Calculation of the hole mobilities of the three homopolynucleotides, poly(guanilic acid), poly(adenilic acid), and polythymidine in the presence of water and Na^+ ions

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Recent high resolution x-ray diffraction experiments have determined the structure of nucleosomes. In it 147 base pair long DNA B superhelix is wrapped around the eight nucleohistone proteins. They have found that there are many hydrogen-bonds (H-bonds) between the negative sites phosphate (PO_4^-) groups DNA, and first of all there is the positively charged lysine and arginine side chains of the histones. This means that there is a non-negligible charge transfer from DNA to the proteins causing a hole current in DNA and an electronic one in the proteins. If the relative positions of the two macromolecules change due to some external disturbances, the DNA moves away from the protein and can be read. If this happens simultaneously at several nucleosomes and at many places in chromatin (built up from the nucleosomes), undesired genetic information becomes readable. This final end can cause the occurrence of oncoproteins at an undesired time point which most probably disturbs the self-regulation of a differentiated cell. The connection of these chain of events with the initiation of cancer is obvious. To look into the details of these events we have used the detailed band structures of the four homopolynucleotides in the presence of water and natrium (Na^+) ions calculated previously with the help of the *ab initio* Hartree-Fock crystal orbital method. We have found that in the case of three homopolynucleotides the width of their valence band is broad enough ($\sim$10 times broader than the thermal energy at 300 K) for the application of the simple deformation potential approximation for transport calculations. With the help of this we have determined the hole mobilities at 300 K and 180 K of poly(guanilic acid), poly(adenilic acid), and polythimidine (polycytidine has a too narrow valence band for the application of the deformation potential method). The obtained mobilities are large enough to allow Bloch-type conduction in these systems. At the end of the paper we discuss briefly the possible mechanism of charge transport in aperiodic DNA as a combination of Bloch-type conduction, hopping, and tunneling.

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I. INTRODUCTION

In two recent papers [1,2] we have calculated the electronic band structures of the four homopolynucleotides: Poly(guanilic acid) [1], polycytidine [2], poly(adenilic acid) [2], and polythimidine [2] in the absence and presence of water and Na^+ ions. For the unit cells of the homopolynucleotides (with the exception of polycytidine) see Figs. 1–3. In these calculations we have used the *ab initio* Hartree-Fock (HF) crystal orbital (CO) method with arbitrary number of basis functions in the unit cell [3–6]. The method does not work only for simple translation, but in the case of a periodic one-dimensional (1D) chain it can be easily generalized also for a combined symmetry operation (like helix operation).

According to the results obtained, with the exception of polycytidine, the other three homopolynucleotides also in the presence of water have valence bands with widths over 0.20 eV [1,2] (the valence band width of polycytidine is only 0.05 eV [2]).

Since in the three mentioned cases there is a possibility of Bloch-type hole conduction (the band width is about one order of magnitude or more larger than the thermal energy at $T=300$ K, $k_BT=0.025$ eV [7]) if they are doped by electron acceptors, we could use the deformation potential approximation.

As a pilot calculation we have performed also a HF +MP2 (Møller-Plesset)2 calculation for the nucleotide base stacks [2] (for the details of the calculations see Methods in [2]). According to the results obtained the correlation effects generally decrease somewhat the valence band widths, but in the case of the cytosine (C) stack it nearly doubles its HF valence band width from 0.06 eV to 0.11 eV [2].

For the three homopolynucleotides with large enough valence band widths we present here the mobility calculations. For this the mentioned deformation potential [8] approximation based on the band structure was used.

