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Quantum entanglement and quantum information in biological systems (DNA)

Ivan Hubač,[1,a] Miloslav Švec[2,b] and Stephen Wilson[3,c]

[1] Institute of Physics, Faculty of Philosophy & Science, Silesian University,
74601 Opava, Czechia
[2] Physics Department, Dawson College, Montreal, Quebec H3Z 1A4, Canada
[3] Physical and Theoretical Chemistry Laboratory, South Parks Road,
Oxford OX13QZ, England
[a] belaxx@gmail.com
[b] svec@hep.physics.mcgill.ca
[c] quantumsystems@gmail.com, ORCID 0000-0001-5651-5669

ABSTRACT

Recent studies of DNA show that the hydrogen bonds between given base pairs can be treated as diabatic systems with spin-orbit coupling. For solid state systems strong diabaticity and spin-orbit coupling the possibility of forming Majorana fermions has been discussed. We analyze the hydrogen bonds in the base pairs in DNA from this perspective. Our analysis is based on a quasiparticle supersymmetric transformation which couples electronic and vibrational motion and includes normal coordinates and the corresponding momenta. We define qubits formed by Majorana fermions in the hydrogen bonds and also discuss the entangled states in base pairs. Quantum information and quantum entropy are introduced. In addition to the well-known classical information connected with the DNA base pairs, we also consider quantum information and show that the classical and quantum information are closely connected.

Keywords: Quantum entanglement – quantum entropy – biological systems

1 INTRODUCTION

There is growing evidence that living systems harness some of the unique features of quantum mechanics (Lambert et al., 2012). Examples include quantum coherent energy transport in photosynthesis (Blankenship, 2014; Cogdell et al., 2008) and avian magnetoreception (Wiltschko et al., 2010).

In this paper, we find theoretical support, using the Majorana quasiparticle formalism, for the conjecture that quantum mechanics and quantum information may also have a role in the DNA molecule. In particular, we propose that Majorana fermions can be connected with the delocalized electron associated with each of the hydrogen bonds between the base pairs. These Majorana fermions can define Majorana qubits which give rise to entanglement and

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form the foundation of quantum information processes. As Lambert et al. (2012) have asked in their review of quantum biology:

> "…has nature already beaten us in leveraging quantum effects to achieve something an equivalent classical system cannot? Certainly nature can harvest energy extremely efficiently, sense weak magnetic fields and *create human minds complex enough to even be asking these questions*."

where the italics have been added by the present authors. In this work, we demonstrate a mechanism for quantum information processes in the DNA molecule. This quantum mechanism may be exploited alongside the well-known classical information processes in DNA.

In the following section, we present some of the background to this work.

2 BACKGROUND

Recent years have witnessed a growing interest in Majorana fermions (Wilczek, 2009; Franz, 2013; Leijnse and Flensberg, 2012; Elliott and Franz, 2015; Alicea, 2012) which were first proposed eight decades ago by Ettore Majorana (1937). (A review of the work of Majorana is given in the recent volume by Esposito (2014).) Majorana developed a specific representation of Dirac equation (Dirac, 1928) with solutions describing a neutral spin $\frac{1}{2}$ particle which is its own antiparticle. This particle is termed a Majorana fermion. The Majorana fermion has been extensively investigated in nuclear and particle physics as a potential fundamental particle with the neutrino being the favoured candidate. In condensed matter systems, the electrons are, of course, Dirac fermions, but Majorana fermions can arise as *emergent quasiparticles* (Wilczek, 2009; Franz, 2013; Leijnse and Flensberg, 2012; Elliott and Franz, 2015). Majorana quasiparticles were found (Mourik et al., 2012; Williams et al., 2012; Rokhinson et al., 2012; Deng et al., 2012; Das et al., 2012; Finck et al., 2013; Nadj-Perge et al., 2014; Bunkov and Gazizulin, 2015) recently although their existence was predicted theoretically by Kitaev (2001) using a one-dimensional model of a topological superconductor. Here electronic and vibrational motions are coupled together. The Majorana formalism has the potential importance in a number of areas of science, e.g. solid state physics, cosmology and quantum computation.

In this paper, we apply the Majorana quasiparticle formalism to a biological system, in particular, to the hydrogen bonds in DNA. A number of authors have considered biological systems as a source of quantum biological information, e.g. Lambert et al. (2012); Koruga (2012); dos Santos et al. (2015); Guerra et al. (1999); Abbott et al. (2008); Loewenstein (2013); Asano et al. (2015); Arndt et al. (2009); Sergi (2009); Craddock et al. (2014); Vedral (2014). Although Dirac fermions play the central role in solid state physics and practically the whole of chemistry (e.g. in the determination of the structure and properties of condensed matter and molecular systems), we suggest that Majorana fermions can play an important role in quantum biology because of their role in quantum information theory (Hassler, 2014).

Influenced by Schrödinger's book *What is Life?* (Schrödinger, 1944) and his idea of the *aperiodic crystal* as the basis of life, Watson and Crick (1953) established the double helix structure of DNA, deoxyribonucleic acid, consisting of two polynucleotide strands held together by hydrogen bonds between pairs of complementary purines (adenine (A) and guanine (G)) and pyrimidines (thymine (T) and cytosine (C)). Adenine is always hydrogen bonded to thymine and guanine to cytosine. The two strands have a backbone consisting of a repeating unit of a sugar (deoxyribose) connected to a phosphate which form hydrogen bonds to external water molecules, whereas the purine-pyrimidine base pairs are located in

