Mathematical formalism of femtosecond laser-deoxyribonucleic acid interaction: thermal evolution

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ABSTRACT

A novel analytical formalism is proposed based upon Quantum heat transport equation in order to describe the femtoseconds/picoseconds laser pulses interaction with the Deoxyribonucleic acid (DNA). The formalism generates solutions based upon inputs as: voltage, laser beam intensity and laser - DNA interaction time. Thermal waves induced inside irradiated DNA are defined and accounted for. Analytical simulations show that the optimum regime of laser - DNA interaction was reached for a potential carrier generated at the interface equal to $3.5 \times 10^{-3}$ eV. It has to be mentioned that the formalism breaks down if the potential carrier generated at the interface is inferior to $10^{-2}$ eV. Accordingly, for pulse duration inferior to 1 ps, the laser beam spatial-temporal distribution has an essential role in defining the shape and magnitude of the thermal distribution within the irradiated DNA strands.

1. Introduction

The theoretical description of the electronic transport across molecules has been originally published in ‘70s of the last century [1, 2]. A large interest was focused, during last decades, on molecular biology, in particular on Deoxyribonucleic acid (DNA) [3]. Many experimental and simulation studies have been recently carried out to investigate the laser - DNA interaction, mainly for bio-medical purposes. Thus, Chikawa et al. [4] simulated the interaction between an IR laser beam with a DNA strand submerged in a water solution. The elongation of DNA strand was investigated taking into account the convection heat of the laser-water interaction. It was shown that the convection heat plays a key role in the elongation of DNA length up to a long-axis diameter of $(20–25)$ μm after 30 s irradiation time. Oklay et al. [5] developed an analytical model to describe the external forces, including Stokes and hydrodynamic ones, in laser - DNA interaction. Pfits et al. [6] connected the damage in DNA strand to shock waves generated under the laser irradiation. They found that the shockwaves stay at the origin of the thermo-mechanical stress induced inside the irradiated DNA strand. Kim et al. [7] used a mode-locked fiber laser ($\lambda = 1563$ nm) to irradiate an aqueous DNA solution, as a potential saturable absorber. It was shown that the optical non-linearity of the DNA solution can control the lyotropic chiral nematic state that can be further valorized to study the DNA optical properties. Chen et al. [8] used ultra-fast laser pulses to investigate the excited state of DNA. They observed the existence of an emissive state population for nanoseconds period of time. As known [8], ultrafast laser pulses can be used to image processes at atomic level. Measurements of dynamic processes, deep inside an atom, were thus reported using ultrafast spectroscopy [9]. In the experiment, a Krypton atom was stimulated via a brief X-ray pulse which generates a hole inside the inner shell. The electrons rearrangement in the excited atom was monitored by a synchronized laser beam with a resolution of $10^{-18}$ s.

The quantum heat transport equation [10, 11] was applied via a novel mathematical formalism to investigate the heat generation and transport in the DNA structures as an effect of the irradiation with ultra-short (femtoseconds) laser pulses. At variance to Ref. 4, our approach explores the elongation of DNA strands in the case of a direct interaction with fs laser pulses and not only under simple solvent (water) heating. To
the best of our knowledge, there are not available experimental data to check up the predictions of the model. The closest experimental set-up was introduced in Ref. 4. There are however noticeable differences in respect with our work regarding the irradiation time, which was of about 30 s there.

The model is simple and accessible. This work is mainly intended to provide to experimentalists a possible guidance for future laser irradiation experiments of DNA with ultra-short laser pulses (of (1–1000) fs duration).

2. Mathematical formalism

Figure 1(a) depicts schematically the DNA strand interaction with the laser beam in a water medium [4]. Laser beam pulses of fs/ps duration cross and transfer through water medium the energy to the DNA strand causing heating by water convection.

One may consider (Figure 1a) that the propagation length inside liquid medium is long enough so that the laser beam with a power of 2 W does not directly interact with the DNA. Whenever the laser power is superior to 2 W, DNA will break down due to turbulence effects induced in the molecular structure of DNA. Separately to Figure 1a, in Figure 1b we assumed three major changes: i. short water level which involved an interaction between incident laser beam - DNA strand, ii. much less power laser beam, and iii. the laser beam is static and incident onto the beginning of DNA strand. The simulations carried out in the present study refer to experiments according to Figure 1b, only.