In two earlier papers we have computed the mobilities in this approximation in a guanine stack taking into account the compression and dilatation [9] and the torsional motion of

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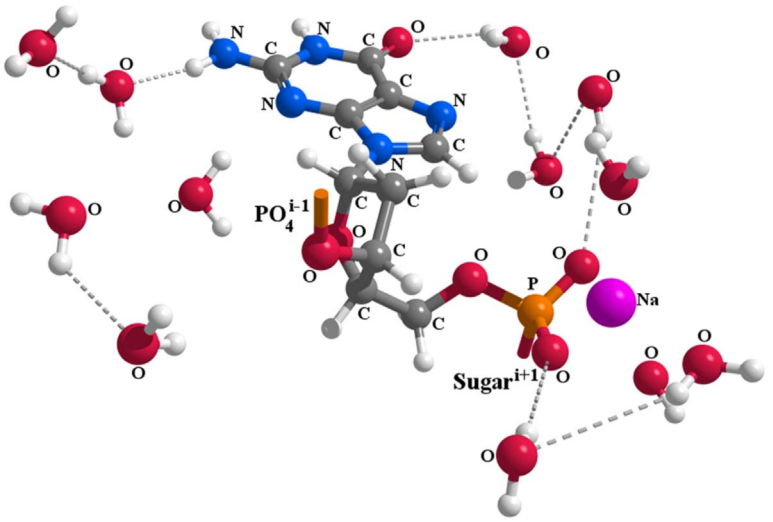

FIG. 1. (Color online) The unit cell of poly(guanilic acid) with water molecules and one Na^+.

the helix in poly(guanilic acid) [10]. To be able to perform these calculations one had to rederive from the available three-dimensional (3D) mobility expression [7] the one which is valid in the 1D case [9].

One should point out that recently the crystal structure of the nucleosomes [11,12] (they build up the chromatin system from which at cell duplication the chromosomes are formed) was determined by high resolution x-ray diffraction (2.8 Å resolution for the nucleohistone proteins and 1.9 Å for DNA [12], respectively). The structure consist of eight nucleohistone proteins which are wrapped around by a left-handed DNA B superhelix of 147 base pairs. The structure is held together first of all by the charge transfer (CT) through the H bonds of the positively charged side chains (lysine and arginine) of the histones and the PO_4^- groups of DNA. For the geometrical structure of a PO_4^- group see Fig. 4 and for the bond distances, valence angles, and torsional angles in it see Table I.

Subsequent investigations [13] have shown that there are 120 possibilities to H-bond formation between PO_4^- groups and the positive side chain of the histones. This makes it possible to estimate the positive charge per base on DNA and the negative charge on the histone side chains. In this way one would expect a positive hole current (due to local electric fields) in DNA and a negative one in the proteins.

In Refs. [11,12] it is described also that if something (binding of foreign molecules like carcinogens or radiations) change the relative conformations of the nucleohistones and DNA, the DNA molecule can unwrap. Since in that case the two molecules become further away from each other, the CT between them (and with it their mutual attraction) diminishes and with it the hole and electron currents can become interrupted.

If this happens simultaneously at many places and most probably the unwrapping of DNA from the histones spreads to a number of nucleosomes, this means that at a certain unexpected time point many genes which take part in the cell regulation become readable. This leads through the biochemically well-known mechanisms (transcription to mRNA and translation to proteins) to the occurrence of a number of so-called oncoproteins in an unexpected time point and locations. It is easy to imagine that all this disturbs the self-regulation of a cell. Most probably this contributes to the onset of cancer [14,15].

The above-described chain of events is the reason that we have started these large scale quantum mechanical calculations on DNA, calculated the charge transfer (CT) between the PO_4^- groups and the lysine and arginine side chains [16], and started to calculate the dc conductivity in proteins [17].

