DNA as scaffolding for nanophotonic structures

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**Abstract.** Deoxyribonucleic acid (DNA) chains can be engineered for diverse nanophotonics applications by the insertion of molecular groups in different spatial configurations. DNA chains can be assembled into wire-like structures, origami structures, photonic crystal-like assemblies, liquid-crystal phases, and thin films. These structures can be made to serve as scaffolds for the organization of various organic molecules and nanoparticles. The properties of nanostructures can be modified by the use of DNA and DNA modified by the surfactants. © 2012 Society of Photo-Optical Instrumentation Engineers (SPIE). [DOI: 10.1117/1.JNP.6.064505]

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1 **Introduction**

The deoxyribonucleic acid (DNA) molecule is well known as a blueprint of life, amazingly rich in information content and very robust. However, its unique structural features and powerful recognition capabilities can also be of interest for assembling artificial structures for a variety of applications. This review mostly concentrates on the perspectives of use of DNA in nanophotonics: the technology of generating and manipulating light quanta on the nanoscale. A DNA helix is itself a nanoobject that can be manipulated in various ways, but it can also be treated as a versatile molecular scaffold for building nanoscale devices from the bottom up. DNA’s unique properties are intensively studied from different points of view: as a molecular wire, as a drug delivery system, a ladder for ordered arrangements of various nanostructures, a spacer to control distances between nanoobjects, etc.

In this article, we review the use of DNA in the material science approaches, mainly through DNA’s ability to provide order to molecules of dyes and/or nanoparticles (NPs) that are attached or interacting with the DNA helices. We describe the potential for a single DNA helix to behave as a robust building block of nanoscale systems, and we also discuss the ordering of DNA and light-responsive elements on a macroscopic scale, where liquid crystalline properties of DNA play an important role. Finally, we present how a modification of DNA, soluble in organic solvents, can be incorporated into optoelectronic and nanophotonic devices.

2 **DNA Helix—Ordering of Molecules and Nanoparticles for Applications in Nanophotonics**

2.1 **DNA as a Molecular Wire**

The elongated shape of the DNA molecule suggests that it is a wire capable of transmitting signals along its length. On the one hand, DNA is a rather poor electrical conductor, and its application in molecular electronics will be difficult. On the other hand, DNA possesses well-defined binding sites, where various molecules, from small dyes to large and complex...
macromolecules, can be precisely located. Biochemists and genetic engineers have developed advanced techniques for the manipulation of DNA chains and multiple chemistry procedures of well-controlled DNA labeling. These robust possibilities of organization of organic markers on the DNA scaffold can be easily adapted for organization of photoactive molecules. Su et al. pointed out some of the latest achievements in the field of energy transfer via dyes organized on single DNA strands and on double-stranded DNA. Unidirectional energy transfer over the length of 40 bp (~13 nm) on one DNA chain was observed with the efficiency of 20%. In order to achieve better efficiency, a template DNA strand, biotinylated at one end and labeled with injector (Rhodamine G) at the other end, was immobilized on a streptavidin-coated surface. Short complementary strands were doped with emitting chromophores (tetramethylrhodamine, ATTO dyes, etc.) and the emitting dye (Cy5.5) and hybridized to the template. This five-color system demonstrated an energy transfer efficiency of 85%. Apart from the hybridized dye-labeled short oligonucleotides, DNA stained with intercalators of groove-binders exhibits efficient energy transfer and can be considered to be photonic molecular wires. Such structures were expanded to two and three dimensions [see Fig. 1(a)].

Shionoya, Muller, and others introduced a concept of combining DNA and functional metal complexes into one molecular wire. More recently, single-molecule break junctions of metallo-DNA with single wall carbon nanotubes as point contacts were applied for charge transport. Incorporation of chelating agents enables the switching of conductive properties and opens up the possibility to build single-molecule devices, such as molecular switches or sensors.

