DNA – the fascinating biomacromolecule in optoelectronics and photonics applications

M.-E. BARBINTA-PATRASCU[1], S. M. IORDACHE[2,*]
[1]*University of Bucharest, Faculty of Physics, Department of Electricity, Solid-State Physics and Biophysics, 405 Atomistilor Street, PO Box MG-11, Bucharest-Magurele, 077125, Romania*
[2]*National Institute for Research and Development for Optoelectronics - INOE 2000, Optospintronics Department, 409 Atomistilor Street, PO Box MG-05, Magurele, 077125, Romania*

Taking into account the global situation regarding environmental pollution, the use of natural resources must be one of the priorities of scientists to find clean technologies for the development of materials with new and interesting properties. Thus, bio-residues can be used as a source of extraction of deoxyribonucleic acid (DNA) – the fascinating and the "smartest" biomolecule. This paper presents a brief survey of the nongenetic properties of DNA and the advances of its applications in optoelectronics and photonics, in the last twelve years. This fascinating biomaterial should be more and better exploited in the field of optoelectronics and photonics. An analysis of the studies from the last twelve years shows a growing interest in the DNA use in these fields, which, however, is still in infancy and therefore, not fully exploited as it deserves. Finally, this paper points out some challenges and perspectives of DNA applications in optoelectronics and photonics.

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1. Introduction

Deoxyribo Nucleic Acid (DNA) is *the bio-informational molecule* with a double right-handed helical structure (Watson–Crick model, Fig. 1) composed of two intertwined strands arranged such that the *sugar-phosphate backbone* (consisting of alternation of sugars and phosphate groups) lies on the outside and the base pairs on the inside [1]. This smart biomolecule stores and conveys the genetic information (required for development and functioning of all living organisms) which is coded in genes composed of *codons* - the triplets made up of the nitrogenous bases.

DNA is an anionic biopolymer; its building blocks being *nucleotides* which are molecules made up of three components: 1) a *phosphate group*; 2) a *sugar molecule* (pentose) - *deoxyribose*; 3) a *nitrogenous base* that can be *pyrimidine*: cytosine (C) and thymine (T), and *purine*: adenine (A) and guanine (G).

According to Chargaff's rule, a purine situated on a DNA strand is always matched by a pyrimidine on the other strand, by a complementary association: G is paired with C through 3 hydrogen bonds, while A paired with T through 2 hydrogen bonds. These hydrogen bondings are crucial for DNA stability, being responsible also for the DNA *nano-oddities* [2] arising from its ability for self-recognition and self-assembly, enabling the interactions with itself or with other biomolecules (e.g., proteins, lipids, polysaccharides) or with nanoparticles [3, 4], properties exploited by scientists to design novel DNA materials with great potential for various applications.

The structural, optical and electrical properties of DNA were exploited by scientists, in various applications including biomedicine and forensic science [5, 6], nanotechnology [7], agriculture [8], energy storage and conversion, catalytic and sensing applications [9, 10], molecular logic systems [11], flame-retardant, materials science [12-14], (bio)electronics and (bio)photonics [15-18].

Herein, some applications of DNA in optoelectronics and photonics are briefly reviewed.

DNA applications in *biotronics* (the research area that uses biologically-based materials for electronics and photonics) were firstly proposed in 1990s by Professor Naoya Ogata (Chitose Institute of Science and Technology, Japan), as stated by Kawabe and Yoshida in their paper "Progress in DNA Photonics and Electronics - Tribute to Naoya Ogata" [19]. He succeded to turn salmon DNA wastes into valuable materials for photonics and electronics applications.

Optoelectronics (also called *optronics*) is a branch of electronics dealing with electronic devices for emitting, modulating, transmitting, and sensing light [20]. Furthermore, optoelectronics also "*concerns the study and application of electronic devices that source, detect and control light*" [21].

Photonics is a branch of physics dealing with the properties and applications of photons especially as a medium for transmitting information [22]. Photonics is known as "*technology for generating and harnessing light, whose quantum unit is the photon*" [23].

In the following, an analysis of the evolution of publications related to DNA in optoelectronics and in

photonics, in the last twelve years (according to Web of Science Core Collection) will be further presented.

Despite the fact that DNA is a unique material with fascinating properties, the studies on DNA applications in optoelectronics and photonics are scarce and still in their infancy. Temporal distribution of publications (for the last 12 years analysis according to WOS) shown in Fig. 2 a and Fig. 2 b revealed the increasing number of publications for „DNA optoelectronics" as compared to „DNA photonics", but the number of publications in „DNA photonics" is greater than that in „DNA optoelectronics".

