Vibronic Dephasing Model for Coherent-to-Incoherent Crossover in DNA

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In this work we investigate the interplay between coherent and incoherent charge transport in cytosine-guanine (GC) rich DNA molecules. Our objective is to introduce a physically grounded approach to dephasing in large molecules and to understand the length dependent charge transport characteristics and especially the crossover from coherent tunneling to incoherent hopping regime at different temperatures. Therefore, we apply the vibronic dephasing model and compare the results to the Büttiker probe model which is commonly used to describe decoherence effects in charge transport. Using the full ladder model and simplified 1D model of DNA, we consider molecular junctions with alternating and stacked GC sequences and compare our results to recent experimental measurements.

I. INTRODUCTION

Scientific interest in charge transport characteristics of biological molecules, in particular double-stranded DNA, has grown over recent decades because of its fundamental importance in live sciences and potential for electronic applications. Many experiments on charge transfer and transport have been realized showing a wide range of contradictory outcomes covering insulating, semiconducting, and conducting behaviour. This large variety of experimental results reflects the complexity of charge transfer processes in DNA and especially the influence of environmental effects characterized by measurement conditions as well as the structural conformation. At low temperatures the transport should be coherent, weakly influenced by interaction of electrons with conducting environment. On the other hand, the main source of dephasing at finite temperatures is elastic electron-vibron interaction due to large number of soft vibrations in biological molecules.

Experimental as well as theoretical studies revealed two fundamental charge transport mechanisms in DNA. Over short distances and at low temperatures (actually, as we will see, even room temperature can be low enough) charge carriers move coherently through delocalized molecular orbitals. In the case of the Fermi level position inside the conductance gap, this coherent tunneling regime is represented by an exponentially length dependent transmission. In the case of coherent resonant transport, the conductance is oscillating function of the length because of interference.

However, in the long-range regime at high temperatures charge carriers localize on the bases and the main charge transfer occurs through incoherent hopping between them. This can be modeled by multistep hopping events between purine bases, i.e., bases with the lowest oxidation potentials and shows a linear length dependence of transmission at high temperatures.

The calculation of coherent charge transport is based on the Landauer-Büttiker approach (also known as the scattering method), which is implemented usually with a help of the Green function technique.

Ⅰ. INTRODUCTION

However, due to external influences and temperature dependent vibrations a coherent theory is often not sufficient. In recent years there were some studies to expand the coherent theories to include decoherence effects.

In the framework of the Green function approach dephasing resulting from environmental effects can be accounted for using the Büttiker probe model. It is based on the idea of virtual probes coupled to the system. This coupling controls the strength of the dephasing and is chosen empirically.

More physical is the vibronic dephasing, the contribution of which is especially important at high temperatures. Vibrational effects can be taken into account through the corresponding self-energy, which is dependent on the Green function at the same energy in the elastic approximation (see Sec. II A). In this paper we consider local and elastic dephasing models, which are most important at low and intermediate temperatures at low applied voltages, corresponding to typical experimental conditions.

The recent experiment has shown intermediate coherent-incoherent charge transport characteristics at room temperature in GC-rich DNAs. The phenomenological explanation based on the incoherent hopping approach with addition of coherent transport was suggested. It is important to understand transition from coherent to incoherent transport starting from unified microscopic approach. The model of such crossover was studied recently by using Büttiker probe model of environmental caused dephasing. In this paper we will investigate decoherence effects in DNA structures based on the vibronic dephasing model proposed by some of us earlier. Besides, we implemented both methods in the open software package DFTB+XT.
II. ELASTIC DEPHASING

In this section we outline the nonequilibrium Green function formalism for systems with elastic dephasing. The total Hamiltonian in the case of two equilibrium electrodes can be written as

\[ \hat{H} = \hat{H}_L + \hat{H}_{LM} + \hat{H}_M + \hat{H}_{MR} + \hat{H}_R, \]

(1)

where \( \hat{H}_{L/R} \) describes the left/right lead and \( \hat{H}_M \) represents the DNA molecule. The coupling between leads and molecule is defined by \( \hat{H}_{LM} \) and \( \hat{H}_{MR} \). The corresponding retarded and advanced Green functions of the molecule are then given by

\[ G^{R}(\varepsilon) = \left[ i\varepsilon - \hat{H}_M - \Sigma^{R}_{L}(\varepsilon) - \Sigma^{R}_{R}(\varepsilon) - \Sigma^{\text{dep}}(\varepsilon) \right]^{-1}, \]

