the overlap integral \( t_0 \) of the discrete model.

For the phonon part of the structural fluctuations in
DNA, we consider that the relative motions between two
different base pairs can be represented by an acoustical
phonon mode and the vibrational motion inside a base
pair by optical phonons. These modes represent lattice
distortions, such as sliding, twisting or bending. The
Hamiltonian that describes these modes is:

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is small, the protons are localized in one side of the hydrogen bonds. However, when it is large, the protons are delocalized. As we can see in Fig. 2(a), the normal G-C base pair is the lower energy state and its tautomeric form \((G^*≡C^*)\) is an excited state. When \(\varepsilon \gg t\), the probability of having a tautomeric form is extremely small. However, in Fig. 2(b), the radical cation of a G-C base pair has almost the same energy as its tautomeric form \(^2\). i.e., \(\varepsilon \lesssim t\). In this case, the proton state becomes delocalized in the hydrogen bonds.

![FIG. 2. Proton transfer in the hydrogen bond of a DNA base pair described by a two-level system. (a) For a neutral G-C base pair, proton transfer is unlikely to cause the tautomeric form and we have \(\varepsilon \gg t\). (b) The radical cation of a G-C base pair only has a little energy difference with the tautomeric form, i.e., \(\varepsilon < t\).](image)

To model the coupling between the protons in the hydrogen bond and the charges in the DNA strand, we use:

\[
H_{e-\sigma} = \gamma_\sigma (\sigma_i^x - 1) n_i ,
\]

where \(\gamma_\sigma\) is the coupling intensity and \(n_i = C_i^\dagger C_i\) is the number of charges on the \(i\)-th site. When the proton is in the lower energy state \((\sigma_i^x = 1)\) or there is no charge around the hydrogen bond \((n_i = 0)\), the coupling vanishes. Although there are many hydrogen bonds in the base pairs of a unit cell, it is not necessary to consider all of the possible proton transfers since at room temperature the transitions to the tautomeric forms are very unlikely to occur. We only need to consider the hydrogen bond with the most probable proton transfer with charge injection into the unit cell, i.e., only one two-level system is required for each unit cell.

By giving a total Hamiltonian as the sum of (1), (2), (3), (4) and (5) we have our complete model for charge motion in DNA.

### III. FORMATION OF POLARON IN DNA

Although polaron formation has already been considered very important for charge motion in DNA\(^{13,14,30}\), the effect of the proton motion in the hydrogen bonds has not yet been discussed in the literature. Here, to investigate this factor, we analytically solve our model by taking the Hamiltonians into the continuum limit. The carrier wavefunction \(\psi(x,t)\) and the phonon modes \(u_v(x,t)\) are considered as functions of time and a continuous coordinate \(x\). The equations of motion for the variables in the problem can be studied in the semiclassical limit \((\hbar \to 0)\) and by a change of variables \(\lambda = x - vt\), where \(v\) is the polaron velocity. In this case, we have:\n
\[
\begin{align*}
\frac{u_{s(v)}(x,t)}{u_{s(v)}(x-\nu t)} &= \frac{u_{s(v)}(\lambda)}{\psi(x,t)} = \phi_0(\lambda) \exp\left[\frac{\gamma_\sigma}{\hbar} (m v x - E_0 t)\right], \\
\end{align*}
\]

where \(E_0\) is the binding energy of the polaron, and the problem reduces to a nonlinear Schrödinger equation for the wavefunction \(\phi_0\):

\[
\begin{align*}
\frac{\hbar^2}{2m} \frac{d^2 \phi_0}{d\lambda^2} + \left(E_0 - \frac{mv^2}{2} + \gamma_\sigma \sin^2 \theta\right) \phi_0 &+ \left(\frac{\gamma_\sigma^2}{\rho (c_s^2 - v^2)} + \frac{\gamma_\sigma^2}{\rho \omega_0^2}\right) \phi_0^3 = 0 ,
\end{align*}
\]

where \(\rho\) is the mass density \(M/a\), \(\sin^2 \theta\) represents the positions of the protons in the hydrogen bonds and is expressed by:

\[
\sin^2 \theta = \frac{1}{2} \left(1 - \frac{\varepsilon}{\sqrt{\varepsilon^2 + t^2}}\right),
\]

where \(\varepsilon = \varepsilon - \gamma_\sigma \phi_0^2\). The interaction energy between the carriers and protons, \(\gamma_\sigma \sin \theta\), is significant only when the charge trapped by the hydrogen bond causes proton delocalization. Therefore, for small value of \(t\) compared to \(\varepsilon\), we can use the approximation:

\[
\sin^2 \theta \approx \frac{\gamma_\sigma \phi_0^2}{2t} - \frac{1}{2t} (\varepsilon - t) , \quad \gamma_\sigma \phi_0^2 \leq \varepsilon - t , \quad \varepsilon - t < \gamma_\sigma \phi_0^2 \leq \varepsilon + t .
\]

\[
\left\{ \begin{array}{ll}
\gamma_\sigma \phi_0^2 > \varepsilon + t \\
\end{array} \right.
\]

\[
\left\{ \begin{array}{ll}
\gamma_\sigma \phi_0^2 < \varepsilon - t \\
\end{array} \right.
\]

(9)
From the continuity of the polaron wavefunction, equation (7) can be analytically solved and the solution is found as combinations of Jacobian elliptic functions. When the charge coupling with the protons is small, i.e., \( \gamma_\sigma (\phi_0)^2 \leq 1 - t \) (where \( \langle \phi_0 \rangle \) is the maximum value of \( \phi_0 \)), the polaron is purely formed by the interaction with phonons and the proton motion has no effect on the wavefunction or the binding energy. The ground state solution is simply expressed as

\[
\phi_0 (\lambda) = \sqrt{g} \sech (2g\lambda),
\]

and the binding energy is

\[
E_0 = -\frac{2\hbar^2 g^2}{m},
\]

where

\[
g = \frac{m}{\hbar^2} \left[ \frac{\gamma_s^2}{\rho (c_s^2 - v^2)} + \frac{\gamma_v^2}{\rho \omega_d^2} \right].
\]

It is easy to see that \( 1/g \) represents the size of the polaron. To calculate the binding energy and the size of a static polaron, we consider that a hole carrier is doped into a poly(G)-poly(C) DNA. The effective mass of the carrier \( m = \hbar^2 / 2t_0 a^2 \) is of order \( 20 m_e=1.8219 \times 10^{-29} \text{kg} \) (\( m_e \) the mass of an electron) obtained from band structure calculations, which give a bandwidth of about 0.04 eV\(^3\). For the charge coupling strength \( \gamma_\sigma \) and \( \gamma_v \) generated by the acoustical and optical phonons in (7), since the energy difference between the adiabatic and vertical ionization potentials of a G-C base pair is about 0.5 eV\(^2\), the optical part is given as about 0.5 eV \( \times 3.38 \text{Å} = 1.7 \text{ eV Å} \). The acoustical part is assumed to be smaller, so we consider the value of the coupling strength as approximately 2 eV Å. From these results, we find that the polaron is 0.75Å wide with binding energy \( E_0 = -1.35 \text{ eV} \) and, therefore, it is entirely localized in a base pair. For the weak coupling to hydrogen bonds (\( \gamma_\sigma \leq 0.2 \text{ eV} \)), the protons are localized in their original positions and the tautomeric base pair is unlikely to be generated.

As the coupling constant \( \gamma_\sigma \) becomes larger, proton transfer can be induced since the charge trapped into a base pair reduces the bias energy \( \varepsilon \) dramatically. By considering the bias energy \( \varepsilon \) and the tunneling matrix element \( t \) of order 0.3 eV Å\(^{-1}\) and 0.03 eV Å\(^{-1}\), respectively, we find that the binding energy of the polaron \( E_0 \) is \(-1.45 \text{ eV} \) for \( \gamma_\sigma = 0.226 \text{ eV} \), which is more stable than the above with the same values of the effective mass \( m \) and the coupling strength \( \frac{\gamma_s^2}{\rho c_s^2} + \frac{\gamma_v^2}{\rho \omega_d^2} \). The ground state wavefunction consists of two parts, the central and the external. For the external part, the wavefunction behaves as a hyperbolic secant function since it should vanish as \( \lambda \to \infty \). In the central part of the polaron, the wavefunction is described by a Jacobian elliptic functions of second kind and the proton in the hydrogen bond is delocalized since the value of \( \sin^2 \theta \) is large. For the base pair in the center of the polaron, its tautomeric form can be easily induced.