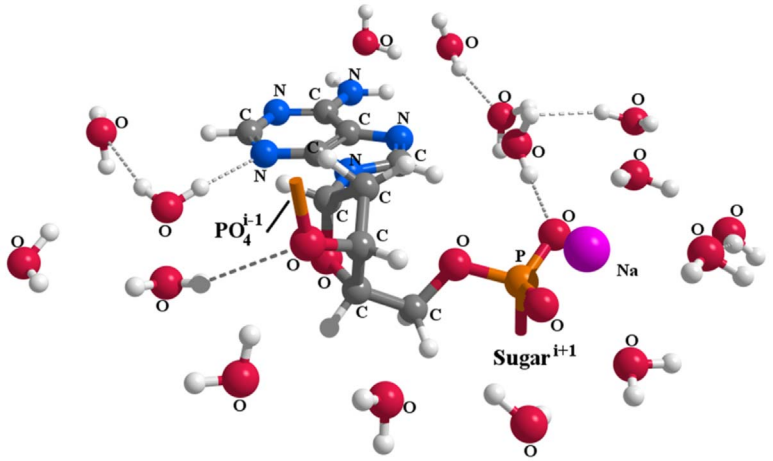

FIG. 2. (Color online) The unit cell of poly(adenilic acid) with water molecules and one Na^+.

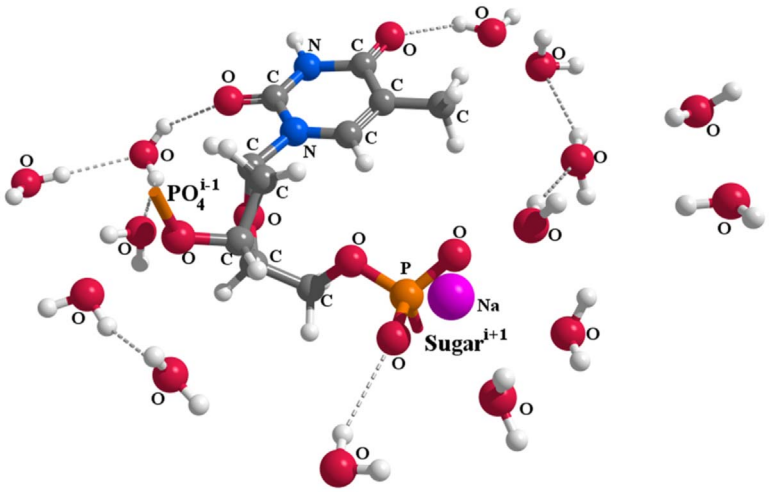

FIG. 3. (Color online) The unit cell of polythymidine with water molecules and one Na^+.

II. METHODS

For the calculation of the hole mobilities the newly derived expression for the hole mobility of 1D systems (as mentioned before [9]) has been applied

$$\mu_h = \sqrt{\frac{2}{\pi}} \frac{c_\perp \hbar^2 e}{\varepsilon_{1h}^2 m_h^{*3/2} (k_B T)^{1/2}}. \tag{1}$$

Here $c_\perp$ is the elastic constant for the motion of holes perpendicular to the base stacks, ε_{1h} is the deformation potential at the upper edge of the valence bands, and m_h^* is the effective mass of the holes. Comparing this expression with the usual one for 3D systems [7]

$$\mu_h = \frac{2^{2/3}\pi^{1/2}}{3} \frac{c_\perp \hbar^4 e}{\varepsilon_{1h}^2 m_h^{*5/2} (k_B T)^{3/2}}, \tag{2}$$

one finds that m_h^* in the denominator occurs on the 3/2th and not on the 5/2th power, $k_B T$ also in the denominator on the 1/2th and not on the 3/2th power, and instead of $\hbar^4$ in the numerator in the 1D case $\hbar^2$ occurs. Besides there is a difference in the numerical factors.

The deformation potentials at the upper edge of the valence bands were calculated with the aid of the expressions

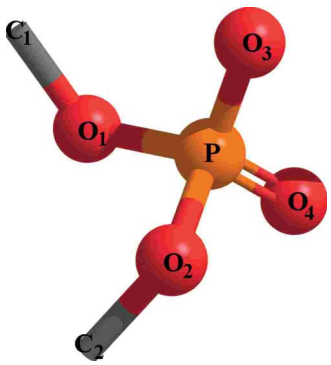

FIG. 4. (Color online) The structural formula of the PO_4^- group. The bond distances, valence angles, and torsional angles are given in Table I.