As shown in Ref. [12], DNA strand was demonstrated to be a good heat conductor. One may therefore assume that the two complementary DNA strands can provide a molecular-switch [13, 14, 15]. As known [16], a molecular-switch device can be operated between high- and low-resistance states, and was recognized as one of the key elements in molecular electronics.

According to the Kozlowski theory [16], whenever one irradiates DNA with ultra-short laser pulses, the one-dimensional (1–D) non-Fourier heat equation can be written as:

\[
\frac{\partial T}{\partial t} + \frac{\partial}{\partial x} \left( \frac{\nu \partial T}{h} + \frac{2 V \nu m_e T}{h^2} - \nu^2 \frac{\partial^2 T}{\partial x^2} \right) = 0
\]  

(1)

Here, \( T \) stands for the electron temperature, \( m_e \) is the mass of heat carrier in DNA, i.e., electrons, \( V \) is the potential carrier generated at the interface, \( t \) is the current time, \( x \) is the Cartesian coordinate, \( \nu \) is the velocity of heat propagation and \( h \) is the reduced Planck’s constant \( h = \hbar / 2 \pi \); with \( \hbar \) the Max-Planck constant.

\( \varepsilon \) (Eq. 2), \( K \) (Eq. 3) and \( \alpha \) (Eq. 4) are calculated based upon \( \nu \), \( m_e \) and \( V \), as follows:

\[
\varepsilon \equiv \frac{\nu^2 m_e}{\hbar}
\]  

(2)

\[
K \equiv \frac{2 V \nu^3 m_e}{\hbar^2}
\]  

(3)

\[
\alpha \equiv \nu^2
\]  

(4)

Eq. (1) can be rewritten accordingly, as:

\[
\frac{\partial^2 T}{\partial x^2} + \frac{\partial I}{\partial t} + K T - \alpha \frac{\partial^2 T}{\partial x^2} = 0
\]  

(5)

The reason to convert Eq. (1) into Eq. (5) is that the first one is more complex, difficult to handle and requires accordingly more computational time.

The source temperature, under given boundary conditions, is defined by Eqs. (6) and (7) [17]:

\[
T(x, 0) \propto I_0
\]  

(6)

\[
T(x, \infty) = 0
\]  

(7)

In Eq. (6), \( I_0 \) stands for the incident laser beam intensity.

Based upon the boundary conditions, the final solution was inferred as [17]:

\[
T(x,t)=I_0 \frac{e^{-\frac{\nu t}{2h\nu^3}}}{2\pi \nu^2} \left( \frac{m_e^2}{t^2} + 4 \left( \frac{2 \nu^2 m_e}{h} \right)^2 \right)^{1/4} \sqrt{\pi} \sqrt{K_1} \sqrt{K_2} \left[ \frac{m_e^2}{t^2} + 4 \left( \frac{2 \nu^2 m_e}{h} \right)^2 \right]^{-1/2} \left[ \left( \frac{m_e^2}{t^2} \right) \left( 1 + \frac{2 \nu^2 m_e}{h} \right) \right]^{1/2}
\]  

(8)

In Eq. (8), \( K_1 \) is the modified Bessel-function. In the current study, the value of \( \nu \) was assumed of 0.05 nm/fs [16].

One should mention that the notion of “heaton”, i.e., quanta of heat in Ref. 16, was not used in our model which resort to “classical” physics concepts only.

3. Simulation data

Table 1 compiles data from literature [16] on DNA and characteristics of laser pulses used in our simulations based on “MATHEMATICA” software.

A Core i7, 8th generation with 32 GB Ram computer has been used for the “MATHEMATICA” software computations. The computation time was of only 20s, i.e., quite efficient for molecular electronics simulations.
is why we continued the simulations with \( V_{3.5} \). We conclude that the optimum interaction regime corresponds to 1 ps and \( V_{jump} \) to 9 K in the case of 1 fs laser pulses, respectively. One may notice from Figures 2, 3, and 4 that the temperature on sample surface (\( x_{0} \)) is rapidly changing during laser irradiation because of the high heat conductivity of DNA, which intermediates the higher heat transfer from the surface to the sample bulk. Thus, in the case of 1 ps and 100 fs, the surface temperature variation increases from 0.03 to 2.3 K to jump to 9 K in the case of 1 fs laser pulses, respectively. One may observe that the thermal field in the case of 1 fs laser beam was incident onto the starting point of the DNA strand, i.e., from \( x = 0 \) μm. One can observe that the thermal field is almost constant around 300 K. In fact, the temperature variation in this case keeps very small, of about 1 K. One should take into account that the heat equations used in the present model are non-Fourier ones and this will lead to thermal waves propagation in the DNA sample.