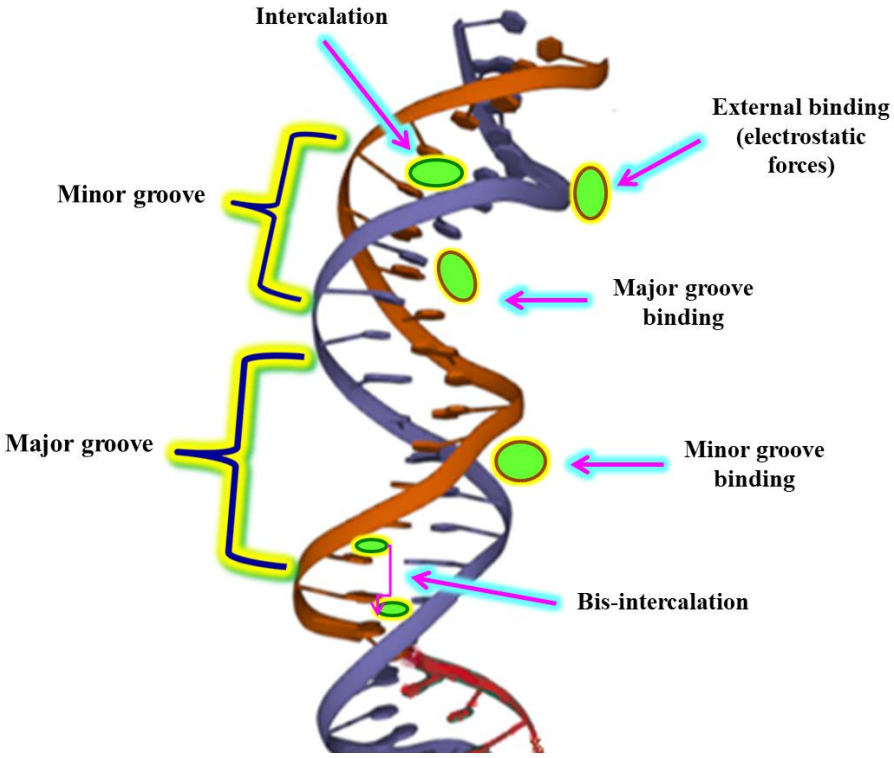

Fig. 1. The Watson-Crick model of DNA double-helix, and the main binding modes of fluorescent dyes on dsDNA, according to [6,24,25]. The DNA-bis-intercalators contain two intercalating units separated by a linker chain. (The DNA molecule was generated with https://www.rcsb.org/3d-view/2IEF.)

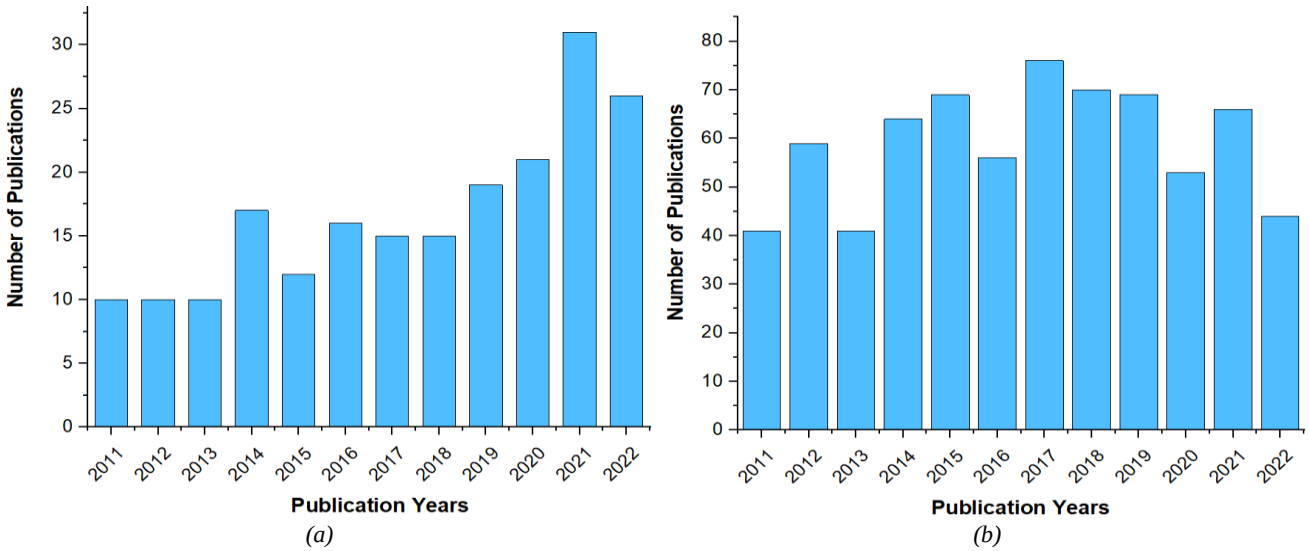

Fig. 2. The analysis results of publications, for the last 12 years, selected from Web of Science Core Collection, related to the number of publications by using the keywords: "DNA optoelectronics" (a) and "DNA photonics" (b), according to publication years. (Source WEB OF SCIENCE, WOS, accessed on 19th November 2022)

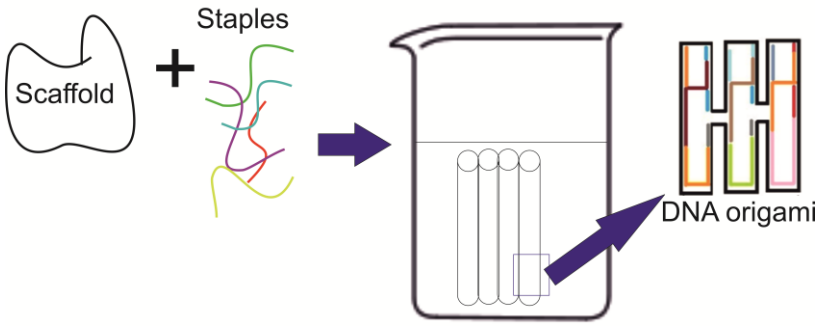

Fig. 3. Schematic presentation of principle of the DNA origami assembly (Figure adapted upon Loretan et al. [26])

2. DNA origami nanostructures

Similar to paper folding, DNA origami is created by folding a single, long strand of DNA (usually a viral DNA) into different architectures by using staples placed at different intervals. This process was developed by Paul Rothemund, in 2006 [27].

There are three main strategies to design DNA nanostructures [28] as follows: (a) *multi-stranded design* or the conventional approach used to construct a supramolecular design by assemblying short oligonucleotides (commonly known as "DNA tiles"); (b) *single-stranded design* composed of one single, long scaffold DNA and few or no ''helper strands", and (c) *scaffolded design*, composed of a single, long DNA strand with a multitude of staples placed arbitrary lengthwise. The (b) and (c) categories are commonly known as "origami DNA" because of their capability to fold on the staples and form various structures with erratic geometries.

The new structures resulted from the folding of single strands of DNA have different properties for specific applications.

As stated by Loretan *et al*. in a recent review [26], DNA origami is an emerging technology for the engineering of fluorescent and plasmonic-based biosensors. The DNA origami method consists of an annealing process (taking place in solution under temperature and salt concentration conditions) of long single-strand DNA ("scaffold") and several hundreds of short ssDNA strands ("staples"), resulting in the design of highly diverse, complex, multi-functional, 2D, and 3D structures. The principle of the DNA origami assembly is presented in Fig. 3.

DNA origami technique allows a facile design and development of nanoarchitectures with fascinating properties: self-assembling, reconfigurability, and self-repairing [29].