(2)

\[ G^{A}(\varepsilon) = (G^{R}(\varepsilon))^\dagger, \]

(3)

where \( \Sigma^{R/L}_{L,R}(\varepsilon) \) is the retarded self-energy of the left/right electrode and \( \Sigma^{\text{dep}}(\varepsilon) \) is the self-energy due to dephasing. In the case of elastic dephasing, considered here, the self-energy at some energy does not depend on other energies, but can be a function of the Green function at the same energy. We do not take into account electron-electron interactions. In the case of dephasing in strongly interacting many-body systems other approaches may be required.

In order to determine the transport properties, we use the many-body nonequilibrium Green function formalism. The details can be found in Refs.\textsuperscript{[24,25]} Within this approach the current can than be calculated via the Meir-Wingreen formula

\[ I = \frac{ie}{\hbar} \int_{-\infty}^{\infty} \text{Tr} \left[ (\Gamma_L(\varepsilon) - \Gamma_R(\varepsilon)) G^{<}(\varepsilon) \right. \\

\left. + (f_L(\varepsilon)\Gamma_L(\varepsilon) - f_R(\varepsilon)\Gamma_R(\varepsilon)) (G^{R}(\varepsilon) - G^{A}(\varepsilon)) \right] d\varepsilon, \]

(4)

where the broadening function can be determined by:

\[ \Gamma_{L/R}(\varepsilon) = i(\Sigma_{L/R}(\varepsilon) - \Sigma^R_{L/R}(\varepsilon)). \]

The lesser/greater Green functions are defined through the Keldysh equation

\[ G^{<}(\varepsilon) = G^{R}(\varepsilon)\Sigma^{<}_{\text{tot}}(\varepsilon)G^{A}(\varepsilon), \]

(5)

where the total self-energy \( \Sigma^{<}_{\text{tot}}(\varepsilon) = \Sigma^{<}_{L}(\varepsilon) + \Sigma^{<}_{R}(\varepsilon) + \Sigma^{\text{dep}}(\varepsilon) \) contains contributions from the leads as well as from interactions. The former can be expressed by the equilibrium fermi functions of the leads \( \bar{f}_{L/R}(\varepsilon) \) via

\[ \Sigma^{<}_{L,R}(\varepsilon) = i\bar{f}_{L/R}(\varepsilon)\Gamma_{L/R}(\varepsilon), \]

(6)

\[ \Sigma^{<}_{L/R}(\varepsilon) = -i(1 - \bar{f}_{L/R}(\varepsilon))\Gamma_{L/R}(\varepsilon). \]

(7)

By assuming that the total self-energy \( \Sigma_{\text{tot}} \) contains just contributions of the leads, eq. (6) would yield the Landauer formula for coherent transport.

A. Vibronic dephasing (VD) model

Incoherence effects on charge transport can be taken into account by including the electron-vibron interaction. This can be realized by an additional self-energy \( \Sigma_{\text{vib}} \).

In order to obtain an expression for this self-energy we assume that the Hamiltonian of the central device can be written as:

\[ \hat{H}_M = \hat{H}_el + \hat{H}_{e-v}. \]

(8)

The electronic structure of the molecule is given by \( H_{el} \) and the electron-vibron interaction \( H_{e-v} \) can be written as:

\[ \hat{H}_{e-v} = \sum_{i,j,\alpha} M_{ij}^{\alpha} (a_\alpha + a_\alpha^\dagger) d_i^\dagger d_j, \]

(9)

where \( a(a^\dagger) \) describes the annihilation (creation) operators of vibrons and \( d(d^\dagger) \) the same for electrons. The coupling between electrons and vibrons in mode \( \alpha \) is described by \( M^{\alpha} \). The corresponding self-energy reads (see details in Ref.[22], actually it is enough to calculate the lesser self-energy)

\[ \Sigma^{<}_{\text{vib}}(\varepsilon) = \sum_{\alpha} \frac{i}{2\pi} \int M^{\alpha} G^{<}(\varepsilon - \omega) M^{\alpha} D_{0,\alpha}(\omega) d\omega, \]

(10)

where \( D_{0,\alpha} \) is the free-vibron Green function.