$$\varepsilon_{1h\perp} = \frac{\delta\varepsilon_{v,u,l}}{\frac{\Delta l}{l_0}} \quad (l_0 = 3.32\ \text{Å}), \quad \varepsilon_{1h\varphi} = \frac{\delta\varepsilon_{v,u,\varphi}}{\frac{\Delta\varphi}{\varphi_0}} \quad \left(\varphi_0 = \frac{2\pi}{10}\right), \tag{3}$$

for the electron-acoustic phonon scattering corresponding to the contraction and dilatation of the base stack and to the torsion of the helix formed by the nucleotides, respectively. For the equilibrium geometry we have used the one of Olson *et al.* [18], who obtained for the stacking distance $l_0 = 3.32$ Å and for the torsional angle $36° = \frac{2\pi}{10}$ by averaging the results of several x-ray diffraction investigations for double stranded DNA in absence of water. In the case of the contraction and dilatation as well as for the torsion, the ge-

TABLE I. The internal coordinates (bonds, valence angles, and torsion angles) of the PO_4^- group.

Coordinate	Value	Unit
$r(C_1\text{-}O_1)$	1.42	Å
$r(C_2\text{-}O_2)$	1.44	Å
$r(O_1\text{-}P)$	1.60	Å
$r(O_2\text{-}P)$	1.60	Å
$r(O_3\text{-}P)$	1.48	Å
$r(O_4\text{-}P)$	1.48	Å
$\alpha(C_1\text{-}O_1\text{-}P)$	119.04	deg
$\alpha(C_2\text{-}O_2\text{-}P)$	119.01	deg
$\alpha(O_1\text{-}P\text{-}O_2)$	101.43	deg
$\alpha(O_1\text{-}P\text{-}O_3)$	109.65	deg
$\alpha(O_1\text{-}P\text{-}O_4)$	109.67	deg
$\alpha(O_2\text{-}P\text{-}O_3)$	109.58	deg
$\alpha(O_2\text{-}P\text{-}O_4)$	109.58	deg
$\alpha(O_3\text{-}P\text{-}O_4)$	115.95	deg
$\tau(C_1\text{-}O_1\text{-}P\text{-}O_3)$	−16.83	deg
$\tau(C_1\text{-}O_1\text{-}P\text{-}O_4)$	−145.23	deg
$\tau(C_2\text{-}O_2\text{-}P\text{-}O_3)$	155.04	deg
$\tau(C_2\text{-}O_2\text{-}P\text{-}O_4)$	−76.68	deg

ometry of the PO_4^- groups were repeatedly optimized using the GAUSSIAN 03 program [19]. Taking this geometry [18] as starting point we have performed band-structure calculations also at the stacking distances 3.30 Å and 3.34 Å, respectively, keeping the torsional angle in all cases with $\phi_0 = 36° = \frac{2\pi}{10}$ constant. In a second series of calculations we have kept the stacking distance at $l_0 = 3.32$ Å constant and have changed the torsional angle from $\phi_0 = 36° = 0.2\pi$ to $34° = 0.181\pi$ and $38° = 0.211\pi$, respectively. This means that we had to perform, for all three homopolynucleotides, five calculations.

To obtain the band structures of the three aforementioned homopolynucleotides in the presence of water and Na^+ ions we have applied the *ab initio* Hartree-Fock (HF) crystal orbital (CO) method in its linear combination of atomic orbitals (LCAO) form. For the description of this method for arbitrary number of basis functions in the unit cell and for a general symmetry operation on a 1D periodic chain see Refs. [3–6]. In the case of a helix operation one must apply besides the translation along the helical axis also a rotation of the positions of the nuclei and all basis functions which have a component perpendicular to the helix axis (this generalization of the CO method in the 1D case is therefore possible, because the symmetry group of the simple translation is in the 1D periodic case isomorphic with the group of the combined symmetry operation). For a concise, but still detailed description of the theory see [20].