DNA electric properties are still a question being explored and debated, as various reports describe DNA as insulator, semiconductor, or conductor. It has been shown in the paper by Pawlicka et al. that the conductivity can be improved by adding the plasticizer such as poly(3,4-ethylenedioxythiophene)/poly(styrenesulfonate) (PEDOT:PSS), poly(o-ethoxyaniline) (POEA) or electrochromic dye Prussian Blue (PB). However, the DNA structure itself can serve as a reproducible scaffold where photoactive molecules can be positioned with nanometric precision.

### 2.2 DNA as a Template for Synthesis of Nanoparticles

The predictable order exhibited by DNA helices and their chemical stability make them attractive as agents used for controlling the synthesis of various NPs. Both single-strand DNA (ssDNA) and double-strand DNA (dsDNA) have been proved to be effective templates for the synthesis and ordering of nanometric objects. Noble metal (plasmonic) NPs are an example of a class of nanoobjects that can be successfully grown onto DNA templates. Standard synthesis of such

![Fig. 1](https://www.spiedigitallibrary.org/journals/Journal-of-Nanophotonics on 01 Dec 2019)

*Fig. 1* (a) Schematic energy transfer (ET) in a tetrahedron DNA nanotag loaded with YOYO-1 intercalated dyes and covalently attached Cy3 acceptor dyes. (b) Pictures of emissive solutions of ss-DNA-encapsulated Ag nanoclusters created in: 5'-CCCTTTAACC-3' (blue), 5'-CCCTTTAACC-3' (green), 5'-CCCTTTAATCC-3' (yellow), 5'-CCCTTTACC-3' (red). (c) Schematic diagram of logic operations based on HP26-tuned fluorescent Ag nanoclusters. (Right) Corresponding symbols of logic gates. K⁺ and H⁺ serve as two inputs to trigger the allosterism of HP26 and modulate the fluorescence output.

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NPs involves the reduction of metal ions with reducing agents (e.g., citrate or borohydride). As an example of the advantage of performing such reactions in the presence of DNA, reducing Ag\(^+\) ions in the presence of additional cytosine bases of short ssDNA was shown to lead to formation of NIR-emitting, water-soluble Ag nanoclusters.\(^7\) These few atom clusters possess high photostability, very good dispersibility in water, and very high single-molecule emission rates with strong antibunching. All these features yield great promise for the application of DNA-capped nanoclusters, introduced both in vitro and in vivo, in single-molecule imaging. The influence of the sequence, length, and arrangement of ssDNA on nanoparticle properties has been investigated.\(^8,17\) Modifications of the sequence resulted in the tuning of excitation and emission spectra of nanocrystals [Fig. 1(b)], with the photostability of DNA-NP complexes increasing with the increase of the emission wavelength. Moreover, when a DNA sequence producing a secondary intramolecular i-motif structure was introduced in Ag nanocluster formation, strong red luminescence was observed at pH 6, giving way to green luminescence at pH 8 to 9.\(^17\) Multiple logic operations can also be performed, evoked by changes in the concentrations of K\(^+\) and H\(^+\) ions, which in turn modify the structure of DNA.\(^9\)

The presence of DNA molecules with various sequences during the nanoparticle synthesis determines their morphology. Gold NPs synthesized with the addition of poly(A), poly(C), and poly(G) adopt a flower like shape, whereas poly(T) results in a spherical shape.\(^18\) Synthesis in the presence of short ssDNA oligonucleotides (1 to 12 nucleotides) was also performed,\(^19\) and the homogeneity of nanoparticle size and shape (as well as stability in salt solutions) was found to increase with the length of the oligonucleotides. These DNA-mediated changes of nanoparticle morphology allow one to easily modify the optical properties, as well as influence the cellular uptake and biocompatibility of gold NPs. It appears that tuning of optical properties in nanoclusters using DNA oligonucleotides may be a simple and efficient technique compared to adjusting the optical properties of fluorescent dyes or of larger NPs. An interesting feature of DNA-tuned Ag nanoclusters is that they can also work as molecular logic gates exhibiting multichannel fluorescence output [Fig. 1(c)].