To study the decoherence effects at low voltages (linear conductance) we use the elastic dephasing model\textsuperscript{[23,25]} which will be referred to as Vibronic Dephasing (VD).

Within this model it is assumed that the electron-vibron interaction is localized at atomic sites \( \alpha = i \) and is identical at all sites. Moreover, the energy of vibronic quanta \( \hbar \omega_{\alpha} \) is assumed to be small compared to the other energy scales in the system, so that no vibrons are excited inelastically. This approximation works well for low-frequency acoustic type vibrations.

In the elastic approximation one can integrate \[ \int \text{d} \varepsilon \] over \( \omega \) assuming \( G^{<}(\varepsilon - \omega) \approx G^{<}(\varepsilon) \). The retarded and lesser self-energies are then represented by:

\[ \left[ \Sigma_{\text{VD}}^{R(<)}(\varepsilon) \right]_{ij} = \gamma^2 G^{R(<)}_{ii}(\varepsilon) \delta_{ij}, \]

(11)

where \( \gamma \) is the strength of dephasing. It can be estimated as\textsuperscript{[23,25]}

\[ \gamma \approx \sqrt{k_B TV_{e-v}}, \]

(12)

where \( V_{e-v} \) describes the electron-vibron-coupling and \( T \) is the temperature.

Since the self-energy \( \Sigma_{\text{VD}}^{R(<)}(\varepsilon) \) depends explicitly on \( G^{R(<)}(\varepsilon) \) we have to solve this problem self-consistently.
B. Büttiker probe (BP) model

Another way to include dephasing in charge transport calculations is the so called Büttiker Probe (BP) model\cite{20,13}. As depicted in Fig. 1, in this model virtual probes are connected to every site of the chain. This leads to an additional self-energy (in the simplest case site-independent)

\[ \Sigma_{\text{BP}}^{R} = -i \frac{\gamma_{\text{BP}}}{2}, \]

where $\gamma_{\text{BP}}$ is the coupling strength of the probes to the sites. Electrons may tunnel to the probes and back instead of travelling from left to right lead directly. To describe model dephasing, in contrast to the real conducting environment, the probes have to fulfill a zero net current condition i.e. the number of electrons entering the probe has to be the same as the number of electrons leaving it again to the same site. The effective transmission between left and right leads is then given by\cite{28}

\[ T_{\text{eff}} = T_{LR} + T_{\text{corr}} \]

where $T_{LR}$ is the coherent transmission from left to right lead.

The second term includes the transmission corrections due to the probes

\[ T_{\text{corr}} = \sum_{m,n} T_{mn} T_{\text{corr}}^{m,n} \]

where $W$ describes the Markov matrix:

\[ W_{mn} = (1 - R_m) \delta_{mn} - (1 - \delta_{mn}) T_{mn}. \]

The reflection function is defined as:

\[ R_m = 1 - \sum_{n \neq m} T_{mn}, \]

where $T_{ij}$ describes all transmissions between probes $i,j = m,n$ and two leads $i,j = L,R$:

\[ T_{ij} = \text{Tr} \left( \Gamma_i \mathbf{G}^R \Gamma_j \mathbf{G}^A \right). \]

This approximation is within the Landauer-Büttiker picture and we can use the coherent expression\cite{18} for transmissions.

This model introduces decoherence effects due to re-emission of electrons with random phase. This means that the coupling strength $\gamma_{\text{BP}}$ of the probes controls the dephasing strength. The transmission correction in eq.\cite{15} is only valid for small voltages and small temperatures. Otherwise a numerical approach to adjust the chemical potential $\mu_n$ of the probes should be used to ensure zero net current\cite{37,40,44}.

Although the vibronic dephasing model and the Büttiker probe model have different origins of dephasing, both describe elastic scattering caused by local interactions.

III. THE MODEL OF DNA

Our aim is to analyze the interplay between coherent and incoherent charge transport in DNA. Therefore, we will investigate stacked ACnGnT and alternating A(CG)nT sequences following Xiang et al.\cite{19} where A,C,G,T stand for the nucleobases adenine, cytosine, guanine and thymine respectively. These structures are double-stranded and self-complementary.