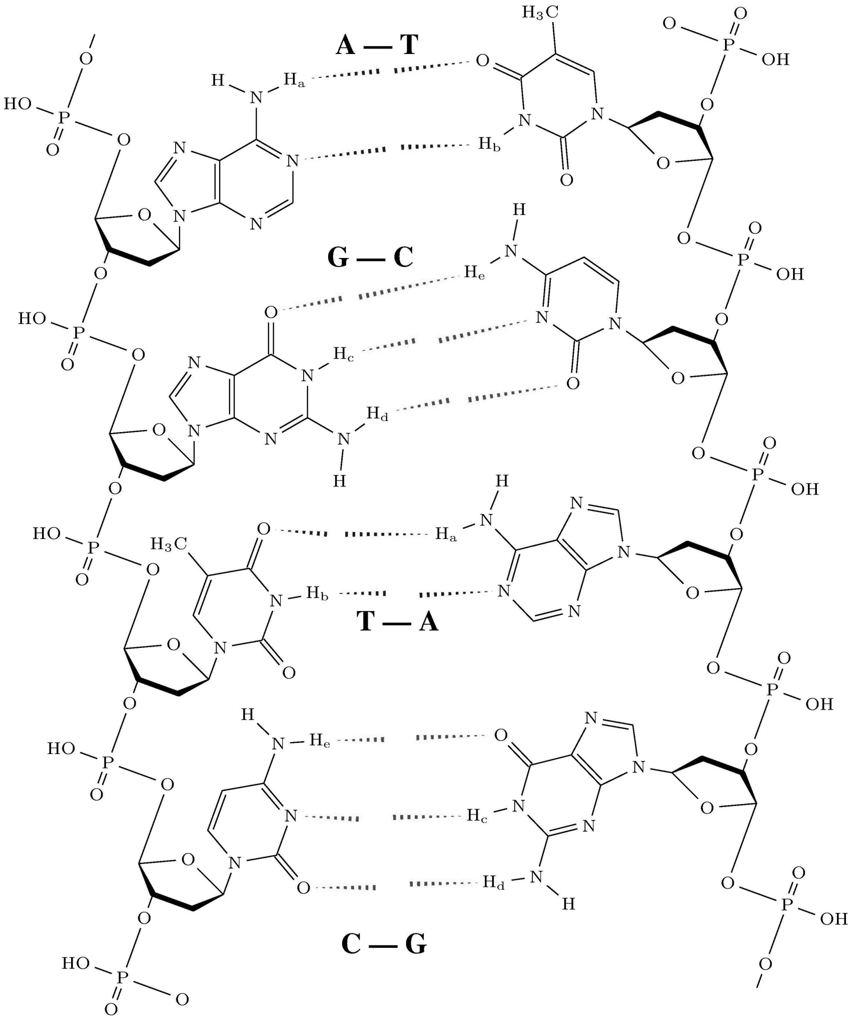

Figure 1. Structure of the DNA macromolecule with the sugar-phosphate backbone of the two helices on the left- and right-hand side and the Adenine (A)-Thymine (T) and Guanine (G)-Cytosine (C) base pairs between them. There are two hydrogen bonds (a one qubit systems) between A and T and three hydrogen bonds (a two qubit system) between G and C. The base pairs are located in the centre of the DNA molecule where they are protected from the aqueous environment.

the centre of the DNA molecule where they are protected from the aqueous environment. The structure of the DNA molecule is shown schematically in Fig. 1. DNA provides the genetic code for protein synthesis. Each triplet of nucleic acids forms a *codon*. These codons translate into amino acids. The one or more polypeptide chains, from which proteins are made, are synthesized by condensing together amino acids with the elimination of water (Crick, 1963, 1967; Watson et al., 2014). This model provides a classical approach to the DNA-protein scheme. In this model DNA is a classical information system using a ternary coding system with $4^3 = 64$ coding words (Crick, 1963). The vast majority of the ~ 3000 million base pairs of the human genome do not code for proteins. The ENCODE project (ENCODE Project Consortium, 2004, 2012; Djebali et al., 2012; Thurman et al., 2012; Neph et al., 2012; Gerstein et al., 2012; Sanyal et al., 2012; Birney et al., 2007) has shown that these stretches of what had been called 'junk' DNA contain regions that bind proteins and RNA molecules, bringing them into positions from which they regulate the function and level of expression of protein-coding genes. Furthermore, it appears that transcription from non-coding DNA has the potential to provide a reservoir for the creation of new functional molecules, such as regulatory RNAs. Parrington's recent book *The deeper genome* (Parrington, 2017) provides an account of developments in our understanding of the genome over recent years.

The hydrogen bond involves the attraction of a hydrogen atom bound to an electronegative atom, such as N, O or F, to another electronegative atom (Jeffrey, 1997; Gilli and Gilli, 2009; Desiraju and Steiner, 2001; Scheiner, 1997). Normal hydrogen bonds are primarily electrostatic in nature and generally linear. Other types of hydrogen bond are described as 'proton-shared' and 'blue-shifting' (Joseph and Jemmis, 2007; Buckingham et al., 2008; McDowell and Buckingham, 2010; Arunan et al., 2011a,b). Experimentally, hydrogen bonding can be identified by crystal structure analysis, by vibrational spectroscopy and by gas-phase rotational spectroscopy (Desiraju and Steiner, 2001). Infrared spectrum analysis, for example, can be used to identify changes in vibrational spectra attributable to hydrogen bonding. The hydrogen bond in DNA is a diabatic system (Buckingham et al., 2008) and therefore it should be treated with diabatic Hamiltonian. Computational quantum chemistry allows the displacements of nuclei, which comprise the normal modes of vibration, to be studied with an accuracy beyond that afforded by experimental data (Scheiner, 1997). The importance of hydrogen bonding in the structure and function of biological macromolecules was recognized at an early stage (Pauling et al., 1951). In DNA and RNA hydrogen bonds between complementary pairs of bases stabilize the double helix structure with guanine-cytosine and adenine-thymine pairs in DNA and thymine replaced by uracil in RNA (Watson et al., 2014). In the 1960s, Löwdin (1962, 1966) published pioneering studies of the quantum tunnelling of hydrogen in the DNA and RNA molecules which helped to established the field of 'quantum biology.' (For a recent overview of quantum tunnelling of hydrogen in DNA see (Al-Khalili, 2013).) Santos et al. have recently studied quantum confinement in the hydrogen bonding for proteins in the NH and OH groups (dos Santos et al., 2015). In very recent work (dos Santos et al., 2015), these authors consider the quantum confinement of the hydrogen in the NH group of the nucleic acids. Figure 2 shows the quantum confinement of hydrogen in the X-baseH$\cdots$Y system. Quantum entanglement between the electron clouds of nucleic acids in DNA was studied by Rieper et al.

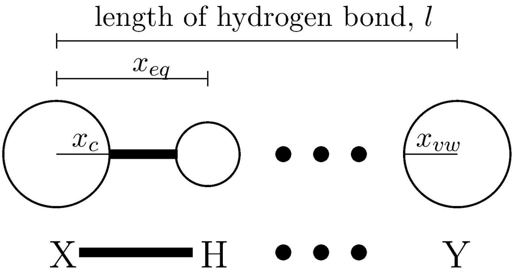

Figure 2. Schematic representation of a hydrogen bond showing the interaction between the proton donor X-H and a proton acceptor Y. x_c is the covalent radius of the donor atom, x_{vw} is the van der Waals radius of the acceptor atom, and x_{eq} is the equilibrium distance of covalent bond X-H.

(2010) using a model in which the electron clouds of the nucleic acids were viewed as a chain of coupled quantum harmonic oscillators.