Interestingly, the chiral nature of the DNA helix may give rise to induced optical activity of the NPs synthesized on DNA templates. Chiral silver and gold nanoclusters\(^20\) and NPs\(^21\) have been prepared. The chirality of metal nanoclusters originates mainly from a high probability of packing Ag and Au atoms into low-symmetry chiral configurations, as was confirmed with theoretical computations [Fig. 2(a)].\(^22,23\) On the other hand, larger NPs exhibit circular dichroism, most probably due to their chiral organization on DNA helix [Fig. 2(b)]. Several review articles describing DNA and other chiral molecules inducing chirality in metal NPs have been published recently.\(^23-27\) They demonstrate the increasingly growing interest in optically active nanostructures and their potential applications in chemical synthesis, biosensing, drug delivery, production of optical components, chiral negative index materials, and nanophotonic devices.

DNA has the ability to control the growth and morphology of quantum dots (QDs) as well. CdTe nanocrystals were synthesized in a one-pot procedure with a bifunctional DNA strand.\(^29\) One part of DNA, with a phosphorothioate end, served as a ligand binding to the QD surface, whereas the other part of DNA was a molecular recognition element. Differences in structural and optical properties of CdTe QD capped with A, G, T, or C oligonucleotides of various number of nucleotides were investigated.\(^30\) Stable and highly luminescent DNA-labeled QDs were then assembled in a well-controlled manner. QD complexes of up to three different sizes of nanocrystals were constructed. Their optical properties were dependent on the energy transfer between incorporated QDs and could be controlled by external stimuli (e.g., pH). Syntheses of DNA-capped cadmium and lead chalcogenide-based nanocrystals were also presented in the literature,\(^31\) and a strong influence of DNA on morphology and optical properties of the products was confirmed.

### 2.3 Ordering and Assembling of Gold NPs with DNA Strands

Thiol-functionalized DNA can be easily attached to gold surfaces and thus made to serve as a perfect link between gold NPs and nanoclusters, with full reversibility of binding of linking
chains. High specificity of the binding comes from the hybridization of two complementary ssDNAs on two different NPs or hybridization of these strands through a duplex-bridge DNA. Numerous review papers and monographs have been recently published on this broad subject. The linking can lead to the formation of various size plasmonic molecules (dimers, trimers, etc.), with NPs of the same or different sizes; furthermore, one can obtain regularly spaced plasmonic nanoparticle chains in polymerization-like reactions. Finally, three-dimensional ordered arrays of gold NPs can be created with advanced procedures of the synthesis of well-defined plasmonic super-crystals, which have been recently described in Macfarlane et al. Among various geometries, chiral species were demonstrated, with gold NPs of four sizes, assembled with DNA linkers into chiral pyramids [Fig. 2(c)]. Many of the structures presented can successfully incorporate various lengths of the DNA strands and sizes of metal NPs, as well as inorganic nanocrystals. Changes of the geometry and relative positions of these building blocks have been shown to induce modifications of the optical properties. As a result, multifunctional, tunable and environmentally-sensitive nanostructures can be obtained. They are finding various interesting applications in nanophotonic systems (e.g., plasmon-based nanolenses).

Not only can the structural properties of DNA be exploited in nanotechnology, but also the DNA synthesis techniques, developed by biologists and genetic engineers, may be adopted there...
as well. The polymerase chain reaction (PCR) is the standard DNA synthesis procedure, which offers exponential amplification of DNA strands identical with the template one. PCR, with DNA primers attached to gold and magnetic NPs, was performed, and nanoparticle aggregates were obtained. Then, PCR was applied to computer-controlled assembling of DNA-gold NP conjugates, with the size of the obtained nanostructures dependent on the number of PCR cycles. Interestingly, when the procedure is performed in a solution with a distribution of NP sizes, optically active dimers, trimers, tetramers, and pentamers are obtained. The product mixture is not racemic; it exhibits a strong positive circular dichroism signal whose origin has not yet been explained.