We follow the idea of previous authors\cite{10,44,15} and model these structures by using a next neighbor tight-binding (TB) ladder models as depicted in Fig. 2 (gray lines). Within this approach we neglect any geometrical effects and assume that they are included in the electronic parameters. On top of that we consider low-energy charge transport so that just the frontier orbitals of the bases are relevant. This means that we take one orbital per base into account, i.e., we have $N = 4n + 4$ electronic states per molecule with $n$ base pairs.

The corresponding Hamiltonian reads:

\[ \hat{H} = \sum_{i=1}^{n} \left( \sum_{s=1,2} \varepsilon_{i,s} \hat{c}_{i,s}^\dagger \hat{c}_{i,s} + \sum_{s \neq s' = 1,2} t_{i,s,s'} \hat{c}_{i,s}^\dagger \hat{c}_{i,s'} \right) + \sum_{s,s' = 1,2} t_{i,i+1,s,s'} (\hat{c}_{i,s}^\dagger \hat{c}_{i+1,s'} + \text{h.c.}) \]

where $\varepsilon_{i,s}$ describes the onsite energy of site $i$ in strand $s$, $t_{i,s,s'}$ the interstrand hopping within a base pair and $t_{i,i+1,s,s'}$ charge transfer between the base pairs (interstrand as well as intrastrand).

Furthermore we adopt the beforehand mentioned assumption that charge transport occurs only through the purine bases (T, G).

We compare the ladder models with the corresponding 1D models which just include these bases. For a visualisation see the black lines in Fig. 2.

We use the electronic parameters which were calculated on a DFT level by Senthilkumar et al.\cite{19} and modify the data set to reproduce experimental results from
Xiang et al. A short explanation of this adjustment is given in the appendix. While most of the charge transfer integrals can be used directly we have to pay some attention to the onsite energies. In the study these values were estimated depending on the two neighboring bases and show, e.g. for guanine, a maximal variation of $\Delta \varepsilon_G \approx 0.5 \text{ eV}$. Since the outer bases, i.e., A and T are not sandwiched between two bases, we follow the idea of previous studies and average the onsite energies. The numerical values of the parameter set can be found in the appendix.

We investigate a single double-stranded DNA sandwiched between two leads. The influence of the leads will be modelled in the wideband limit, i.e. we neglect any energy dependence of the coupling. Furthermore we assume that contacts are just coupled to the first and last base (thymine) of the DNA molecule. This assumption is in fair agreement with the experiment of Xiang et al.

The hybridization functions in Eq. (4) then reduces to

$$[\Gamma_L]_{1,1} = \gamma_L,$$

$$[\Gamma_R]_{N,N} = \gamma_R.$$

Therefore the coupling to the leads is controlled by the adjustable parameters $\gamma_{L/R}$.

With these parameters we can calculate the transport properties for the ladder as well as for the 1D models.

IV. RESULTS

A. Computational Details

Our calculations were performed using DFTB$^+$XT package based on the DFTB$^+$ source code. The calculation of the vibrational self-energies is done self-consistently.

The coupling to the electrodes is estimated to be $\gamma_{L/R} = 0.30 \text{ eV}$. This value is typical for the couplings of molecular orbitals of small organic molecules to gold surfaces.

The vibronic dephasing parameter we estimate as $\gamma_{VD} = \sqrt{0.03 \text{ eV}^2/K \cdot T}$ from the formula (12). The voltage is taken to be 0.002 V to be in the linear regime. The Fermi energy of the leads are aligned with the HOMO energy $\varepsilon_G$, which is the standard approximation explained by the level alignment.

B. Comparison of Ladder and 1D DNA models

In order to get insight into the charge transport characteristics of the two different TB models we calculate the current spectral density which is the integrand of Eq. (4). The results for the stacked and alternating DNA molecules with 8 base pairs with dephasing are shown in...
FIG. 3. Current spectral densities of the DNA ladder models (solid lines) and the corresponding 1D models (dashed lines) for \( n = 8 \) at room temperature (\( \gamma_{VD} = 0.03 \text{ eV} \))