Tekoglu *et al*. [30] used DNA-cetyltrimethylammonium (DNA-CTMA) as ion-solvating component for light-emitting electrochemical cells (LECs). The researchers observed that upon blending with the Super Yellow emissive polymer, the structure exhibits wide electrochemical window and a visible light emission covering the entire visible spectrum, which ensures an optimized LEC performance. Although improved, the performance of the biopolymer does not surpass the performance of the synthetic ion-solvating polymers in LECs. Another group of researchers focused on the development of eco-friendly devices in the field of nanoelectronics is that at the Brigham Young University, USA [31]. These researchers have annealed at 170 ^{0}C, Au and Te nanorods onto DNA origami, thus developing metal-semimetal junctions simply by controlling seeding on DNA origami templates. The pH of nanocomposites can be easily assessed by using DNA origami nanocalipers, developed by Zhang *et al*. [32]. This nano-device has a scissor-like structure that switches between open and close angles, when the environmental pH varies. The variation of open/close transition is visible *via* TEM imaging.

The wide range of applications of the DNA origami nanoassemblies is due to four fundamental properties: elasticity, pliability, plasticity and stability [33]. Elasticity and pliability are reversible changes in the structure of the supramolecular system, while plasticity and mechanical stability are irreversible and inherent to the fragmentation of the DNA-assembly. In the world of nanoelectronics, the properties of the material tailor the application of the DNA-based composites towards a certain field. Besides the four fundamental properties, other characteristics such as conductivity, charge transport, electronic level, current distribution, and surface potential [34] modulate the material either for energy capture, biosensing or nanorobotics.

The disadvantages of this new, emerging technology are in terms of the accurately positioning the staples and other single molecules that are drafted on the DNA – origami.

Cervantes-Salguero *et al*. [35] presented two strategies based on density functional theory and molecular dynamics simulations for controlling the polar and in-plane azimuthal angular orientations for the immobilized molecule.

Wang *et al*. [36] constructed AND gate (which is an electrical circuit acting in the same way as the logical "and" operator) and NAND gate (negative AND gate; it operates as an AND gate followed by a NOT gate, https://www.techtarget.com/whatis/definition/logic-gate-AND-OR-XOR-NOT-NAND-NOR-and-XNOR) models based on DNA origami platform. The gates use polymerase strand displacement reaction and hybridization chain reaction in order to give True or False judgment,

which is produced by the presence of fluorescence on the origami platform.

The perfect placement of the metalic nanoparticles on DNA origami is paramount to surface-enhanced Raman spectroscopy (SERS) [26], with applications in plasmonic-based biosensors. Several researchers have used Au, Ag and dyes to enhance the SERS signal to 9 orders of magnitude to bring the sensibility down to single molecule [37, 38]. By processing the data with spectral inverse quantum (Spectral-IQ) method [39], the sensibility could be pushed to lower concentrations, making DNA origami structures effective plasmonic optical antennas.

A step further was perfomed by Zhao *et al*. [40] who created "the origami of origami", which is a scale-up DNA origami technology. It uses a loose DNA framework into which large origami tiles are introduced and act as large staples to supra-organize the structure into larger structures. Daems and co-workers [41] introduced for the first time, two distinct DNA origami structures as bioreceptor carriers to functionalize a fiber optic surface plasmon resonance sensor with nanoscale precision. This constitutes potential applications in biomedical field as biosensing platforms, being beneficial both for scientists and clinicians looking for improved diagnostics.

A novel application of the DNA origami is the development of "DNA walkers". A DNA walker is a dynamic molecular nanomotor composed of walking strands and walking tracks and endowed with the ability to generate a driving force [42]. The most important feature of the nanomotor is the driving force, which uses strand displacement reaction, enzymatic reaction, optical and chemical stimulus to "sense" the analyte and "walk". The walking strands (i.e., the legs) could be uni-, bi-, and multipedal. The main strenghts of these DNA walkers are their high programmability and integrability. DNA nanostructures can be easily integrated onto sensing platforms such as "lab-on-a-chip" which could detect many molecules down to ppb. The rapid development of DNA computing leads to improvements in the recognition algorithm which lowers even more the detection limit of the analytes. Nevertheless, one has to keep in mind that the amplification and sensing patterns are complicated, involving at least 4 elements which limit its efficiency. Another drawback is the fact that side-reactions are likely to occur due to the sophistication of the sensing path, thus decreasing the selectivity towards a specific analyte.

3. Non genetic properties of DNA in relation to the suitable applications in optoelectronics/photonics

An insight into the nongenetic properties of DNA related to its applications will be further presented.

3.1. DNA charge

DNA is negatively charged due to the presence of negative phosphate groups in its nucleotides. This charge allows the electrostatic attraction with cations (Na^+, K^+, etc.) or with positively charged molecules [including histones, chitosan (CTS), etc.]. Therefore, DNA is an anionic biopolymer and its interactions with polymeric polycations have been exploited to obtain DNA-based biosensors, with distinctive physical-chemical properties caused by small size, quantum confinement, and macro quantum tunnel effects [43,44]. Thus, Peyman *et al.* [45] developed through a fast, simple, sensitive, and cost-effective method, a DNA-based electrochemical biosensor using DNA immobilized on CTS and single walled carbon nanotubes (SWCNTs) modified Au electrode for biodetection of Pirazon - a herbicide used for general weed management which inhibits photosynthesis.

The process of complexing DNA with different polymeric polycations is pH dependent. For example, Amaduzzi and co-workers [46] pointed out that by increasing pH, the CTS amount required to complex DNA "to saturation" (i.e., to the point that excess "free" CTS remains uncomplexed in the solution) increases, because at high pH values, the effective CTS charge is reduced.

On the other hand, the negative charges of DNA allow its separation in an electric field, by electrophoresis.

3.2. Conductive properties of DNA. DNA metallization

For a long time DNA has been considered to be an insulator [47], but the report of Fink and Schönenberger in 1999 [48] demonstrated that DNA could be a very good conductor, and surprisingly, after two years, Kasumov *et al*. reported DNA as a superconductor [49]. From this moment, a special interest was given to DNA in the optoelectronics field. These huge differences between these results highlighting conductive properties of DNA from insulating to poorly conductive to highly conductive behaviour come from the different experimental conditions [50] such as: various DNA lengths and different DNA sequences, altered surrounding conditions, different contacts between DNA and electronic circuits, showing that conductance properties of DNA are very sensitive to experimental parameters.