In solid state theory, Majorana fermions can arise as emergent quasiparticles which can be regarded as collective excitations of the quantum many-body state describing a system of interacting electrons (Leijnse and Flensberg, 2012; Elliott and Franz, 2015). Any fermion can be written as a combination of two Majorana fermions. Essentially, a fermion can be divided into a real part and an imaginary part, each of these components being a Majorana fermion. This purely formal mathematical operation becomes interesting when the Majorana fermions are spatially well separated. In this situation, they are protected from decoherence since local perturbations cannot affect only one of the Majorana fermions. However, the state can be manipulated by physical exchange of Majorana fermions because of their non-abelian statistics. This leads to the idea of low-decoherence topological quantum computation (Nayak et al., 2008).

Here we try to identify the Majorana fermions in the hydrogen bond in DNA and to introduce qubits in connection with this hydrogen bond. Since the Majorana fermions were discussed in connection with superconductivity, where electronic and vibrational motions are coupled together, we start with a short discussion about coupling electronic and vibrational motions in molecules.

The structure of the remainder of this paper is as follows: In Section 3, we present a canonical transformation which couples the electronic and vibrational motions in molecular Hamiltonian by building on our previous work (Hubač and Wilson, 2008). Majorana transformations are then applied to this diabatic transformation in Section 4 leading to Majorana fermions in the molecule. In section 5, we introduce Majorana qubits and quantum information entropy. Quantum information in the DNA molecule is considered in section 6. Section 7 contains our conclusions and our perspective on the outlook for future work.

3 DERIVATION OF THE ADIABATIC VIBRATIONAL-ELECTRONIC HAMILTONIAN OPERATOR USING CANONICAL TRANSFORMATIONS

We have presented a detailed derivation of the adiabatic and the diabatic Hamiltonian by canonical transformations previously (Hubač and Wilson, 2008; Hubač and Svrček, 1988, 1992b,a). Here we summarize the details required for the application to the hydrogen bonds in the DNA molecule considered in this paper.

We can allow the electrons to experience the R-dependence of the nuclear terms by coupling the electronic and vibrational motions by means of a series of canonical transformations. The transformation from a basis set defined with respect to a fixed geometry R_0, $\{|p\rangle, |q\rangle, \ldots\}$, to an R-dependent basis set, $\{|p(R)\rangle, |q(R)\rangle, \ldots\}$, can be written:

$$|p\rangle \rightarrow |p(R)\rangle = \sum_q C_{qp}(R)\,|q\rangle \ .$$

This transformation employs a mixed set of second quantized operators and leads to a new representation in terms of *fermions* and *bosons* rather than *electrons* and *phonons*. The new fermion creation and annihilation operators are defined as $\{\bar{a}_p, \bar{a}_q, \ldots\}$ and $\{\bar{a}_p^\dagger, \bar{a}_q^\dagger, \ldots\}$, and new boson operators as $\{\bar{b}_r, \ldots\}$ and $\{\bar{b}_r^\dagger, \ldots\}$. The new fermion operators can be required to commute with the new boson operators. The quasi-particle transformation relating the 'crude adiabatic representation' with our new representation may then be written as follows:

$$\bar{a}_p = a_p + \sum_q \sum_{k=1}^{\infty} \frac{1}{k!} \sum_{r_1 \ldots r_k} C_{pq}^{r_1 \ldots r_k} B_{r_1} \ldots B_{r_k} a_q \,, \tag{1a}$$

$$\bar{a}_p^\dagger = a_p^\dagger + \sum_q \sum_{k=1}^{\infty} \frac{1}{k!} \sum_{r_1 \ldots r_k} C_{pq}^{r_1 \ldots r_k} B_{r_1} \ldots B_{r_k} a_q^\dagger \,, \tag{1b}$$

$$\bar{b}_r = b_r + \sum_{pq} \sum_{k=0}^{\infty} \frac{1}{k!} \sum_{s_1 \ldots s_k} D_{rPQ}^{s_1 \ldots s_k} B_{s_1} \ldots B_{s_k} a_p^\dagger a_q \,, \tag{1c}$$

$$\bar{b}_r^\dagger = b_r^\dagger + \sum_{pq} \sum_{k=0}^{\infty} \frac{1}{k!} \sum_{s_1 \ldots s_k} D_{rpq}^{s_1 \ldots s_k *} B_{s_1} \ldots B_{s_k} a_q^\dagger a_p \,. \tag{1d}$$

where B_{r_i} are the normal coordinates and C_{pq} and D_{rpq} are expansion coefficients.

Equations (1b) can be cast in the more compact forms:

$$\bar{a}_p = \sum_q \sum_{k=0}^{\infty} C_{pq}^{(k)} a_q = \sum_q C_{pq} a_q \quad \text{and} \quad \bar{a}_p^\dagger = \sum_q \sum_{k=0}^{\infty} C_{pq}^{(k)} a_q^\dagger = \sum_q C_{pq} a_q^\dagger$$

and similar formulae can be written for equations (1d) in terms of the D_{rpq} coefficients.

Operators for the fermions satisfy anticommutation relations whilst those for the bosons satisfy commutation relations. We require that operators for the fermions and bosons commute and therefore the total vibrational-electronic wave function can be written

$$\Psi(r,R) = \psi_k(r,R)\chi_k(R) \,,$$

where $\psi_k(r,R)$ is the fermion wave function and $\chi_k(R)$ is the boson wave function. It is found that there are two invariants of the transformation – the number of fermions is conserved, i.e. $\bar{N} = N$ so that we are free to redefine the Fermi vacuum, and the normal coordinate is unchanged, i.e. $\bar{B} = B$.