Diamond-like lattices composed of gold NPs and viral particles, woven together and held in place by strands of DNA, were created. Fabrication of such a structure—a distinctive mix of hard, metallic NPs and organic viral pieces known as capsids, linked by DNA—marks a big step toward creating a DNA-based photonic crystal that can manipulate visible light. The specific pieces of DNA were attached to gold NPs and viral particles, choosing the sequences and positioning them exactly to force the particles to arrange themselves into a crystal lattice that has a sodium thallide crystal structure. Such a self-assembled crystal lattice is potentially a central ingredient to a photonic crystal, which can be used for precise manipulation of light through blocking certain wavelengths of light while letting other colors pass. Applications for such...
DNA-base photonic crystal structures in fields like optical computing and telecommunications are foreseen, but manufacturing and durability remain serious challenges.

### 2.4 DNA-Origami as a Multifunctional Nanoscaffold

The unique property of complementary behavior of DNA bases became the basis of the controllable folding of DNA chains on a nanoscale. This method of manipulation of the chains, called DNA-origami, was introduced by Rothemund in 2006. It facilitates bending a single, long DNA strand with the aid of short staple strands with carefully designed sequences [Fig. 3(a)]. Since then, multiple periodically patterned two- and three-dimensional structures of DNA have been reported: twisted bundles of DNA, bent in a controllable way, three-dimensional spherical shells and nanoflasks, and three-dimensional monoliths and crosses made out of honeycomb lattices of DNA-origami [Fig. 3(b)]. Dietz and coworkers approached the automatic design and synthesis of DNA-origami structures by developing a computational tool (computer-aided engineering for DNA-origami: CanDo) for predicting the structure and providing information about the conditions under which DNA-origami objects can be expected to maintain their structure [Fig. 3(c)]. Recently, Zhao et al. presented a method to produce large area of DNA-origami by the superorigami or origami of origami method. Fabrication of large-area, mesoscopic arrays of gold NPs was made possible by depositing DNA-origami onto a lithographically patterned substrate, and the 5-nm gold NPs were then precisely bound to specific points on the DNA scaffold. The combination of top-down lithographies with bottom-up self-assembly was accomplished with DNA-origami nanotubes. Due to appropriate functionalization with thiols, they formed connections between gold patterns produced with electron beam lithography [Fig. 3(d)]. Recently, the thermal stability of DNA constructs was improved by photo-cross-linking with 8-methoxypsoralen, which brings DNA-scaffolds closer to optoelectronic and nanophotonic applications.

DNA-origami-based elements of nanodevices have already been presented [e.g., nanomechanical and tensegrity (tensional integrity) structures]. DNA-origami bundles were successfully used for controllable positioning of dye molecules in artificial light-harvesting antennas. DNA-scaffolds and DNA-nanoparticle assemblies find multiple applications in biodiagnostics and optical, electrochemical, or electrical biosensors. The combination of both of them may form sophisticated systems; e.g., a computational device with DNA nanomechanical self-assembled elements, which can divide a number by 3. In this system, various ssDNAs serve as an input and the sequence of gold NPs bound to specifically ordered sticky ends of the DNA sequence is an output signal.

Liu and coworkers examined the metallization of DNA-origami, as an enabling step toward the use of such DNA as templates for nanoelectronic circuits. DNA-origami makes it feasible to increase the complexity and flexibility needed for both the design and assembly of useful circuit templates. In addition, selective metallization of the DNA template is essential for circuit fabrication. Metallization of DNA-origami presents several challenges, including (1) the stability of the origami in the processes used for metallization, (2) the enhanced selectivity required to metallize small origami structures, (3) the increased difficulty of adhering small structures to the surface so that they will not be removed when subject to multiple metallization steps, and (4) the influence of excess staple strands present with the origami. The DNA templates were seeded with Ag and then plated with Au via an electroless deposition process. Both staple strand concentration and the concentration of ions in solution were found to have a significant impact. Selective continuous metal deposition was achieved, (height as small as 32 nm), and the shape of branched origami was also retained after metallization. These results represent important progress toward the realization of DNA-templated nanocircuits. A novel method for producing complex metallic nanostructures of programmable design was presented. DNA-origami templates, modified to have DNA binding sites with a uniquely coded sequence, were adsorbed onto silicon dioxide substrates. Gold NPs, functionalized with the cDNA sequence, were then attached. These seed NPs were later enlarged, and even fused, by electroless deposition of silver. Using this method, a variety of metallic structures, including rings, pairs of bars, and H shapes, was constructed.
3 DNA as a Matrix for Ordering of Molecules and NPs