The relation between the coupling constant \( \gamma_\sigma \) and the binding energy is shown in Fig. 3, where we have 1.7 eV Å for the value of the coupling strength \( \frac{\gamma_s^2}{\rho c_s^2} + \frac{\gamma_v^2}{\rho \omega_d^2} \). When \( \gamma_\sigma \) is larger than 0.24 eV, the proton becomes delocalized in the hydrogen bond and the charge binding energy increases rapidly. The local deformations from the charge interaction with phonons can also stabilize the polaron as shown in Fig. 4 with \( \gamma_\sigma = 0.25 \text{ eV} \). The increase of the couplings \( \gamma_\sigma \) or \( \gamma_v \) also induces proton transfer since the carrier is strongly attracted in the base pair.

The effect of proton transfer has been observed in experiments on the influence of mismatches in the DNA long-range charge transfer. Mismatched base pairs are found to cause a large drop in the charge transfer rate due to the weakening of hydrogen bonds. For a normal G-C base pair, the positive charge in the radical cation can be distributed by both nucleotides through the delocalization.
IV. POLARON DIFFUSION IN DNA

The dynamic properties of a polaron induced by the carrier interaction with acoustical and optical phonons in one dimension were explored many years ago\(^\textsuperscript{15}\). In the strong coupling limit, it is found that the polaron moves as a Brownian particle interacting with the light particles of the environmental heat bath. To investigate these results in the context of DNA, we first consider the case without proton coupling (i.e., \(\gamma_\sigma = 0\)).

The damping parameter of the optical polaron motion at high temperature \((k_B T \gg \hbar \omega_0)\) is given by\(^\textsuperscript{15}\):

\[
\gamma_o = \frac{\pi k_B T g_o^2}{6 \omega_0 M_o},
\]

where \(M_o\) is the classical polaron mass:

\[
M_o = \frac{m}{8} \left( \frac{E_o}{\hbar \omega_0} \right)^2
\]

and

\[
g_o = \frac{m}{2} \frac{\gamma_o^2}{\hbar^2 \rho \omega_0^2} = \frac{\sqrt{8mE_o}}{\hbar^2}.
\]

Here, we use \(E_o\) as the binding energy from the optical phonons. For acoustical phonons we have the damping parameter:

\[
\gamma_a = \frac{315 k_B T g_a}{32\pi^4 c_s M_o},
\]

where the classical polaron mass for the acoustical case is

\[
M_a = \frac{32m}{3} \left( \frac{E_a}{\hbar c_s g_a} \right)^2
\]

and

\[
g_a = \frac{m}{2} \frac{\gamma_a^2}{\hbar^2 \rho c_s^2} = \frac{\sqrt{8mE_a}}{\hbar^2}
\]

with \(E_a\) the binding energy from the acoustical phonons. The random walk motion of the polaron in the long time limit \((t \gg 1/\gamma_o \text{ or } t \gg 1/\gamma_a)\) is governed by the one-dimensional diffusion equation:

\[
\frac{dn}{dt} = D \frac{\partial^2 n}{\partial x^2},
\]

where \(n(x,t)\) is the density of the particles and \(D\) is the diffusion coefficient. The relation between the damping parameter and the diffusion coefficient is given by the Einstein relation:

\[
D_o(a) = \frac{k_B T}{\gamma_o(a) M_o(a)},
\]

where we use \(D_o(a)\) for the diffusion coefficient in the optical (acoustical) case.

As we can see from the scheme of the multiple-step hopping mechanism in Fig. 1, the hole hopping from a G-C base pair to its nearest neighbor can be viewed as an oxidation-reduction reaction with a rate constant \(k\). According to the rate law of a first-order reaction, the concentration of the charge at the \(i\)-th G-C base pair, \(n_i\), is described by

\[
\frac{dn_i}{dt} = k (n_{i+1} + n_{i-1} - 2n_i).
\]

In the continuum limit, when the distance between two nearest neighboring G-C base pairs, \(a\), is very small, we have

\[
\frac{dn_i}{dt} = ka^2 \left( \frac{n_{i+1} - n_{i-1}}{a} - \frac{n_i - n_i}{a} \right) \to ka^2 \frac{\partial^2 n_i}{\partial x^2}.
\]

By comparing this equation with (19), we have the correspondence from the diffusion coefficient in the continuous medium to the charge transfer rate of the discrete model:

\[
D \to ka^2.
\]

Thus, instead of a diffusion coefficient, we can use the discrete characteristic \(t_0\) to estimate the rate constant. From (20) and (13), we have

\[
D_o = \frac{k_B T}{\gamma_o M_o} \frac{6 \omega_0}{\pi g_o^2} = \frac{3 \omega_0}{2\pi E_o} \frac{\hbar^2}{2ma^2} \to \frac{3 \omega_0}{2\pi E_o} a^2.
\]