The canonical transformations (1) leads to an expression for the total vibrational-electronic Hamiltonian operator which can be written a sum of two parts: $\mathrm{H} = \mathrm{H}_A + \mathrm{H}_B$. Here the operator H_A can be written:

$$\begin{aligned}\mathrm{H}_A = E_{NN}(B) - E^{(2)}_{NN}(B) - V^{(2)}_N(B) + E_{\text{SCF}} + \sum_{pq} f_{pq} N\left[a^\dagger_p a_q\right] \\ + \frac{1}{2}\sum_{pqrs} v_{pqrs} N\left[a^\dagger_p a^\dagger_q a_s a_r\right],\end{aligned}$$

where the terms $E_{NN}(B)$ and $E^{(2)}_{NN}$ arise from a Taylor expansion in normal coordinates (Hubač and Svrček, 1988) and $V^{(2)}_N(B)$ is the new effective potential. The new quasi-particle Fermi vacuum allows a new quasi-particle Hartree–Fock energy to be defined as:

$$E_{\text{SCF}} = \sum_{rsi} h_{rs} C_{ri} C_{si} + \frac{1}{2}\sum_{rstkij}\left(v^0_{rtsu} - v^0_{rstu}\right) C_{ri} C_{si} C_{tj} C_{uj}\,,$$

together with a corresponding Hartree–Fock operator f with matrix elements

$$f_{pq} = \sum_{rs} h_{rs} C_{rp} C_{sq} + \sum_{rtsui}\left(v^0_{rtsu} - v^0_{rstu}\right) C_{rp} C_{sq} C_{ti} C_{ui}\,,$$

and the new two-particle integral

$$v_{pqrs} = \sum_{tuvw} v^0_{tuvw} C_{tp} C_{uq} C_{vr} C_{ws}\,.$$

The coefficients C_{pq} can be determined by solving the *coupled perturbed Hartree–Fock (CPHF)* equations (Stevens et al., 1963; Gerratt and Mills, 1968a,b; Pople et al., 1979; Takada et al., 1983; Pulay, 1977; Simons et al., 1984). Carrying out the inverse transformation of the transformations (1) and then applying Wick's theorem, we obtain the result (Hubač and Svrček, 1988):

$$\begin{aligned}\mathrm{H}_A = E^0_{NN} + E^0_{\text{SCF}} + \sum_p N\left[a^\dagger_p a_p\right] + \sum_{n=1}^{\infty}\sum_{k=0}^{[n/2]} E^{(k,n-2k)} B^{(n-2k)} \\ + \sum_{n=1}^{\infty}\sum_{k=0}^{[n/2]}\sum_{pq} f^{(k,n-2k)}_{pq} B^{(n-2k)} N\left[a^\dagger_p a_p\right] \\ + \frac{1}{2}\sum_{n=0}^{\infty}\sum_{k=0}^{[n/2]}\sum_{pqrs} v^{(k,n-2k)}_{pqsr} B^{(n-2k)} N\left[a^\dagger_p a^\dagger_q a_s a_R\right].\end{aligned}$$

The second part of the vibrational-electronic Hamiltonian, H_B, can be handled in a similar fashion to give:

$$\begin{aligned}
H_B = \sum_r \hbar\omega_r \left(b_r^\dagger b_r + \tfrac{1}{2}\right) &+ \sum_{air} \hbar\omega_r (D_{rai})^2 \\
&+ \sum_{pqr} \hbar\omega_r \left(b_r^\dagger D_{rpq} + D_{rpq} b_r\right) N\left[a_p^\dagger a_q\right] \\
&+ \sum_{pqair} \hbar\omega_r \left(D_{rpa} D_{rqa} - D_{rpi} D_{rqi}\right) N\left[a_p^\dagger a_q\right] \\
&+ \sum_{pqrsr} \hbar\omega_r D_{rps} D_{rqr} N\left[a_p^\dagger a_q^\dagger a_s a_r\right] .
\end{aligned}$$

3.1 Generalization of the canonical transformations

Up to this point in our discussion, we have based the quasi-particle transformation on the normal coordinates using the follow expressions:

$$\begin{aligned}
\bar{a}_p &= \sum_Q C_{PQ}(B) a_Q \,, & \bar{a}_p^+ &= \sum_Q C_{PQ}(B)^+ a_Q^+ \,, \\
\bar{b}_r &= b_r \sum_{PQ} D_{rPQ}(B) a_P^+ a_Q \,, & \bar{b}_r^+ &= b_r^+ \sum_{PQ} D_{rPQ}(B)^+ a_Q^+ a_Q \,,
\end{aligned} \qquad (2)$$

where we have used the normal coordinate: $B = b + b^+$.

For the non-adiabatic representation, we can generalize the transformations (2). By analogy with the normal coordinate operator B_r, we can introduce the momentum operator: $\tilde{B} = b - b^+$ and then generalize the transformations (2) so as to define new fermion operators which represent the dependence of the motion of electrons on both the coordinates and the momenta of the nuclei. Because of their finite mass, the electrons do not follow to the motion of the nuclei adiabatically. The motion of the electrons is phase-shifted with respect to the motion of the nuclei.

The case of strong coupling has been consider in the recent work by Dahnovsky (2007) on *Ab Initio electron propagators in molecules with strong electron-phonon interaction*. Dahnovsky uses the exponential transformation:

$$T = e^S \,,$$

where

$$S = \sum_{i,g} c_i^+ c_i \frac{N_g^i}{W_g} (b^+ - b) \quad \text{and} \quad \bar{C}_i = e^S c_i e^{-S} \,.$$

In the present approach, the general canonical transformation from the old set of second quantized operators $\{a_P\}$, $\{a_P^+\}$, $\{b_r\}$ and $\{b_r^+\}$ to the new set of operators $\{\bar{a}_P\}$, $\{\bar{a}_P^+\}$, $\{\bar{b}_r\}$ and

$\{\bar{b}_r^+\}$ should be (Svrček, 1986):

$$
\begin{aligned}
\bar{a}_P &= \sum_Q C_{PQ}(B,\tilde{B})a_Q\,, & \bar{a}_P^+ &= \sum_Q C_{PQ}(B,\tilde{B})^+ a_Q^+\,,\\
\bar{b}_r &= b_r + \sum_{PQ} D_{rPQ}(B,\tilde{B})a_P^+ a_Q\,, & \bar{b}_r^+ &= b_r^+ + \sum_{PQ} D_{rPQ}(B,\tilde{B})^+ a_P^+ a_Q\,.
\end{aligned}
$$

These general transformations are rather complicated. The general coefficients, $C_{PQ}(B,\tilde{B})$ and $D_{rPQ}(B,\tilde{B})$, satisfy complicated conditions. To make progress, we introduce a simple approximation in which the coefficients are written as products, i.e.

$$C_{PQ}(B,\tilde{B}) = C_{PQ}(B)\cdot\tilde{C}_{PQ}(\tilde{B})\,.$$

This is expected to be a good approximation since coordinates and momenta are independent variables. The coefficients $C_{PQ}(B)$ correspond to the adiabatic transformation describe above whilst coefficients $\tilde{C}_{PQ}(\tilde{B})$ correspond to the new transformation which we term the non-adiabatic transformation. The non-adiabatic representation results from the combination of both transformations. The canonical transformation from the old set of second quantized operators $\{a_P\}$, $\{a_P^+\}$, $\{b_r\}$ and $\{b_r^+\}$ to the new set of operators $\{\tilde{a}_P\}$, $\{\tilde{a}_P^+\}$, $\{\tilde{b}_r\}$ and $\{\tilde{b}_r^+\}$ is (Svrček, 1986):

$$
\begin{aligned}
\tilde{a}_P &= \sum_Q \tilde{C}_{PQ}(\tilde{B})a_Q\,, & \tilde{a}_P^+ &= \sum_Q \tilde{C}_{PQ}(\tilde{B})^+ a_Q^+\,,\\
\tilde{b}_r &= b_r + \sum_{PQ} \tilde{D}_{rPQ}(\tilde{B})a_P^+ a_Q\,, & \tilde{b}_r^+ &= b_r^+ + \sum_{PQ} \tilde{D}_{rPQ}(\tilde{B})^+ a_P^+ a_Q\,.
\end{aligned}
\tag{3}
$$