DNA is a "dream molecule" suitable for the design and fabrication of the new nano-electronic devices. Priyadarshy reviewed some reports related to DNA conductance [51]. He pointed out that DNA molecule acts as an electronic *molecular wire*, facilitating the charge (electron and/or hole) transport which is closely related to intramolecular electron transfer in a donor-bridge-acceptor (D═B═A), when the bridge (DNA) acts as a molecular wire. Priyadarshy presented the theoretical background on long range electron transfer and concept of electron mobility in molecular systems. In such a D═B═A system, when the molecules of donor [the excited state of ethidium bromide (EB^+)] and the acceptor [N,N'-dimethyl-2,7-diazapyrenium (DAP^{2+})] were covalently bound to the DNA (the bridge), and intercalated within the DNA stack, an electron transfer rate of 2.5×10^6 s^{-1} for a distance of 17 Å was observed.

To increase the electronic conductivity, DNA has been functionalized with metallic nanoparticles or with

semiconducting polymers, such as polyaniline, polypyrrole, polythiophene, and their derivatives. Kim *et al* [52] prepared DNA:CTMA, PEDOT-S:CTMA, and DNA:CTMA:PEDOT-S:CTMA complexes which showed an electronic conductivity of as much as 10^{-6} - 10^{-5} S/cm in the conductivity range of semiconductors.

Metal incorporation into the DNA-structure, process called *DNA metallization* resulting in metallized DNA (M-DNA), is a method used to overcome the problems related to the use of native DNA in electronic circuits, due to the poor conductance properties of DNA [50].

The concept of *DNA metallization* was firstly proposed in 1998 by Braun and coworkers [53] who reported the first work of DNA metallization in order to fabricate a conductive Ag nanowire on the contour of bacteriophage λ-DNA.

The preparation of M-DNA involves a bottom-up process. In their review, Li *et al.* [54] pointed out that self-assembled DNA nanostructures have shown remarkable potential in the engineering of biosensing interfaces. They summarized the current applications of electrochemical biosensors based on the framework nucleic acids (FNAs, which are DNA nanostructures with controllable sizes, such as 3D shells or skeleton DNA frameworks), including the detection of nucleic acids, ions, small molecules, proteins, and cells. Moreover, the electrochemiluminescence properties of a "nanoswitching-type" electrochemical DNA biosensor (with FNAs) were plasmon-enhanced by the addition of Au nanodendrites in this system [55].

The DNA metallization provides new strategies for DNA-based memristive technology and also for a broad range of applications such as optical storage, plasmonics, and catalysis [56].

3.3. Bioelectromagnetism within DNA

DNA has a very weak dipole moment. Hydrogen bonds between nitrogenous bases in the DNA double helix, may act as dipoles, and their movement could lead to spontaneous emission or absorption of electromagnetic radiation. Similar to other polymers, DNA does not seem to exhibit any intrinsic bioelectromagnetism, but acts as a complex antenna by absorbing and re-transmiting any incoming electromagnetic waves [57]. Bukhari *et al*. stated that "*even if there were a possibility of existent bioelectromagnetism within purified DNA, its amplitude would be extremely faint and impossible to measure using human methods, including some of the most sensitive and highest precision methods available today*" [57].

3.4. Optical properties of DNA

3.4.1. UV–Visible absorption spectra of DNA

Useful information (both qualitative and quantitative) regarding the DNA structure and its analytical control can be obtained by recording the UV absorption spectrum of the DNA solution. DNA molecules absorb radiation from the UV range, namely, in the 200-290 nm region with a characteristic maximum at 260 nm (Fig. 4), due to the $\pi \rightarrow \pi^*$ transitions assigned to the aromatic structures of the nitrogenous bases (purines and pyrimidines) in which are the determining chromophores of nucleic acids due to the presence of the heterocyclic structures with conjugated double bonds. The absorption band of natural DNA below 225 nm originates from the overlapping electronic absorptions of phosphate groups and sugar moieties [58].

The characteristic absorbance at 260 nm is used to calculate the DNA concentration in a sample, by using the Lambert-Beer-Bouguer law.

UV-Visible absorption spectroscopy also allows the study of the thermal denaturation of the DNA that is, the transition: native, double-stranded DNA → single-stranded DNA (dsDNA → ssDNA) (Fig.4). Denatured DNA exhibits the phenomenon of hyperchromicity, i.e. a stronger absorption than native DNA. Double-stranded DNA shows a lower absorption (hypochromicity) than the sum of the absorptions of the component mononucleotides in the free state, because the nitrogenous bases, the chromophores of nucleic acids, are "masked" inside the DNA molecules, where they are linked by hydrogen bridges [1]. The characteristic hyperchromic effect of denatured DNA molecules is explained by the opening of hydrogen bridges, the purine and pyrimidine bases no longer being "stacked" in the helix, resulting in reduction of their optical absorption capacity. In this way, the purine and pyrimidine bases in ssDNA behave spectrophotometrically as if they are free in solution, causing ssDNA to exhibit up to 40% higher absorbance, compared to their position into dsDNA. The degree of optical absorption increase depends on the DNA type.

Strong interaction between a ligand and the right-handed double helix DNA could result in hyperchromic or hypochromic effect in the DNA electronic absorption spectra. Hypochromism is due to the axial compression of DNA double helix, by electrostatic binding and hydrophobic interactions, thus causing a slight change in the DNA conformation [45, 59]. On the contrary, the hyperchromism occurs when the DNA double helix structure is damaged [45].

The studies of Nowak *et al*. [59] showed that the addition of three ionic liquids, ILs [3 different cationic surfactants with short chains (allyl, *n*-butyl, and methyl)] into DNA solution caused a hypochromic effect on the DNA absorption spectra at 260 nm, without the peak shift. These optical changes make the DNA–IL complexes potential candidates for biosensor applications.