By the correspondence (23), the rate constant for the optical case is obtained as

\[
k_o = \frac{3}{2\pi} t_0 \omega_0.
\]

We can also derive the rate constant for the thermal activation by the acoustical phonons as

\[
k_a = \frac{64\pi^4 \rho c_s^3 t_0}{315 \gamma_s^2}.
\]

For poly(G)-poly(C) DNA, the oscillation frequency is \(\omega_0 \approx 10^{11}\) Hz from the theoretical and experimental results\(^\textsuperscript{32}\). The theoretical calculation has shown the valence bandwidth of 0.04 eV\(^\textsuperscript{31}\). By considering that the binding energy \(E_o\) is about 1 eV, we have the reaction rate \(k_o \approx 10^9\) s\(^{-1}\). This estimate is in agreement with the experimental result for electron transfer in DNA which gives \(k_o \approx 10^8 \sim 10^{9}\) s\(^{-1}\).
This low reaction rate is generated by two factors in equation (25), the hopping probability $\frac{t_0}{E_0}$ and the oscillation frequency $\omega_0$. Due to the strong structural fluctuations at room temperature, the charge hopping could only happen when the local structure of the polaron achieves an effective configuration. For each period of structural oscillation, the probability to have a successful hopping is of order $\frac{t_0}{E_0}$. Thus, the charge transfer rate is determined by the frequency of reaching the effective configuration $\omega_0$ and the probability $\frac{t_0}{E_0}$. The behavior of the incoherent charge hopping is now described by the polaron motion under thermal fluctuations.

Another way to study the diffusion coefficient in the optical case is to consider the mean square displacement:

$$\left\langle \langle \Delta x \rangle^2 \right\rangle = \left\langle x^2 \right\rangle = \int_{-\infty}^{\infty} x^2 \phi_0(x) \phi_0(x) \, dx = \frac{\pi^2}{3\sigma^2}, \quad (27)$$

and we may rewrite (24) as:

$$D_o = \frac{72}{\pi^2} \frac{\omega_0}{2\pi} \left\langle \langle \Delta x \rangle^2 \right\rangle \sim \frac{\omega_0}{2\pi} \left\langle \langle \Delta x \rangle^2 \right\rangle \quad (28)$$

Now, we can see that, for every oscillation period $2\pi/\omega_0$, the collisions by the light particles from the environment spread the polaron with the mean-square displacement $\left\langle \langle \Delta x \rangle^2 \right\rangle$. The diffusion in the continuous medium, however, needs to be corrected when we make the correspondence since the size of the polaron is smaller than the lattice spacing. Therefore, the real reaction rate of charge transfer between the discrete base pairs should be smaller than our results. Nevertheless, the order of the charge transfer rate can still be estimated from the simple expression:

$$k \sim \omega_0 \frac{\left\langle \langle \Delta x \rangle^2 \right\rangle}{2\pi \sigma^2} \quad (29)$$

We can also estimate the reaction rate $k_o$ in the acoustical case from the experimental data for the sound velocity $c_s \approx 10^5$ cm s$^{-32}$. Assuming $\frac{2\pi^2}{\rho\sigma^2} \approx .34$ eV Å, we have $k_a \approx 10^{13}$ s$^{-1}$ which is four order of magnitude larger than $k_o$. A faster charge transfer rate is expected if the polaron is purely generated by the acoustical phonons. However, if optical phonons or proton transfer are involved, a smaller value of the reaction rate should be expected due to the increase of the binding energy in those cases.

V. CONCLUSIONS

In this paper we studied the charge conduction in periodic DNA focusing on the effect of the structural fluctuations which includes the proton motion in hydrogen bonds. We found that the charge trapped by hydrogen bonds or phonons can generate a polaron state and the coupling to the protons can induce proton transfer in a base pair. The polaron moving with small velocity is seen as a Brownian particle colliding with the light particles of the environment. This diffusion process corresponds to a multiple-step hopping mechanism in the discrete model. From this correspondence, the reaction rate of the long-range charge transfer in DNA can be derived from the diffusion coefficient, and it was found that the result predicted in the optical case is in agreement with the experimental results.

Although we did not include the effect of proton transfer on the polaron diffusion, it is expected to be crucial in the dynamics of charge hopping. From electrochemical experiments, a decrease of charge transport along the DNA with mismatched base pairs is found. This behavior might be explained by the effect of the proton motions in the hydrogen bonds and we will explore this possibility in future work.

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