To determine the water structure around a nucleotide we have applied a multistep procedure which is described in detail in Refs. [1,2]. We have found in this way for poly(guanilic acid) 13 water molecules and one Na^+ ion, for poly(adenilic acid) 16 water molecules and one Na^+ ion, and for polythymidine 14 H_2O molecules and one Na^+ ion in the unit cell. The Na^+ ions are located between those two oxygen (O) atoms of the PO_4^- group which do not take part in H bonds in the plane determined by the P atom and the mentioned two O atoms [1]. In the actual calculation the electronic structure of the H_2O molecules and of the Na^+ ion has been explicitly taken into account [1,2].

We have used for these rather large scale calculations Clement's double-ζ basis set [21] [in this way the number of basis functions in the presence of water and Na^+ ions is 479 for poly(adenilic acid) and 443 both for polythymidine and poly(guanilic acid) per unit cell] and 16 different k points [the quasimomentum k is defined in our calculation on the combined symmetry (helix) operation and not on the simple translation] in the half Brillouin zone.

Though we have kept only two-electron Coulomb integrals larger or equal than 10^{-6} a.u., the handling of the enormous number of integrals was a large task.

It should be further mentioned that for long-range Coulomb integrals, if they were smaller than 10^{-6} a.u., the multiple expansion method of Delhalle and Piella [22,23] was applied.

In the case of the exchange integrals only those were kept which have two basis functions with their centers smaller than 30 Å apart. In large gap systems (as in our cases) the exchange integrals decay rather quickly.

After calculating the HF band structures of the three homopolynucleotides at three different stacking distances and three different torsional angles the shifts of the upper edges of their valence bands were determined (one had to take average of these shifts in both directions). From these using Eq. (3) the deformation potentials for the changes of the stacking distances and the torsional angles were determined.

The effective hole masses at the upper edge of the valence bands occurring in the mobility, Eq. (1), were computed in the usual way by fitting a sixth-order polynomial to the dispersion curves of the valence bands.

One knows the elastic constant $c_\perp$ of graphite which was applied previously for the mobility calculation of a guanine stack in the case of dilatation and contraction [9], and for poly(guanilic acid) in the presence of water and Na^+ ions. The elastic constant c_φ for the torsional motion is known experimentally of a single DNA helix [24]. On the other hand, no elastic constants $c_\perp$ and c_φ are available for the other cases. Therefore, we have determined all of them theoretically from the band structure, to be on the same footing.

The elastic constant of 1D polymer fiber can be determined as [25]

$$c_\perp = l_0 \left.\frac{\partial^2 E}{\partial l^2}\right|_{l=l_0}, \quad c_\varphi = \varphi_0 \left.\frac{\partial^2 E}{\partial \varphi^2}\right|_{\varphi=\varphi_0}. \tag{4}$$

Here E is the calculated total energy per unit cell, l and φ are the unit cell length and torsion angle, respectively, l_0 and φ_0 are their equilibrium values. The derivatives in Eq. (4) were obtained from a harmonic fitting of the calculated total energy at three different values of l, (l_0 and $l_0 \pm \Delta l$) and φ (φ_0 and $\varphi_0 \pm \Delta\varphi$).

The c values obtained in this way must be substituted into the equations corresponding to $\mu_\perp$ and μ_φ [see Eq. (1)]. To obtain the effective mobility in the case of simultaneous contraction-dilatation and torsion one must apply the expression

$$\frac{1}{\mu_{h_{\text{eff}}}} = \frac{1}{\mu_{h\perp}} + \frac{1}{\mu_{h\varphi}}. \tag{5}$$

In a recent calculation we have computed the CT from a PO_4^- group to the lysine$^+$ and arginine$^+$ side chains of the histones. We have used a triple ζ + polarization function on the non-H atoms in our HF+MP2 calculation. We have assumed that there are three H_2O molecules and a kalium K^+ ion in the unit cell. After geometry optimization [applying the basis set superposition error (BSSE) correction and using the natural bond orbital (NBO) analysis] we have obtained $0.067e$ CT in the case of a lysine and $0.050e$ CT for arginine [26]. Taking their algebraic average $0.059e$ and multiplying it by $\frac{60}{147} = 0.41$ (there are 120 H bonds between the 147 base pair long DNA B superhelix and the nucleohistones in a nucleosome [13]). Since, however we have calculated only a single helix, this means that there are only 60 H bonds. One must multiply the CT of $0.059e$ by this number to obtain the charge transfer from a DNA single helix per nucleotide base to the histones, $0.024e$.