3.4.2. DNA FTIR spectroscopy

The FTIR spectrum of a pure salmon DNA in B form possesses the main vibrational bands located in the following wavenumber regions [59]:

- 3,000-3,400 cm^{-1}: broad band assigned to N–H-stretching modes, C=N vibrations, OH symmetric, and antisymmetric stretching modes;

- 1,800-1,400 cm^{-1}: bands comming from nucleobase vibrations, which are extremely sensitive to base sets and base-pairing interactions.
- 1,750-1,600 cm^{-1}: broad bands including stretching and bending mode (C=C, C–N, C=O, and NH_2) vibrations arising from base molecules.
- 1,500-1,250 cm^{-1}: marker bands originating from vibrations connected with base and base-sugar units. This region gives information about nucleoside-specific interaction and conformation.
- 1,250-1,000 cm^{-1}: markers of backbone conformation (A-, B-, or Z-form) due to vibrations along the sugar–phosphate bone. The A-, B-, and Z-form marker bands are located at 1,240 cm^{-1}, 1,221 cm^{-1}, and at 1,215 cm^{-1}, respectively [60].
- 900-800 cm^{-1}: absorption bands assigned to vibrations of the sugar–phosphate groups; these bands are intense markers for the various sugar-puckering modes (N- and S types).

Table 1 displays the assignment of the main FTIR absorption bands for pure salmon B-DNA.

FTIR spectroscopy is a useful tool to assess the nature of the DNA-ligand interactions, and also to confirm the formation of DNA-based complexes.

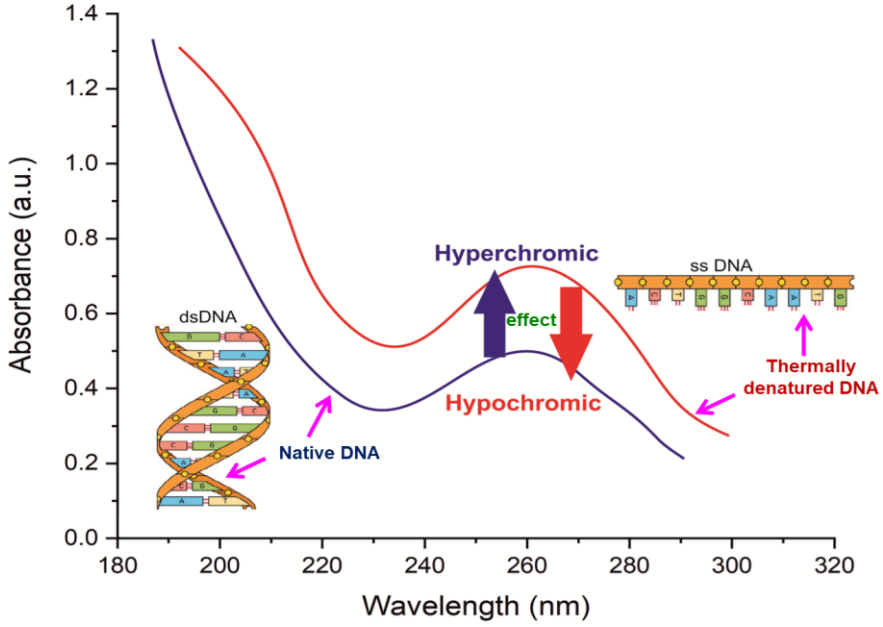

Fig. 4. Thermal denaturation of the DNA molecule monitored by UV-Visible absorption spectroscopy (adapted upon https://www.biologyexams4u.com/2018/01/ds-or-ssdna-has-more-absorbance-double.html)

Table 1. The main FTIR absorption bands assignment for pure salmon B-DNA

FTIR absorption bands (cm^{-1})	Assignment	Ref.
3,400–3,000 cm^{-1}	Broad band that could be attributed to N–H-stretching modes, C=N vibrations, OH symmetric, and antisymmetric stretching modes.	[59]
1,683 cm^{-1}	Sharp band assigned to the vibrations of C6=O of guanine and C4=O of thymine	[59]
1,643 cm^{-1}	Vibration of C2=O of cytosine	[59]
1,603 cm^{-1}	Vibrations of C=N7 of guanine	[59]
1,530 cm^{-1}	N–O-stretching vibration of secondary amine groups	[59]
1,456 cm^{-1}	C=C-stretching vibration attributed to purine imidazole ring vibration	[59]
1,221 cm^{-1}	Marker band of backbone B- conformation due to vibrations along the sugar–phosphate bone	[59] [60]
1,057 cm^{-1}	Vibrations of the –C–O–P sugar–phosphate chains	[59]

3.4.3. DNA fluorescence

As compared to other biomolecules, DNA molecules exhibit extremely weak intrinsic fluorescence [59, 61].

In order to enhance its fluorescence, the DNA molecule must be labelled with fluorescent dye molecules which could interact with DNA through intercalation, groove binding or external stacking [6, 24, 25, 62] as displayed in Fig.1. The intercalation of DNA-binding dyes involves firstly the interaction with phosphate chains by electrostatic forces, and then the dye intercalation between DNA base-pairs [62]. One example of fluorescent intercalator is Rhodamine 6G (Rh6G), one of the most efficient laser dye, which contains benzene rings with cationic NH- groups [62] involved in hydrogen bonding that facilitates Rh6G binding on DNA. Some applications

of fluorescent properties of DNA-Rh6G will be further presented.

The fluorescence is a very sensitive method allowing detection of very small amounts of samples. Thus, it could be mentioned the fluorescence method developed by Ma *et al*. [63], for simultaneous recognition of ssDNA and dsDNA, based on the quenching of the fluorescence of fluorophore labeled DNA probes by gold nanoparticles (AuNPs), with detection limits as low as 330 pM.

On the other hand, the fluorescence detection and quantification of low amounts of dsDNA using fluorescent dyes is very important in forensic DNA-analysis [6].

The unique features of DNA associated with enhanced optical performance of dye-doped DNA complexes bring perspectives for organic solid-state lasers and lasing applications. Thus, Hung *et al*. [64] developed optical films made of DNA modified by two types of surfactants [cetyltrimethylammonium (CTMA) chloride, and benzyltrimethylammonium (BTMA) chloride] and doped with Rh6G which exhibited fluorescence enhancement. Kitazawa *et al*. [62] used another surfactant – tetradecyltrimethylammonium (TTA) for preparation of Rh6G-doped DNA-TTA film with enhanced optical properties and photostability.