The completely coherent current spectral densities are equivalent to the transmission functions from Landauer-Büttiker theory. The current spectral densities in Fig. 3 of the stacked ladder and the corresponding 1D model show good agreement, i.e., charge transport mainly occurs through the purine bases. Contrary the alternating models show a large discrepancy. The current spectral density of the ladder model is shifted to lower energies and suppressed by a factor of \( \approx 2 \) corresponding to the density of the 1D model. This large difference is caused by the strong intrstrand coupling \( t_{\nu' - GC - \nu'} = t_{\nu' - CG - \nu'} \) in the alternating model which is neglected in the 1D model. This has a stronger influence on the alternating than on the stacked DNA because it appears just once in the latter system. Nevertheless the current spectral density of the 1D alternating DNA model shows the same features as the ladder model, i.e. it can be used to investigate qualitative transport properties. It should be emphasized that the 1D alternating model will lead to a higher current and thus a lower resistance than the stacked model. Therefore we will investigate dephasing effects utilizing the ladder model to get comparable results.

C. Dephasing in the Ladder models

We apply the vibronic dephasing model and the ladder models described before to investigate the effect of dephasing. Therefore we calculate the current spectral densities for both DNA molecules for \( n = 3, \ldots, 8 \) base pairs at room temperature. The results for the coherent and incoherent cases are shown in Fig. 4. The two DNA molecules show quite narrow transmission windows. For the coherent calculations (upper panel in Fig. 4) we find gaps at the Fermi energy of the leads \( E_F = \epsilon_G \), which is in agreement with experimental results from Shapir et al. [53]. The transmission functions of the 6 alternating DNA molecules in Fig. 4a are grouped in two discrete transmission windows above and below the Fermi energy of the leads. The stacked molecules (Fig. 4b) show broad distributed transmission peaks. Furthermore the molecules with an odd number of base pairs show transmission peaks very near \( E_F \) while in the alternating molecules there are no features near \( E_F \).

Including decoherence effects leads to broadening of the current spectral densities of both DNA molecules. Moreover it can be seen that this broadening leads to the disappearance of the transmission gap at \( E_G \) for alternating DNA (Fig. 4b) and the appearance of a local maximum slightly below \( E_F \). Additionally it can be seen that the current spectral densities of all alternating molecules have the same shape where the maxima decrease monotonously with the system length.

The incoherent current spectral densities for the stacked DNA molecules (Fig. 4c) show two different shapes. For the molecules with an odd number of base pairs the transmission gap is closed in contrast to the molecules with even number of base pairs. This gap decreases with larger system sizes. It is also noticeable that all stacked curves shows additional gaps apart \( E_F \). For example for \( n = 7 \) (orange lines in Fig. 4b) we can find transmission gaps at \(-0.097\text{ eV}, -0.039\text{ eV}, 0.028\text{ eV} \) and \( 0.086\text{ eV} \) relative to the Fermi energy.

To compare the vibronic dephasing model to the Büttiker probe model we calculated the current spectral densities for a stacked DNA molecule with \( n = 3 \) base pairs (see Fig. 5). We have chosen a dephasing strength \( \gamma_{VD} = \gamma_{BP} = 0.01\text{ eV} \) which is smaller than in Fig. 4 to emphasize the difference between the dephasing models. The peak positions are identical for the three models but the shape differs. While the peaks of the vibronic dephasing model (grey lines in Fig. 5) are rather sharp the BP models show a more smeared shape. Furthermore the minima are not that pronounced in the BP models as in the VD model leading to the disappearance of the gap at Fermi energy.

Additionally the BP model without zero net current condition (black lines) shows a much smaller current spectral density caused by leaking currents through the probes. This model may be important and corresponds to the other physical case, namely the conducting environment.

D. Length-dependent resistance

With the previously discussed current spectral densities we can calculate the current through the molecules according to Eq. 1 and thus the resistance. We used a vibronic dephasing strength of \( \gamma_{VD} = 0.03\text{ eV} \) as in Fig. 4 and estimated \( \gamma_{BP} = 0.09\text{ eV} \) to reproduce a similar behavior in the length dependence. The resistance at room temperature for the VD model (solid lines) as well as for the BP model with zero net current condition (dashed
FIG. 4. Current spectral densities calculated using the ladder models at room temperature \((T = 295 \text{ K})\). Left panel: Alternating \(A(CG)_nT\) DNA for a) coherent case and b) case with dephasing. Right panel: Stacked \(AC_nG_nT\) c) coherent case and d) case with dephasing. Dashed lines indicated the position of the Fermi energy of the leads \(E_F = \varepsilon_G\).