The coefficients $\tilde{C}_{PQ}(\tilde{B})$ and $\tilde{d}_{rPQ}(\tilde{B})$ are given by the Taylor expansions

$$\tilde{C}_{PQ}(\tilde{B}) = \sum_{k=0}^{\infty}\frac{1}{k!}\sum_{r_1\ldots r_k}\tilde{C}_{PQ}^{r_1\ldots r_k}\tilde{B}_{r_1}\ldots\tilde{B}_{r_k}, \qquad \tilde{D}_{rPQ}(\tilde{B}) = \sum_{k=0}^{\infty}\frac{1}{k!}\sum_{s_1\ldots s_k}\tilde{D}_{rPQ}^{s_1\ldots s_k}\tilde{B}_{s_1}\ldots\tilde{B}_{s_k}.$$

This transformation has the form of a supersymmetric transformation. We make the approximation that terms beyond first order can be neglected.

3.2 Fermionic part of the vibrational-electronic Hamiltonian

By performing the generalized canonical transformation defined in equations (3), we can obtain an expression for the fermionic part of the non-adiabatic Hamiltonian in the non-adiabatic representation. To simplify the notation, we shall omit the tilde from the operators $\{\tilde{a}_P\}$, $\{\tilde{a}_P^+\}$, $\{\tilde{b}_r\}$ and $\{\tilde{b}_r^+\}$ in the following. Our expression for the fermionic part of the non-adiabatic Hamiltonian can then be written in the form

$$
\begin{aligned}
H_F = {} & \overbrace{E^0_{NN} + E^0_{SCF}}^{1.} + \sum_{AI} \hbar\omega_2 \left(\left| C^r_{AI} \right|^2 - \left| \tilde{C}^r_{AI} \right|^2 \right) + \overbrace{\sum_{P} \varepsilon^0_P N\left[a^+_P a_P\right]}^{2.} \\
& + \sum_{PQr} \hbar\omega_2 \left[\sum_{A} \left(C^r_{PA} C^{r*}_{QA} - \tilde{C}^r_{PA} \tilde{C}^{r*}_{QA} \right) - \sum_{I} \left(C^r_{PI} C^{r*}_{QI} - \tilde{C}^r_{PI} \tilde{C}^{r*}_{QI} \right) \right] N\left[a^+_P a_Q\right] \\
& - 2 \sum_{PQr} E^{r*} \tilde{C}^r_{PQ} N\left[a^+_P a_Q\right] \\
& + \sum_{PQr} \left[(h(P) - p(P))\, \varepsilon^{r*}_P + (h(Q) - p(Q))\, \varepsilon^r_Q \right] \tilde{C}^r_{PQ} N\left[a^+_P a_Q\right] \\
& - \sum_{PQAIr} \left[\left(v^r_{PIQA} - v^r_{PIAQ} \right) \tilde{C}^r_{IA} + \left(v^r_{PAQI} - v^r_{PAIQ} \right) \tilde{C}^{r*}_{AI} \right] N\left[a^+_P a_Q\right] \\
& + \overbrace{\frac{1}{2} \sum_{PQRS} v^0_{PQRS} N\left[a^+_P a^+_Q a_S a_R\right]}^{3.} + \sum_{PQRSr} \hbar\omega_2 \left(C^r_{PR} C^{r*}_{SQ} - \tilde{C}^r_{PR} \tilde{C}^{r*}_{SQ} \right) N\left[a^+_P a^+_Q a_S a_R\right] \\
& - 2 \sum_{PQSr} \varepsilon^r_P \tilde{C}^{r*}_{SQ} N\left[a^+_P a^+_Q a_S a_R\right] \\
& + \sum_{PQRSTr} \Big\{ \sum_{I} \left[v^0_{PQTS} C^r_{TI} - v^0_{PQTI} C^r_{TS} + \left(v^0_{TQSI} - v^0_{TQIS} \right) C^r_{PT} \right] \tilde{C}^{r*}_{RI} \\
& + \sum_{I} \left[v^0_{TIRS} C^r_{QT} - v^0_{TQRS} C^r_{IT} + \left(v^0_{IQTS} - v^0_{IQST} \right) C^r_{TR} \right] \tilde{C}^{r*}_{IP} \\
& - \sum_{A} \left[v^0_{PQTS} C^r_{TA} - v^0_{PQTA} C^r_{TS} + \left(v^0_{TQSA} - v^0_{TQAS} \right) C^r_{PT} \right] \tilde{C}^{r*}_{RA} \\
& - \sum_{A} \left[v^0_{TARS} C^r_{QT} - v^0_{TQRS} C^r_{AT} + \left(v^0_{AQTS} - v^0_{AQST} \right) C^r_{TR} \right] \tilde{C}^{r*}_{AP} \Big\} N\left[a^+_P a^+_Q a_S a_R\right] ,
\end{aligned}
\tag{4}
$$

where we have overbraced the terms which arise in the usual electronic Hamiltonian operator. We can see immediately that if we set the $\tilde{C}_{PQ}$ coefficients to zero then we obtain the adiabatic Hamiltonian. If we put both the $\tilde{C}_{PQ}$ coefficients and the C_{PQ} coefficients equal to zero, we get the electronic Hamiltonian.

Let us now summarize, what we have achieved so far. We have derived a new kinetic energy term, as well as a new potential energy term, depending on the coefficients C_{PQ} and $\tilde{C}_{PQ}$.

$$
\begin{aligned}
H &= H_A + H_B , & H_B &= E_{\text{kinetic}}(\tilde{B}) + E_{\text{potential}}(B) , \\
E_{\text{potential}}(B) &= E^{(2)}_{NN}(B) + V^{(2)}_N(B) , & E_{\text{kinetic}}(\tilde{B}) &= T_N(\tilde{B}) + W^{(2)}_N(\tilde{B}) .
\end{aligned}
$$

We have to specify the C_{PQ} and $\tilde{C}_{PQ}$ coefficients from our transformation. We shall require that our new fermions, which we call *renormalized fermions*, are such that when partitioning the Hamiltonian H_A into an unperturbed part and a perturbation, the first order corrections vanish.