Manea-Saghin *et al.* [58] enhanced the fluorescence of DNA-CTMA complex by doping DNA with two organic molecules: 5- formyl-13-hydroxy[2.2]paracyclophane and 5-formyl-12-hydroxy[2.2]paracyclophane. Under the UV irradiation, both materials exhibited a large transparency in visible range and fluorescence. Moreover, the fluorescence emission band in visible range is well separated from the absorption band, showing a high potential for LED application.

4. DNA-based materials

Based on DNA properties which are in close correlation with its strucure, valuable materials have been developed.

4.1. Methods of fabrication

Self-assembling

Self-assembling DNA techniques pioneered by Seeman, Rothemund, and many others now provide a nanoscale design of DNA nanomaterials [65].

Self-assembling process of the DNA leads to 0D (dots, nanoparticles), 1D (wire, nanotubes), 2D (sheets) and 3D (hydrogels) structures [10]. Thin films (2D) are the most researched structures for DNA application. Anton *et al*. [66] used natural chromophores extracted from fruits, leaves or roots to functionalize DNA and DNA-CTMA chloride complex to imprint nonlinear optical properties to the thin film. The authors observed that the nonlinear optic susceptibility varies with the conjugation length, the largest one being obtained from DNA-CTMA-curry extract. Kesama *et al.* [67] functionalized DNA with semiconducting ZnO and CuO nanoparticles to produce composites with tunable properties, such as: increased absorbance, larger band edge shift and higher resistance. Shi *et al.* [56] used Ag for the functionalization of a solid DNA film to investigate the UV light driven metallization process and to elucidate the kinetics and mechanism of particle evolution.

Mousavi *et al*. [68] have calculated the band structure, density of states and electrical conductivity for the DNA – based wires since their fabrication is extremly difficult. Kim *et al*. [69] fabricated NiO nanowires using *Escherichia coli* DNA as a structural template. The new anode exhibited a large reversible capacity of 850 mA h g^{-1} and stable retention capability for 150 cycles. The process used by authors was „DNA metallization" (see section 3.2), a method that has been employed for the fabrication of a variety of composite biomaterials with tunable physical properties for optical storage, plasmonics, and catalytic applications.

Bottom–up and Top–down nanofabrication

Bottom-up approach is usually employed to design novel biomaterials. This method starts from the smallest particles which are "grown" into larger structures either by self-assembly or by reaction with/and incorporating other nanoparticles. The techniques commonly used are atomic layer deposition, sol–gel nanofabrication, molecular self-assembly, vapor phase deposition and DNA-scaffolding.

Shani *et al*. [70] used octahedral DNA frames to scale-up to superconducting 3D nanostructures. The authors started with self-assembling DNA superlattice which was transformed into a solid structure by coating it with SiO_2 and Nb. The final structure was used as Josephson junctions and amplifiers for quantum information systems.

Top-down approach is less used because it involves the break-down of large particles into nanoparticles. For biological materials this implies irreversible damaging if classical methods are used. Thus, the techniques used for this approach should provide multi-directional patterning ≤ 100 nm. Lithographic techniques, such as optical, electron beam, nanoimprint, scanning probe, and block copolymer lithography are perfect techniques for this approach [71]. Other disadvantages of this technique involve the need of an extremely clean, low-particulate working environment and the use of toxic chemicals in photolithography [72].

4.2. Examples of DNA-based materials

DNA/phytoextracts/bio-dyes

Conjugation of DNA with antioxidants could prevent its oxidative damage, and, therefore, preserving its structure. The most common antioxidants are of plant origin. The conjugation of DNA with phytoextracts gives rise to novel biomaterials with interesting optical features. Thus, the research of Rau *et al*., from Politehnica University of Bucharest, Romania [16] developed DNA–bio-dyes films using the natural extracts of some spices: turmeric (*Curcuma longa*), paprika (*Capsicum annuum*),

black pepper (*Piper nigrum*), and curry leaves (*Murraya koenigii*) which are rich in antioxidant photo-active molecules such as curcumin, capsaicin, piperine, and karapinchamine A & chlorophyll *a*, respectively. These DNA-natural dye complexes present a large π electron conjugation, being an eco-friendly alternative for polluting synthetic dyes suitable for application in photonics. These photoresponsive natural materials were optically characterized by UV-Visible absorption and FT-IR spectroscopy. On the other hand, in the last decade, the spices were investigated for applications as sensitizers for the dye-sensitized solar cells [73] or in photonics and optoelectronics [66].

Moreover, the research team of Barbinta-Patrascu from University of Bucharest developed an ecological strategy to design novel materials based on DNA and the natural extract of *Paeonia officinalis*, a common Romanian ornamental plant [74]; these DNA-phytoextract complexes showed a great potential for applications in photonics.

DNA-amphiphiles complexes

Nizioł and co-workers highlighted the importance of DNA as an ‘‘optical material” in the field of organic electronics and photonics. In this regard, the complexes of DNA with amphiphilic cationic surfactants constitute a new class of optical materials. Thus, in the last decade, the DNA-CTMA complexes are increasingly used as host material for optically active dyes [75] such as Disperse Red [76], Rhodamine 610 [17, 18], or Nile Blue [77].

DNA-CTMA complexes were used in development of photonic devices, because such structures are endowed with self-assembly properties and can generate nanosized supramolecular assemblies [59].

Dyes such as coumarin 480 (Cm 480) and 4-[4-(dimethylamino)styryl]-1-docosyl-pyridinium bromide were used in DNA-CTMA as a supporting matrix for optoelectronics applications (sensors, photovoltaics cells, LEDs, lasers, and solid-state lighting).

Mamangun *et al*. [78] compared different DNA sources and molecular weights with the energy transfer between dyes. The results showed that the DNA films prepared from onions were 2-5% more efficient in transfering the energy than the films obtained from salmon DNA.

Mariyappan *et al*. [79] used 3D printing method to develop DNA flat and curved films containing water-soluble/thermochromatic dyes and di/trivalent ions and CTMA-modified DNA films embedded with organic light-emitting molecules (OLEM). These materials presented improved optoelectric characteristics as compared to pristine DNA, and could be used to construct energy harvesting systems and chemo-bio-sensors.