The resistance of the stacked DNA molecule shows overall a slight slope and additionally an even-odd-oscillation. This oscillations are slightly more pronounced by the BP model than by VD model. It is well known that these oscillations are caused by a partial delocalization of charge carriers in the stacked G-sequences. This can also be seen in Fig. 4d: The current spectral densities of molecules with odd numbers show a maximum at \(E_F\) and thus a minimum in resistance.

FIG. 5. Current spectral densities calculated using the stacked ladder model for \(n = 3\) at room temperature \((T = 295 \text{ K})\) for vibronic dephasing model (grey) and Büttiker Probe model with (red) and without (black) zero current condition. For better visualization the Büttiker Probe model without zero current condition was enhanced.

FIG. 6. Length dependent resistance for alternating and stacked DNA, calculated with ladder model for vibronic dephasing model (solid lines) as well as Büttiker probe model (dashed lines).
In comparison the resistance of the alternating molecule just shows a linear length dependence. The VD model has a slope of 0.584 MΩ per base pair similar to the experiments\(^\text{[40]}\) (≈ 0.596 MΩ per base pair). In the BP model the slope is slightly steeper with 0.684 MΩ per base pair. This increase can also be understood with the current spectral density in Fig. 4b): In the incoherent case a length dependent decrease of the maxima can be seen. This leads to a lowered current and thus to a higher resistance. Length-dependent resistance measurements of Xiang et al.\(^\text{[39]}\) show also a linear increase for alternating and stacked DNA and oscillations which superimpose the linear increase in the stacked molecule. However these measurements show a similar slope of the \(R(N)\) curves for stacked and alternating DNA sequences which we cannot reproduce with our model calculations.

In a previous study of Kim et al.\(^\text{[10]}\) where decoherence effects caused by environmental effects were investigated using the Büttiker Probe model, a similar difference between alternating and stacked DNA molecules as in our model calculations was found. Note, that in Ref.\(^\text{[10]}\) the probes were considered at finite temperature, and the zero-current conditions for probes with a certain broadening for the derivative of the Fermi function were used.

E. Temperature-dependent resistance

To get a more detailed view on the influence of dephasing we calculate the resistance of single DNA molecules dependent on the dephasing strength. The result for the stacked DNA molecule with \(n = 8\) base pairs is shown in Fig. 7.

In this calculation the dephasing strength was varied proportional to the square root of the temperature according to the formula \((12)\). Analyzing this figure, two distinct temperature dependencies can be found. At low temperatures the resistance shows a strong variation, which is the result of level broadening in the case when the resonant transition window is shifted slightly from the Fermi energy, as is shown in Fig. 4c.

On the other hand for higher temperatures the resistance seems to be constant but if we zoom in we see a slight increase (see inset in Fig. 7). Moreover, the resistance is linear dependent on \(\gamma_{VD}\), thus linear in temperature. Such behavior is consistent with “metallic” ohmic behavior of the resistance as a function of length. The crossover between the low temperature and high temperature regimes can be understood with the spectral current densities (see Fig. 4). As mentioned before decoherence effects cause broadening as well as decrease of spectral current density. For low voltages the current is obtained by the integration of spectral current density in the small voltage interval. In the low dephasing regime the broadening of spectral current density dominates resulting in an increasing current and thus decreasing resistance. On the contrary, the metallic-like regime is mainly affected by the decrease of spectral current density since the broadening appears outside the integration window and therefore we obtain a slightly increasing resistance. Similar results were obtained also for odd number of base pairs as well as for the alternating DNA molecule.

For alternating case there is considerable difference between ladder and 1D models, which we saw already in Fig. 3. From the temperature dependence (Fig. 4) it follows that this difference is larger at larger dephasing. The reason may be that the effects of interference are more essential for the ladder model.

V. CONCLUSION

We have shown that decoherence effects decrease and broaden the spectral current density. This broadening is slightly different in the investigated dephasing models. For alternating DNA molecules this closes the transmis-
sion gaps at Fermi energy occurring in the completely coherent case, leading to a conducting behavior and causes a monotonous length dependent resistance. On the contrary, dephasing in stacked DNA structures just closes the transmission gaps for molecules with odd number of base pairs and therefore molecules with even number of base pairs show a lower conductance. This distinct transport mechanism were measured by Xiang et al.\textsuperscript{[32]} Furthermore we have shown that vibronic dephasing leads to crossover from coherent tunneling regime at low temperature to a metallic-like behavior for higher temperatures. To understand better this crossover and low temperature transport in DNA, the conductance measurements at low temperature would be very desired. We also suggest to investigate the interplay between vibronic and conducting environment dephasing.