The equations for the C_{PQ} and $\tilde{C}_{PQ}$ coefficients are

$$u^r_{PQ} + \left(\varepsilon^0_P - \varepsilon^0_Q\right) C^r_{PQ} + \sum_{AI} \left[\left(v^0_{PIQA} - v^0_{PIAQ}\right) C^r_{AI} - \left(v^0_{PAQI} - v^0_{PAIQ}\right)\right] - \hbar\omega_r \tilde{C}^r_{PQ} = \varepsilon^r_P \delta_{PQ} \tag{5}$$

and

$$\left(\varepsilon^0_P - \varepsilon^0_Q\right) \tilde{C}^r_{PQ} + \sum_{AI} \left[\left(v^0_{PIQA} - v^0_{PIAQ}\right) \tilde{C}^r_{AI} - \left(v^0_{PAQI} - v^0_{PAIQ}\right) \tilde{C}^r_{IA}\right] - \hbar\omega_r C^r_{PQ} = \varepsilon^r_P \delta_{PQ} \,. \tag{6}$$

The new vibrational potential energy V^{rs}_N, which originate from the interaction between the nuclei and electrons, is:

$$V^{rs}_N = \sum_I u^{rs}_{II} + \sum_{AI} \left[\left(u^r_{IA} + \hbar\omega_r \tilde{C}^r_{IA}\right) C^s_{AI} + \left(u^s_{IA} + \hbar\omega_s \tilde{C}^s_{IA}\right) C^r_{AI}\right].$$

Let us compare this expression for the potential energy with that obtained in the representations considered previously in this paper. In the ‘crude adiabatic approximation’ we have

$$V^{rs}_N = \sum_I u^{rs}_{II} \,.$$

In the ‘adiabatic’ approximation, we have

$$V^{rs}_N = \sum_I u^{rs}_{II} + \sum_{AI} \left(u^r_{IA} C^s_{AI} + u^s_{IA} C^r_{AI}\right).$$

The kinetic energy term in the ‘renormalized fermion’ representation has the form

$$W^{rs}_N = 2\hbar\omega_r \sum_{AI} C^r_{AI} \tilde{C}^s_{IA} \,.$$

In a previous paper (Hubač and Wilson, 2008) (see also (Hubač and Svrček, 1992b,a)), we demonstrated that this Hamiltonian is equivalent to the diabatic Hamiltonian used in solid state physics theory. To demonstrate this equivalence we show its transformation to solid state notation.

3.3 Simplifications and connections with solid state theory

In this section, we shall introduce some simplifying approximations to the formalism developed above. In this way we shall establish connections with solid state theory.

In solid state theory, as in quantum chemistry, it is common to work with models or effective Hamiltonians. In general terms, a model is a conceptual representation of some physical phenomenon. Such models usually underpin computer programs which allow simulation and visualization of phenomena and/or processes. Examples of such approximations or models in solid state theory and quantum chemistry include the Hubbard model (Hubbard, 1963), the Hückel Hamiltonian (Hückel, 1931b,a, 1932, 1933; Streitwieser, 1961; Coulson et al., 1978), the Pariser–Parr–Pople (PPP) approximation (Pariser and Parr, 1953; Pople, 1953; Pariser, 1990b,a; Pople, 1990) and the Anderson Hamiltonian (Anderson, 1961).

In this section, we shall gain some understanding of the new terms which arise in the fermionic Hamiltonian given above in equation (4) by introducing some simplifying approximations, by considering model systems.

We have seen above that the C^r_{PQ} and $\tilde{C}^r_{PQ}$ coefficients are solutions of the equations (5) and (6).

Let us explore the simplifications obtained by putting

$$v^0_{PQRS} \to 0\,,$$

that is, by assuming that two-electron Coulomb interactions vanish. We then obtain from equations (5) and (6) the following pair of equations:

$$u^r_{PQ} + \left(\varepsilon^0_P - \varepsilon^0_Q\right) C^r_{PQ} - \hbar\omega_r \tilde{C}^r_{PQ} = \varepsilon^r_P \delta_{PQ}\,,$$
$$\left(\varepsilon^0_P - \varepsilon^0_Q\right) \tilde{C}^r_{PQ} - \hbar\omega_r C^r_{PQ} = \varepsilon^r_P \delta_{PQ}\,.$$

By solving these equations for C^r_{PQ} and $\tilde{C}^r_{PQ}$ we get:

$$C^r_{PQ} = u^r_{PQ} \frac{\varepsilon^0_P - \varepsilon^0_Q}{(\hbar\omega_r)^2 - \left(\varepsilon^0_P - \varepsilon^0_Q\right)^2} \tag{7}$$

and

$$\tilde{C}^r_{PQ} = \begin{cases} u^r_{PQ} \frac{\hbar\omega_r}{(\hbar\omega_r)^2 - \left(\varepsilon^0_P - \varepsilon^0_Q\right)^2} & \text{for } P \neq Q\,, \\ 0 & \text{for } P = Q\,. \end{cases} \tag{8}$$

Let us focus our attention on the term which corrects the ground state energy,

$$\Delta E_0 = \sum_{AIr} \hbar\omega_r \left(\left|C^r_{AI}\right|^2 - \left|\tilde{C}^r_{AI}\right|^2 \right).$$

Substitute the expressions for C^r_{PQ} and $\tilde{C}^r_{PQ}$ given in equations (7) and (8), respectively, we get:

$$\Delta E_0 = \sum_{AIr} \left|u^r_{AI}\right|^2 \frac{\hbar\omega_r}{\left(\varepsilon^0_A - \varepsilon^0_I\right)^2 - (\hbar\omega_r)^2}\,. \tag{9}$$

Now let us re-write equation (9) in a notation more familiar in solid state theory[1]. The boson vibrational modes will be denoted by the index of quasimomentum **q**, and spin orbitals (fermion) by (**k**, σ), where **k** is the quasimomentum of the fermion and σ is the spin

---

[1] See, for example, (Gross et al., 1991).