DNA-lipid complexes (also named *lipoplexes*) are used not only in biomedicine as gene delivery systems [80], but also in optoelectronics to develop light-harvesting antenna [81]. Okahata & Kawasaki described a simple preparation of DNA-aligned cast films from DNA-lipid complexes, with conductive properties and also showing polarization of light [82].

One kind of amphiphiles are phospholipids that are biomolecules occurring in all biomembranes of living systems. Learning from nature, and inspiring from plant kingdom, the scientists designed self-assembled photonic systems mimicking natural light harvesting antennas. Zhou *et al*. [7] reviewed the advances of development of *biomimetic photoactive systems* based on DNA nanostructures with applications in energy production and biomedicine.

The phospholipids - the major components of biological membranes - were used to prepare artificial cells endowed with biomimetic membranes, the so-called *liposomes* or *lipid vesicles*. Liposomes are most commonly exploited to construct optical sensors, and also magnetic and electrochemical sensors [83]. On the other hand, phospholipid bilayers are typically transparent in visible light range and have interesting thermal and electronic properties [84]. Barbinta-Patrascu [3] designed, through a bottom-up approach, a material based on DNA and phospholipid biomimetic membranes labelled with a natural fluorofor, chlorophyll *a* (Chla), with great potential in biophotonics' applications. Chla is an optically active phytomolecule and is the key molecule in the photosynthesis process. It was already used as optical sensor to monitor the changes occurred at molecular level, in artificial lipid bilayers [85, 86].

Moreover, Rubio-Sánchez *et al*. highlighted in their paper [87] that the biomimetic membrane-anchored DNA nanostructures could be used as materials for artificial cell construction. These biohybrids could also be exploited in biosensing devices. These authors pointed out that the attachment of the DNA nanostructures to lipid bilayers gives rise to an amphiphilic character which can be used to develop complex supramolecular structures through self-assembly.

DNA-polysaccharides complexes

The conjugation of nucleic acids and carbohydrates have been explored in development of smart devices for sensing multiple analytes with greater precision, sensitivity, and selectivity.

Kantak and Shende [88] reviewed the importance of Nucleic acid-based Carbohydrate Sensors (NAbCSs) in the field of nanomedicine, gas sensing, and gene therapy.

Postnova *et al*. [89] used native ds-DNA - chitosan complex (the so-called *chitoplex*) to form a translucent homogeneous hydrogel with extraordinary mechanical strength and elasticity even after rehydration from aerogel form. Other authors [90] used DNA-chitosan hydrogels as catalyst for the reduction of nitroaromatics.

DNA-CNTs/fullerenes hybrids

Carbon nanotubes (CNTs) surfaces allow electronic π–π interactions with the nucleobases of nucleic acids, which is the key for developing a DNA/electrode interface [91]. Wrapping of multiwalled carbon nanotubes

(MWCNTs) with DNA molecules have received considerable attention in applications in materials engineering, energy harvesting, biotechnology, and detection systems [14].

Bilge and co-workers [92] developed a DNA biosensor for direct electrochemical monitoring of palbociclib-DNA interaction. Palbociclib (PLB) is a drug used in the breast cancer therapy, its binding site being the major groove of DNA. Interestingly, to fabricate this nanobiosensor, the researchers were used NH_2-MWCNT "green" synthesized from human hair wastes by the hydrothermal carbonization method. The human hair is a valuable biomaterial rich in pyridinic nitrogen which increases the electrical conductivity, providing a significant benefit in biosensor and various optoelectronics applications [92, 93]. The DNA biosensor developed by the research team of Bilge [92] was applied to the quantification of PLB in tablet dosage forms.

Mainly, DNA-CNTs biohybrids have been used as label-free rapid electrochemical sensors. DNA-wrapped CNT aerogels [94] have been prepared *via* a single-step freeze-drying and used as platform for biomimetic devices. The most important behavior of these aerogels is the resistive switching as function of the applied voltage, having a similar behavior as a volatile memristor.

Another type of DNA-CNT nanosensor is that developed by the Babenka *et al*. [95] and Cai *et al*. [96] who manufactured a low-cost, flexible CNT network-based DNA sensor using diferent printing techniques (e.g., ink-jet printing). The detection mechanism is based on the immobilizing of ssDNA on the CNT surface and the recognition of their complementary DNA target *via* electrochemical means.

The are only a few studies focusing on the complexes DNA-fullerenes. Ramesan and co-workers [97] used the mutual assisted assembly of the fullerenes and DNA to form an ultrathin, crystalline nanosheet. Wang *et al*. [98] used the complex fullerene-DNA/hyaluronic acid as nanovehicles to control the drug delivery and achieve more intelligent and powerful targeting of HepG2 cells.

DNA-based photonic-devices

Gheorghe *et al*. [15, 17, 99] pointed out that DNA is a new "green" photonic material which has been intensively applied in organic photonics and organic opto-electronics, in the last years. For the moment, the DNA-based bio-photonic materials are biodegradable.

In the field of photonic devices, the DNA-based structures are becoming important, especially as fiber optic sensors. Besides being just a genetic material, DNA is able to control light-matter interactions and thus, controlling laser emission [84]. In integrated photonics, the role of DNA is that of matrix for active molecules, particularly in LEDs and lasers [100, 101].

However, the incorporation of DNA in photonic devices has rise some concerns. The most important one is the phototoxicity, which occurs either when the light is focused on a small area, the optical power being too high or when the UV light is operating the device. Phototoxicity acts in two ways: by phototermal damage, which causes DNA denaturation (particularly above 67 °C) or by photochemical generation of free radicals that damage the integrity of the cell [84].

The most important argument for switching to DNA-based polymers in photonics and electronics is the fact that DNA is renewable. Secondly, it could be obtained from waste from the food industry and third, it is biodegradable [102].

Dagar and Brown [103] reported for the first time, the fabrication of organic solar cells with electron transport layers consisting of a ZnO compact layer covered by a thin DNA layer, with performance at low light levels and working very efficiently under indoor artificial light illumination.