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Appendix A: Effective 1D models for DNA

In section 3 we presented the DNA ladder and the corresponding 1D models, which were compared in section 4. The 1D alternating DNA molecule can be described by:

\[
\hat{H}^A = \sum_{i=2}^{2n+1} \varepsilon_G \hat{c}_i^\dagger \hat{c}_i + \sum_{i=1,2n+2} \varepsilon_T \hat{c}_i^\dagger \hat{c}_i + \\
\sum_{i=1}^{n} t_{3'G-G-3'} \left( \hat{c}_{i+1}^\dagger \hat{c}_{i+2} + \hat{c}_{i+2}^\dagger \hat{c}_{i+1} \right) + \\
\sum_{i=1}^{n-1} t_{5'G-G-5'} \left( \hat{c}_{i+1}^\dagger \hat{c}_{i+2} + \hat{c}_{i+2}^\dagger \hat{c}_{i+1} \right) \\
\sum_{i=1,2n+1} t_{3'G-TG-5'} \left( \hat{c}_{i+1}^\dagger \hat{c}_{i+2} + \hat{c}_{i+2}^\dagger \hat{c}_{i+1} \right), \tag{A1}
\]

and the stacked DNA reads:

\[
\hat{H}^S = \sum_{i=2}^{2n+1} \varepsilon_G \hat{c}_i^\dagger \hat{c}_i + \sum_{i=1,2n+2} \varepsilon_T \hat{c}_i^\dagger \hat{c}_i + \\
\sum_{i=2}^{n} t_{3'G-G-3'} \left( \hat{c}_{i+1}^\dagger \hat{c}_{i+2} + \hat{c}_{i+2}^\dagger \hat{c}_{i+1} \right) + \\
\sum_{i=n+2}^{2n} t_{3'G-G-3'} \left( \hat{c}_{i+1}^\dagger \hat{c}_{i+2} + \hat{c}_{i+2}^\dagger \hat{c}_{i+1} \right) + \\
\sum_{i=1,2n+1} t_{3'G-TG-5'} \left( \hat{c}_{i+1}^\dagger \hat{c}_{i+2} + \hat{c}_{i+2}^\dagger \hat{c}_{i+1} \right) + \\
t_{3'G-G-3'} \left( \hat{c}_{n+1}^\dagger \hat{c}_{n+2} + \hat{c}_{n+2}^\dagger \hat{c}_{n+1} \right). \tag{A2}
\]

Appendix B: Charge transfer integrals and onsite energies

For the sake of completeness we show the used charge transfer integrals as well as the averaged onsite energies of Senthilkumar et al.\(^{15}\)

| X | Y | Z | T |
|---|---|---|---|
| G | A | C | T |
| G | 0.053 | -0.077 | -0.114 | 0.141 |
| A | -0.010 | -0.004 | 0.042 | -0.063 |
| C | 0.009 | -0.002 | 0.022 | -0.055 |
| T | 0.018 | -0.031 | -0.028 | 0.072 |

Appendix C: Modification of CT integrals

As mentioned before we adjusted the DFT charge transfer integrals from Senthilkumar et al.\(^{15}\) to reproduce experimental results obtained by Xiang et al.\(^{16}\) For low temperature calculations the original parameter set reproduces the measured behavior quite well. For room temperature the calculations do not show any significant oscillations, which were found in the experiments, for the stacked DNA molecule. It is well known, that the oscillations are caused by partially delocalization of charge carriers within the stacked G-sequence.\(^{17}\) That is why they can be controlled by \(t_{3'G-G-5'}\), i.e. the intrastrand charge transfer integral between neighboring G bases. For our calculations we increased \(t_{3'G-G-5'}\) from 0.053 eV to 0.080 eV. This can be possibly explained by the fact that DFT calculations were made at zero temperature ground state and the effects of vibrations (geometry changes) at high temperatures are not taken into account.