function. Then we can write:

$$
\begin{aligned}
&r \to \mathbf{q}\,, \\
&\tilde{r} \to -\mathbf{q}\,, \\
&I \to (\mathbf{k}, \sigma)\,, \text{occupation factor: } f_{\mathbf{k}}\,, \\
&A \to (\mathbf{k}', \sigma')\,, \text{occupation factor: } 1 - f_{\mathbf{k}'}
\end{aligned}
$$

together with

$$\epsilon^{0}_{I} \to \epsilon_{\mathbf{k}}\,, \quad \epsilon^{0}_{A} \to \epsilon_{\mathbf{k}'} \quad \text{and} \quad u^{r}_{AI} \to u^{\mathbf{q}}_{\mathbf{k}'\mathbf{k}} = u^{\mathbf{q}} = u^{\mathbf{k}'-\mathbf{k}}\,.$$

In the notation of solid state theory defined above, expression (9) for the energy correction becomes:

$$\Delta E_0 = 2 \sum_{\mathbf{k},\mathbf{k}'} \left|u^{\mathbf{k}'-\mathbf{k}}\right|^2 f_{\mathbf{k}} \left(1 - f_{\mathbf{k}'}\right) \frac{\hbar\omega_{\mathbf{k}'-\mathbf{k}}}{\left(\epsilon_{\mathbf{k}'} - \epsilon_{\mathbf{k}}\right)^2 - \left(\hbar\omega_{\mathbf{k}'-\mathbf{k}}\right)^2}\,.$$

This can be recognized as exactly the expression first given by Fröhlich (1950).

4 MAJORANA TRANSFORMATION OF THE DIABATIC HAMILTONIAN

In the previous section we demonstrated that our diabatic molecular Hamiltonian based on quasiparticle transformations through normal coordinates and corresponding momenta is fully equivalent to diabatic Hamiltonian used in solid state theory. This Hamiltonian plays a central role in theory of superconductivity and, more generally, in situations where electron-phonon interactions are important. Recently, this Hamiltonian was studied in connection of diabaticity and Majorana fermions (McKenzie, 2014).

In section 2, we recalled that the hydrogen bonds in the DNA molecule are known to be strongly diabatic systems (Scheurer and Shnirman, 2013). In the present study, we examine the diabaticity of the hydrogen bonds in the DNA molecule by using our diabatic molecular Hamiltonian with a Majorana transformation similarly to that studied in solid state theory (McKenzie, 2014).

In next section, we shall show that the hydrogen bonds in the DNA molecule carry quantum information through Majorana fermions. We show that the A-T and/or T-A segments (i.e. two hydrogen bonds, as can be seen in Fig. 2) segments represent one qubit. (Four Majorana fermions are necessary to form one qubit.) The segments C-G and/or G-C (i.e. three hydrogen bonds, as seen in Fig. 2) are entangled states and, therefore, carry quantum information (Tichy et al., 2011).

Majorana fermions for the molecular diabatic Hamiltonian can be introduced in the following way. The diabatic quasiparticle fermions are given by

$$\bar{a}_P = \sum_Q C_{PQ}(B, \tilde{B}) a_Q$$

and

$$\bar{a}^{+}_P = \sum_Q C_{PQ}(B, \tilde{B})^{+} a^{+}_Q\,.$$

We can put

$$\bar{a}_P = \gamma_1 + i\gamma_2$$

and

$$\bar{a}_P^+ = \gamma_1 - i\gamma_2\,,$$

where γ_1 and γ_2 are Majorana fermions. They are emergent quasiparticles. They play a central role in the quantum information theory associated with in the hydrogen bonds in the DNA molecule to which we turn in the following section.

5 MAJORANA QUBITS AND QUANTUM INFORMATION ENTROPY

Let us consider any fermion f with spatially well separated Majorana fermions γ_1 and γ_2. The Majorana fermions satisfy anti-commutation relations

$$\left\{\gamma_i, \gamma_j\right\} = 2\delta_{ij}\,. \tag{10}$$

It follows from (10) that $\gamma_i^2 = 1$. As a result there is no Pauli principle for the Majorana fermions. If we define a Majorana number operator $n_i^{MF} = \gamma_i^+\gamma_i$ we find $n_i^{MF} \equiv 1$. Similarly $\gamma_i\gamma_i^+ \equiv 1$. Experimentally measurable are the well-defined occupation numbers $n^f = 0, 1$ of the fermion f which are the eigenvalues of the fermion number operator $n^f = f^+f$ corresponding to the eigenvectors $|0, f\rangle$,$|1, f\rangle$.

To define Majorana qubits we need at least two fermions (Leijnse and Flensberg, 2012; Bravyi, 2006) with well separated Majorana fermions. Let us consider a system of three fermions f_1, f_2, f_3 each of them with spatially well separated Majorana fermions $\gamma_{1,k}$ and $\gamma_{2,k}$, $k = 1, 2, 3$. We define two subsystems of two fermions $X = (f_1, f_2)$ and $Y = (f_2, f_3)$. Let $|0, f_k\rangle, |1, f_k\rangle$ be the eigenvectors of the number operator $f_k^+ f_k$, $k = 1, 2, 3$.

For the subsystem X we define Majorana qubits as follows (Leijnse and Flensberg, 2012)

$$|0, X\rangle = |0, f_1\rangle\,|0, f_2\rangle\ ,$$
$$|1, X\rangle = |1, f_1\rangle\,|1, f_2\rangle\ .$$

These states may exist in a qubit superposition state

$$|q, X\rangle = a_0(X)\,|0, X\rangle + a_1(X)\,|1, X\rangle$$

with a density matrix

$$\rho(X) = |q, X\rangle\,\langle q, X| = \sum_{i,j=0}^{1} a_i(X)a_j^*(X)\,|i, X\rangle\,\langle j, X|\ . \tag{11}$$

Similarly, we define Majorana qubits for the subsystem Y

$$|0, Y\rangle = |0, f_2\rangle\,|0, f_3\rangle\ ,$$
$$|1, Y\rangle = |1, f_2\rangle\,|1, f_3\rangle$$

and their qubit superposition state

$$|q, Y\rangle = a_0(Y)\,|0, Y\rangle + a_1(Y)\,|1, Y\rangle$$

with a density matrix

$$\rho(Y) = |q, Y\rangle\,\langle q, Y| = \sum_{i,j=0}^{1} a_i(Y)a_j^*(Y)\,|i, X\rangle\,\langle j, X|\ .$$

The system S of subsystems X and Y may be entangled

$$|X, Y\rangle = a_{00}\,|0, X\rangle\,|0, Y\rangle + a_{01}\,|0, X\rangle\,|1, Y\rangle + a_{10}\,|1, X\rangle\,|0, Y\rangle + a_{11}\,|1, X\rangle\,|1, Y\rangle$$

with a density matrix

$$\rho(X, Y) = |X, Y\rangle\,\langle X, Y| = \sum_{i,j=0}^{1}\sum_{i',j'=0}^{1} a_{ij}a_{i'j'}^*\,|i, X\rangle\,|j, Y\rangle\,\langle i', X\big|\,\langle j', Y\big|\ . \tag{12}$$

A quantum state described by a density matrix ρ carries a quantum information $S(\rho)$ quantified by von Neumann entropy

$$S(\rho) = -\mathrm{Tr}(\rho\log\rho)\,,$$

where log is a logarithm with a base 2. The von Neumann entropy is zero $S(\rho) = 0$ for pure states ρ and is non-zero $S(\rho) \neq 0$ for mixed states. The values of the entropy are limited to the range

$$S_{\min} = 0 \leq S(\rho) \leq S_{\max} = \log N\,,$$

where N is the dimension of the Hilbert space.