Using Eq. (1) for the mobility we have calculated it at the temperatures $T = 300$ K and $T = 180$ K, respectively.

TABLE II. The characteristics of the valence and conduction bands together with their effective masses of poly(guanilic acid), poly(adenilic acid), and polythymidine in the presence of water and Na^+ ions. The energies are given in eV s and the effective masses in units of the free electron mass.

		Poly(guanilic acid)	Poly(adenilic acid)	Polythymidine
Conduction band	u.l.[a]	3.965	6.059	4.814
	l.l.[b]	3.656	5.943	4.666
	w[c]	0.309	0.116	0.148
Valence band	u.l.[d]	−6.536	−6.175	−6.551
	l.l.[e]	−6.889	−6.423	−6.933
	w[f]	0.353	0.248	0.382
	Gap	10.192	12.118	11.217
	m^*[g]	2.354	2.202	2.280

[a]u.l., conduction band upper limit.
[b]l.l., conduction band lower limit.
[c]w, width of conduction band.
[d]u.l., valence band upper limit.
[e]l.l., valence band lower limit.
[f]w, width of valence band.
[g]m^*, the effective mass at the upper limit of the valence band.

III. RESULTS

In Table II we show the characteristics of the valence and conduction bands of the three homopolynucleotides under consideration (the detailed band structures were given in Refs. [1,2]), together with their effective masses.

In Table III, for the same system, the deformation potentials, the elastic constants for the contraction-dilatation motion of the stacks, and for the change of the torsion are given together with the corresponding hole mobilities and the effective mobilities (the hole-mobility values were calculated at 180 and 300 K, respectively).

IV. DISCUSSION

The effective hole mobility at 300 K of 153.05, 18.01, and 20.90 $cm^2/V\,s$, respectively, obtained for poly(guanilic acid), poly(adenilic acid), and polythymidine, in the presence of water and Na^+ ions are medium large in the latter two cases, while for poly(guanilic acid) (first case) μ_{eff} is by one order of magnitude larger. This is also true for $\mu_\perp$ and μ_φ, respectively.

TABLE III. The deformation potentials (in eV s), the elastic constants ($c_\perp$ and c_φ, respectively, in dyn s), the mobility values $\mu_\perp$, μ_φ, and μ_{eff} (all in $cm^2/V\,s$ units) at 300 and 180 K, respectively, for poly(guanilic acid), poly(adenilic acid), and polythymidine in the presence of water and Na^+ ions. The mobility calculations have been performed for two temperatures, $T=300$ K and $T=180$ K, respectively.

	Poly(guanilic acid)	Poly(adenilic acid)	Polythymidine
$\varepsilon_{1h\perp}$	4.32	3.57	14.11
$\varepsilon_{1h\varphi}$	0.94	0.93	1.97
$c_\perp$	1.21×10^{-1}	7.90×10^{-3}	1.52×10^{-1}
c_φ	3.47×10^{-2}	1.49×10^{-2}	4.69×10^{-2}
$\mu_\perp^{300}$	178.57	18.77	22.23
μ_φ^{300}	1070.61	514.93	348.60
μ_{eff}^{300}	153.05	18.10	20.90
$\mu_\perp^{180}$	230.36	24.21	28.68
μ_φ^{180}	1381.09	664.26	449.69
μ_{eff}^{180}	197.43	23.36	26.96