3.1 DNA Liquid Crystals

The previous section described methods of ordering discrete DNA strands in two and three dimensions. A large-scale ordering of a high number of DNA chains is another opportunity for using DNA as a photonic material. In fact, the nature has already provided this behavior, and in aqueous solutions, DNA is known to exhibit liquid crystal (LC) ordering under certain conditions (lyotropic liquid crystallinity). LC phases of DNA were found in chromatin in cell nuclei, bacteria carrying Blue Script plasmids, sperm heads, and capsids of viruses. Depending on the temperature, DNA concentration and length, occurring counterions, and other polyions in the solution, DNA can be disorganized or can organize itself into a cholesteric, columnar hexagonal, or crystal phase. In the cholesteric phase, layers of DNA create a helical superstructure with interesting optical properties, such as selective reflection and transmission of circularly polarized light. DNA helices in the columnar hexagonal phase are packed parallel to one another, in a hexagonal structure. In both phases, various defects can occur, which can bend and deform DNA patterns. DNA-binding dyes follow the orientation of DNA in LC phases and can be perfect markers of DNA orientation. We have recently shown that, using a polarization-sensitive, two-photon fluorescence microscopy (ps-TPFM), we can map the exact position of dyes, and thus DNA, in three dimensions (Fig. 4). On the other hand, by controlling the ingredients of the functionalized DNA solution and external conditions, a specific LC phase can be obtained, with dyes ordered in a desirable manner. Not only standard DNA markers, but also photochromic dyes, were organized in such a system, proving the propensity to incorporate various functional molecules. LC phases are formed in sealed LC cells with appropriate conditions, but also in a simple system of drying droplets, where contact line pinning and radial forces produce a zigzag pattern [Fig. 4(a)]. This is an easy way of creating a millimeter-scale columnar phase of DNA and dye-doped DNA.

Fig. 4 (a) Polarized light microscopic (PLM) image of characteristic zigzag pattern at the perimeter of the dried DNA-dye solution drop and two-photon fluorescence intensity raster scan of the border region between two LC domains (white square in PLM image). Polar graphs (1 to 5) were measured in different points across the border line, and on that basis, the position of DNA in each domain was determined. (b) ps-TPFM scans at different depths of cholesteric DNA-dye phase and the y, z cross-section of the sample.
3.2 DNA Thin Films

DNA doped with the intercalating cyanine dyes were mixed with polyvinyl alcohol (PVA) and its alignment was studied. It has been proven that the DNA-PVA thin film is a highly anisotropic material that can be used, for example, for studies on structural properties for nucleic acids, but also its potential applications in photonics are envisaged. Thin layers of DNA/Polyaniline/Ru(bpy)$_3^{2+}$ complex and tris(8-hydroxyquinolinato)aluminum were combined in organic light-emitting diode (OLED) as the hole transport layer and the electron transport layer, respectively. Another approach is the exploitation of the DNA thin films for orientation of the LCs, such as 8CB and MBBA (see Fig. 5); the chirality of such systems has also been explored.

The mechanical properties of DNA-doped nanofiber hydrogels were shown in Shin et al. The elastic modulus of DNA is on the order of 0.3 to 1 GPa, similar to hard plastics, but at the same time, the molecule is extremely flexible, having the persistence length of 50 nm.

4 Modified DNA Soluble in Organic Solvents—the Polymer Matrix for Optoelectronics

DNA, as a biological material, is soluble in water. To introduce the solubility in organic solvents, substitution of sodium counter cations with cationic amphiphilic molecules is performed. Cationic lipids, hexadecyltrimethylammonium chloride (CTMA), and other surfactants are applied as ligands, binding to phosphate groups of DNA backbone. DNA-CTMA may also serve as a template for organizing dye molecules. Trapping of organic molecules between ligand molecules, or inside the DNA helix, was proposed in systems with fluorescent rhodamine G6, which then can be used for its lasing properties. Drying films of DNA-CTMA have self-organizing properties and very good processability for integration into large-area devices.