6 QUANTUM INFORMATION IN DNA

We now apply the formalism developed in the Section 5 to the hydrogen bonds between complimentary base pairs in the DNA molecule. The hydrogen bonds in the DNA molecule can be viewed as a one-dimensional chain (or 'wire') of quasiparticles. In solid state physics, Majorana fermions have been found in systems containing a one-dimensional wire as well as superconductivity. Hirsch (2010) points out that in 1937 'London suggested that the diamagnetic currents in aromatic rings are analogous to supercurrents superconductors' (London, 1937). Aromatic rings are present in the pairs of complementary purines and pyrimidines in DNA. Figure 1 shows the structural formulae of the DNA molecule with the sugar-phosphate backbone of the two helices on the left- and right-hand side and the Adenine (A)-Thymine (T) and Guanine (G) -Cytosine (C) base pairs between them. There are two hydrogen bonds in A-T/T-A base pairing and three hydrogen bonds in G-C/C-G base pairing. Figure 2 shows the asymmetry of the hydrogen bond with a strong covalent bond X-H and weak hydrogen bonding to Y. In DNA complimentary base pairing. the strong bond always involves nitrogen and the weak bond involves either oxygen or nitrogen.

Recall that hydrogen bond can be viewed as a chain of fermion quasiparticles. This chain is delocalized. This means that there is a high probability p of finding $-i\gamma_2$ at the site X and a high probability p of finding γ_1 at the site Y. At the same time there is a low probability $1-p$ of finding $-i\gamma_2$ at the site Y and the same low probability $1-p$ of finding γ_1 at the site X.

This interpretation implies that in the hydrogen bond the Majorana fermions γ_1 and γ_2 are always spatially well separated and we can define Majorana qubits. Four Majorana fermions and therefore two hydrogen bonds are required to form a qubit. In the G-C or C-G base pairing the three electrons f_1, f_2, f_3 define the entangled two-qubit density matrix $\rho(X,Y)$ in (12) which we assume to be a mixed state with a non-zero entropy. In the isolated A-T or T-A base pairing we have single qubit pure state $\rho(X)$ in (11) with zero entropy. However two adjacent A-T and T-A base pairings can combine to form two-qubit states $\rho(X,Y)$ of systems $X = (f_1, f_2)$ and $Y = (f_3, f_4)$ formed by their two pairs of Majorana fermions. This state will also be assumed to be a mixed state with non-zero entropy. Since the probability of finding the Majorana fermion γ_2 at the site X is a high p and a low $1-p$ at the site Y, the combined density matrix is a mixed state

$$p\rho(X,Y) + (1-p)\rho(X,Y) = \rho(X,Y)\,.$$

A similar identity holds for the state $\rho(X)$.

The DNA molecule consists of protein coding and protein non-coding sequences of complimentary base pairs. In each sequence we assign to each base pair C-G or G-C, each adjacent pair of base pairs A-T/T-A and each isolated base pair A-T or T-A the value of the quantum information entropy of their Majorana density matrices. The corresponding sequence of the quantum information entropy we term Quantum Entropy Sequence (QES). The ensemble of all such quantum entropy sequences of the DNA molecule constitutes quantum information in DNA.

The question arises what is the role of the quantum information in the operation and evolution of the living organisms and their systems. Entropy is an important measure of the complexity of a system (Mitchell, 2009). It is well known that the fraction of protein non-coding sequences in DNA rises dramatically with the complexity of the organism and is largest in humans at ~98 % (Koruga, 2012). This suggests that the total quantum information content of the DNA increases with the complexity of the organism. The complex activity of the genome involves junk DNA in many complex ways (Carey, 2017). The expression of this complex activity may involve also the quantum states of the hydrogen bonds of the DNA bases and thus the quantum information sequences of both coding and non-coding sequences, relating the quantum information directly to the complexity of the living organisms. We suggest that the quantum information in DNA is related to the complexity and diversity of living organisms.

7 CONCLUSIONS AND OUTLOOK

We have shown that quasiparticle transformations leading to coupling of electronic and vibrational motion in the vibrational-electronic molecular Hamiltonian can be subjected to Majorana transformations to give relations that describe a chain of Majorana fermions akin to Kitaev chain. Majorana fermions that are localized at the two ends of this chain define a delocalized fermion state. Such delocalized fermions within a complex molecule define Majorana qubits.

We then make the suggestion that these Majorana qubits give rise to entanglement and form the foundation of molecular quantum information processes. We focus on systems with two and three fermions with well separated Majorana fermions and define their Majorana single-qubit and Majorana two-qubit density matrices, respectively.

We show that in hydrogen bonds of the A-T and C-G base pairs of the DNA molecule the Majorana fermions of the hydrogen electrons are spatially well separated. While the two-hydrogen bonds of isolated A-T or T-A base pairs are pure single-qubit states, the three-hydrogen bonds of the C-G or G-C base pairs are mixed two-qubit states. Two adjacent A-T/T-A base pairs are also mixed two-qubit states. In each protein coding or non-coding sequence we assign to each complimentary base pair A-T, C-G and the coupled base pairs A-T/T-A a value of von Neumann quantum information entropy thus forming a quantum information sequence. The ensemble of all such quantum information sequences in the DNA molecule constitutes its quantum information. We suggest that the quantum information in DNA is related to the complexity of living organisms and may play a role in the evolution of Life.

In this work, we have demonstrated a mechanism for quantum information processes in the DNA molecule but we have not considered how these processes might be exploited by nature. It is emphasize that this mechanism is distinct from the classical information processes currently used in explaining the function of the DNA molecule. It would be interesting, for example, to study the role of quantum information on the telomeres which form the end of chromosomes.

Quantum information in DNA could be exploited technologically in data storage systems. The recent paper by Mayer et al. (2016), for example, demonstrates that classical information in DNA can be exploited in practical data storage systems.

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