The effective hole masses are of the same order of magnitude in all three cases. Polythymidine has one order of magnitude larger $\varepsilon_{1h\perp}$ deformation potential and about 2 times as large $\varepsilon_{1h\varphi}$ value than in the other two cases. The elastic constant $c_\perp$ is of the order 10^{-2} in the case of poly(adenilic acid) and of the order of 10^{-1} in the other two cases. On the other hand, the elastic constants of the torsional motion (c_φ) have the same order of magnitude in all three cases (10^{-2}). The large $c_\perp$ values in poly(guanilic acid) and polythymidine [by one orders of magnitude larger than in poly(adenilic acid)], because of their larger interactions, cause a larger $\mu_\perp^{300}$ value. At polythymidine, however, the deformation potential is by one order of magnitude larger than in the other two cases. This is caused by the fact that thymine has two O atoms, each having two lone pairs of electrons. Since the deformation potential occurs on the second power in the denominator of Eq. (1) it compensates the effect of $c_\perp$ [which has the same order of magnitude as in the case of poly(guanilic acid)]. The interplay of these factors causes that poly(guanilic acid) has a $\mu_\perp$ value by one order of magnitude larger than the other two homopolynucleotides.

The μ_φ values are—as in our previous calculations [10]—by one order of magnitude larger than the corresponding $\mu_\perp$ values. Also in this case μ_φ is by one order of magnitude larger for poly(guanilic acid) than in the other two cases most probably caused by the same effects described before for $\mu_\perp$. Therefore, also μ_{eff} becomes one order of magnitude larger in the case of poly(guanilic acid) than in the other two cases.

We have calculated $\mu_\perp$, μ_φ, and μ_{eff} using the same formalism also at 180 K. Since the square root of the temperature occurs in the denominator of Eq. (1), the $\mu_\perp$, μ_φ, and μ_{eff} increase correspondingly.

One could raise the question what happens with the water structure around the homopolynucleotides at 180 K. Certainly there are some changes, but since we have taken into account only the first water shell around the homopolynucleotides and H bonding of H_2O molecules plays an important role in their distribution, most probably the applied decrease of temperature will not influence very strongly the distribution of H_2O molecules.

We have stopped our calculation at the mobilities and did not try to go further to determine the specific conductivity of the homopolynucleotides. The main reason of this is that we do not know the position of Fermi level of the holes when they are generated through charge transfer from the PO_4^- groups of DNA to the positively charged lysines and arginines of the nucleohistones.

We plan further calculations to find out the details of the charge transfer including a band-structure calculation for the PO_4^- lysine or arginine model systems, repeating them infinite times. We are going to study also the effect of water molecules and K^+ ions on the band structure of this system.

Native DNA is of course aperiodic. On the other hand, an adenine-thymine base pair is rather similar to a guanine-cytosine base pair, both contain a purine-type (guanine or adenine) and a pyrimidine-type (cytosine or thymine) base and both have 24 π electrons. Finally it should be mentioned that in many cases there are several or even 15–20 repeated base pairs.

In a periodic (homo)polynucleotide with small interactions of the charge carriers (moving in the direction of the helix) with the optical phonons (vibrations in the planes of the nucleotide bases) the occurrence of energy bands and with it Bloch-type conduction is possible if the band widths are sufficiently large. This is also true if there is a small barrier in the space between the stacked bases. It is also interesting to observe that the sum of the van der Waals radii of two C atoms in two superimposed bases, 2×1.7 Å $= 3.4$ Å, is very close to the stacking distance in DNA B, 3.32 Å.

On the other hand, native DNA is aperiodic. In this case a frequency-independent hopping-tunneling model must be developed if we wish to calculate the dc conductivity. If in a given base sequence, several times (10–20 times) the same base pairs are repeated, followed by an aperiodic sequence, one must develop a charge transport theory in which coherent conduction is combined with hopping. In such a model also the possibility of hopping from one helix to the other in a DNA double helix should be taken into account.

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