Interestingly, the helical structure of DNA is preserved and can serve as a template for chiral organization of dopants. Circular dichroism of films of DNA-CTMA doped with hemicyanine NLO dye, 4[(4-(dimethylamino)styryl)-1-docosylpyridinium (DMASDPB), was measured, and the full orientation structure along the chiral DNA double helices was confirmed. Chiral organization of molecules is an alternative for the polar organization required to produce second-harmonic generation (SHG). Dried films of DNA-CTMA, doped with the dye crystal violet, were SHG-active, without application of any external stimuli.

The DNA-CTMA complex has been proposed as an ideal material for optoelectronics due to its unique electromagnetic and optical properties, which are summarized in Table 1. Intrinsic nonlinear refractive index discourages to apply pure DNA as nonlinear optical material. However, DNA-CTMA has been applied as a cladding and host material in...
nonlinear optical devices. In organic field effect transistors, a thin layer of DNA-CTMA was introduced as gate dielectric, and in combination with quantum dots, it served as the efficient hole-transporting layer in light-emitting diodes and as an electron-blocking layer in OLEDs.

Moreover, DNA-CTMA doped with photochromic dyes, such as Disperse Red 1, provided a favorable matrix for dynamic holography and other nonlinear optical features such as, for example, third-harmonic generation. DNA with cationic lipids containing TEMPO radicals exhibits a reversible two-stage charge/discharge processes with promising application as an organic radical battery, whereas DNA loaded with the DMASDPB dye demonstrated improved photostability when compared to the PMMA matrix (to cite one example), which can be of importance for photonic applications (solid-state laser design). Amplified spontaneous emission was observed under nanosecond optical pumping, when the DNA-CTMA matrix was doped with rhodamine dye or a combination of organic nonlinear optical dye and photochromic polymer. Recently, dielectric tunability in DNA-CTMA film at microwave frequencies was shown, pointing out that the dominant polarization mechanism is ionic in nature and is caused by intentionally retaining excess ions in the DNA-CTMA precipitate during processing. Effective use of the DNA-CTMA matrix for fabrication of waveguides was presented in Zhou et al. The waveguides exhibited excellent single-mode output and high confinement of light; due to the sharp waveguide profile with very smooth surfaces and vertical sidewalls, low propagation losses were measured.

Any unfavorable characteristics of DNA-CTMA can be improved by formation of hybrid materials (e.g. DNA-ORMOCER nanocomposite). ORMOCER provides mechanical strength, and better hardness than the pure DNA-CTMA films and DNA serves as an efficient dye and nanoparticle-binding material.

5 Conclusion and Perspectives

With this review, we set out to convey that DNA is a very unique system, which has been studied for numerous applications, not only coming from its biological significance but also for material science applications. DNA's specific organization allows for the construction of photonic molecular systems, such as what is schematically shown in Fig. 6.
Light-harvesting systems can be arranged in a specific order, where the sequence of energy or charge transfer processes leads to optimum performance of channeling to create a new generation of photonic wires or conducting plasmonic devices (blue, green, and red balls and orange bars represent the photonic components that can serve as light-harvesting and energy-transfer materials). Other species can act as molecular sensing units by energy or electron accepting, where the light is converted to chemical potential (represented by the transformation of the substrate-triangles to the higher-energy product-stars).

Apart from the organization of photoactive species with single DNA helices, we would like to underline the ability of DNA to create LC phases, which can then be used as matrices for organization of other species, such as dyes or nanoobjects. The LC phases of DNA are undoubtedly of important biological importance (chromatin organization), but they also shall be explored for the positioning of the NPs and molecules in precisely chosen places. Then, the phase transition appearing in the LC DNA also can help the manipulation of the NPs, thus preparing a bistable system.

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