DNA-porphyrins

Complexes of DNA-porphyrins are usually used as photodynamic therapy agents, due to the chemical and physical properties of the porphyrin compound. Particularly ruthenium-porphyrin based species have emerged as anti-cancer drugs. Oliveira *et al*. [104] have studied by spectroscopic means, the interaction with DNA of two tetracationic porphyrins peripherally coordinated to $[Ru(bpy)2Cl]^+$ units. The results showed that the Ru-porphyrins promote an efficient DNA photocleavage upon irradiation with white light. Co-porphyrins in combination with DNA have been investigated as electrochemical marker to detect specific DNA sequences [105]. Modifying the DNA-Co-porphyrin with Au and Ag gives the same performance in terms of sensitivity and selectivity for oligonucleotide sensing. For the detection of DNA G-quadruplexes, Dobrovodsky *et al*. [106] have used Cu-porphyrin and oligodeoxynucleotides. However, it is difficult to distinguish between double-stranded and single-stranded DNA because the Cu-porphyrin binds to both forms. Better results are obtained with Au-porphyrin, meso-5,10,15,20-tetrakis[4-(N-methyl-pyridinium-2-yl)phenyl]porphyrinate gold(III) [107]. This gold structure shows a great affinity towards antiviral genome.

Kumari *et al*. reported, for the first time [108], a self-assembling approach to fabricate DNA-porphyrin hybrid nanostructures as antimicrobial agent through light-induced ROS (reactive oxygen species) generation. These DNA-porphyrin hybrid nanonetworks showed no or negligible dark toxicity against mammalian cells, but they displayed high light-killing activity against Gram-negative (*Escherichia coli BL-21*) and Gram-positive (*Staphylococcus aureus*) bacteria.

DNA-Biolasers

Over the past decade, the concept of biolasers (biological lasers or biointegrated lasers) has become an emerging technology. "*Biolaser is a new type of laser that incorporates biological materials as part of its gain medium or cavity, and/or is embedded/integrated within biological materials*" [84].

In their recent review, Zhang *et al*. [84] pointed out that DNA is much more than a simple genetic material, the DNA nanotechnology being exploited for controlling laser emission.

Recently, Zhang *et al.* [109] developed a self-switchable laser by exploiting the biointerface between label-free DNA molecules and a dye-doped liquid crystal matrix.

DNA-based plasmonic nanostructures

In a recent review paper, Bui *et al*. [65] pointed out that plasmonically active DNA-based materials could be used in biosensors development, by exploiting the optical properties (i.e., color changes) of the metallic nanoparticles as a result of their aggregation state in the presence of DNA. DNA molecule acts as a nanoscaffold for arranging plasmonic nanoparticles that give rise to plasmonically active materials.

Liu *et al*. [110] recently reported the fabrication of a plasmonic nanostructure based on two gold-nanorods, co-assembled onto a DNA origami template. This plasmonic sensor showed a high sensitivity for nucleic acids, adenosines, chiral tyrosinamides or specific receptors expressed by tumor cells, which were correctly identified through pattern recognition and amplification elements.

Moreover, biomimetic metallic nanostructures (biometal NPs) "green" synthesized by aqueous plant extracts received a great attention in the last years, in the fields of biomedical engineering and catalysis, clean energy, electronic devices, and data storage, due to their unique optical, catalytic, and electrical properties [111]. Thus, Barbinta-Patrascu [4] used a "green" nano-approach for eco-synthesis of silver nanoparticles from *Cornus mas* L. fruits aqueous extract with good binding capacity to DNA molecules, and potential applications in biomedical field, in biophotonics, and in biosensing.

DNA-based sensors

Supramolecular systems based on DNA have been used as label-free intelligent sensors for the detection of streptomycin in foods by SERS means [112]. By embedding Au nanorods into the DNA hydrogel, the detection limit reaches 4.85×10^{-3} nM. In order to improve the electron transfer between the analyte and the sensor, Qian *et al.* [113] used MWCNTs combined with hydroxylatopillar[5]arene to develop a supramolecular electrochemical complementary DNA sensor. By stabilizing the supramolecular structure of MWCNT-hydroxylatopillar[5]arene with AuNPs, it can be used to subsequently modify the electrode with Rhodamine B-labeled DNA probes. These developed probes are used to electrochemically detect HBV DNA ultrasensitively, the detection range varying between 0.1 fmol/L to 0.1 nmol/L, the detection limit being 0.19 fmol/L.

Besides Pb^{2+} [114] quantification, the microorganisms *Helicobacter pylori* [115], *Orientia tsutsugamushi* [116], and *Staphylococcus aureus* [117] were also detected by DNA-based sensors.

DNA electronic nose

Mimicking the biological olfactory system, the research group of Gaggiotti [118] developed an *optoelectronic nose* using hairpin DNA and virtually screened peptides with improved selectivity as sensing materials, and surface plasmon resonance imaging as detection system, with improved performances for the analysis of volatile organic compounds. Such optoelectronic devices are useful in various applications such as: air quality monitoring, detection of olfactory pollution, detection of fires and gas leak, food quality control, food safety, cosmetics as well as diagnosis of certain diseases through the detection of volatile markers in breath.

5. Conclusions

This review presented in an original manner, a state-of-the art of DNA applications in photonics and optoelectronics, in the last twelve years.

In the current context of the environmental pollution and of the reduction of global energy resources, there is a growing tendency of using natural raws for development of advanced materials with applications in various fields. Thus, i) the valorization of natural sources (such as food wastes, human hair wastes, algae, vegetal raws, etc.) for DNA extraction, ii) the application of *Green Chemistry* principles, and also iii) the 3D printing method for development of multifunctional DNA-based materials, became new trends in photonics and optoelectronics.

The main "green" strategies used to design and fabricate DNA-based materials/devices are bottom-up self-assembling processes. The unique structure of DNA makes it also suitable for drug delivery applications.

Moreover, biomimetism/bioinspiration is another trend in modern photonics/optoelectronics, allowing the development of DNA supramolecular assemblies with unusual properties.

DNA is a wonderful natural molecule and a versatile biomaterial with a special structure allowing self-healing and self-assembling, giving rise to unusual DNA structures that have been exploited to design interesting materials with wide range of applications including biomedicine, nanotechnology, material science, photonics and optoelectronics.

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*Corresponding author: stefan.iordache@inoe.ro