The main difference between ultrashort (fs, ps) versus ns laser pulses irradiation is related to the scarce presence of a liquid (melted) phase in the first case. This is reflected in the constant temperature in liquid (melt) and a very low temperature variation in bulk phase during the ultrashort laser pulses irradiation.

Our simulations showed that the proposed formalism fails down for the spatial-temporal evolution of the temperature field in the case of 1 fs laser pulses - DNA strand interaction for a potential of \( V = 3.5 \times 10^{-3} \) eV. The laser beam was incident onto the starting point of the DNA strand, i.e., from \( x = 0 \) μm. One can observe that the thermal field is almost constant around 300 K. In fact, the temperature variation in this case keeps very small, of about 1 K. One should take into account that the heat equations used in the present model are non-Fourier ones and this will lead to thermal waves propagation in the DNA sample.

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Figures 3 and 4 illustrate the predictions of the proposed formalism for irradiation by pulses of 100 fs and 1 ps duration, respectively. It can be noted that the simulations carried out in the case of longer laser pulses (of 100 fs and 1 ps) better describe the behavior of DNA as a thermal switch [16].

4. Results

In the following, \( \Delta T \) stands for the temperature variation as an effect of laser irradiation, i.e., the difference between the final and initial actual temperature sample values. In all cases, the simulation time range is identical with pulse duration, as also done in Ref. 16. Numerical simulations were conducted within the whole fs range: from 1 to 1000 fs (Figures 2, 3, and 4).

Figure 2 displays the spatial-temporal evolution of the temperature field during laser - DNA interaction in the case of a laser pulse of 1 fs duration for a potential of \( V = 3.5 \times 10^{-3} \) eV. The laser beam was incident onto the starting point of the DNA strand, i.e., from \( x = 0 \) μm. One can observe that the thermal field is almost constant around 300 K. In fact, the temperature variation in this case keeps very small, of about 1 K. One should take into account that the heat equations used in the present model are non-Fourier ones and this will lead to thermal waves propagation in the DNA sample.

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5. Discussion

One notes from Figures 2, 3, and 4 that the temperature on sample surface (\( x = 0 \)) is rapidly changing during laser irradiation because of the high heat conductivity of DNA, which intermediates the higher heat transfer from the surface to the sample bulk. Thus, in the case of 1 ps and 100 fs, the surface temperature variation increases from 0.03 to 2.3 K to jump to 9 K in the case of 1 fs laser pulses, respectively. One may conclude that the optimum interaction regime corresponds to 1 ps and \( V = 3.5 \times 10^{-3} \) eV when the highest temperature variation of 9 K is reached, i.e., under the limit of 10 K mentioned in Ref. [4] to avoid DNA irreversible damage. These results are in accordance with those reported in Ref. 16 but our formalism is direct, simpler and analytically self-consistent.

6. Conclusions

The heat transport is considered in this study within DNA strands submitted to irradiation with \( \leq 1 \) ps laser pulses. We showed via the quantum heat transfer equation that significant thermal waves are generated inside the DNA strands. An exact analytical formula was inferred for thermal distribution in DNA strands heated under the action of laser pulses with duration inferior to \( \leq 1 \) ps. Under context, the model fails however if the potential at the interface is inferior to \( 10^{-2} \) eV. The laser power should not therefore exceed 2 W in the case of the laser - DNA interaction via liquid medium and even less for a direct laser - DNA interaction. The main constrain of the model is therefore related to the correct choice of the interaction potential (V), which if assumed too high can result in the misprediction of the DNA breaking. The presented results were inferred after consistent and rigorous computer simulations
via a mathematical formalism. The current model proved efficient in terms of calculation time and data presentation as compared to Kozlowski formalism [16], as it generates reliable results within no more than 20 s using a Core i7, 8th generation and 32 GB Ram computer.

**Declarations**

**Author contribution statement**

Mihai Oane & Carmen Ristoscu: Conceived and designed the experiments; Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data; Wrote the paper.

Bogdan A. Sava & Ana V. Filip: Analyzed and interpreted the data; Wrote the paper.

Muhammad Arif Mahmood & Ion N. Mihailescu: Conceived and designed the experiments; Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data.

Cristian N. Mihailescu: Conceived and designed the experiments; Analyzed and interpreted the data.

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**Data availability statement**

Data included in article/supp. material/referenced in article.

**Declaration of interest’s statement**

The authors declare no conflict of interest.

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