SWCNT Scaffold by DNA Spiral Nantenna

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Abstract—

Spiral DNA origami structure is synthesized to realize a spiral nanoscale antenna (Nantenna) for biomedical and engineering applications. Single strand DNA (ssDNA) origami structures utilize self-assembly techniques and short ssDNA staple strands to develop the desired spiral structures. We synthesized a circular (dimension) origami template, which can serve as a scaffold foundation for single walled carbon nanotubes (SWCNT). This paper will discuss the methods and protocol to develop DNA origami structure. We further discuss conductivity and present a detailed mathematical model to calculate the conductivity of Carbon Nanotube (CNT) based Nantenna.

I. INTRODUCTION

The last decade has seen an increase use of nanotechnology in biomedical, medicine and engineering applications [1, 2]. CNT and nanowires based technology has also been used in radio-frequency, microwave, power harvesting and bioelectronics [3-5]. Nanoparticles and nanostructures based sensors for improved selectivity and enhanced biomedical sensing is also thriving research area [6], and numerous research groups [6-9] have investigated the applications of nanoparticles in biomedical engineering and medicine. A challenge in this area is to improve the selectivity or efficiency of nanostructures by changing the geometry or altering the physical, chemical or electrical properties of nanomaterials and nanostructures.

Self-assembling 3-D DNA structures combined with CNT based Nantennas have the potential to revolutionize nano-electronics. Bottom-up growth of self-assembled technology has enabled researchers to build nanoscale structures with sound electrical and thermal properties [10, 11]. Advancements in nanotechnology and the synthesis of scaffold based nanostructures and nanoscale devices have moved toward a cost-effectively future [12]. One of the most interesting developments in nanotechnology is the use of biopolymers, DNA, for the synthesis of nanostructures. In this paper, we present the technique to develop DNA based nanostructure which

leads to the synthesis of nanowires and Nantennas. Electrical transport of such biological Nantennas can be improved by using conductive nanoparticles coating on DNA [13] or by attaching nanowire to DNA structures [14]. Though several DNA origami structures have been reported [15-17], very few applications are proposed due to the poor conductivity of DNA. The focus of our research is to fabricate a DNA scaffold based Nantennas. A DNA based spiral structure has been synthesized to serve as a template for CNT based Nantenna. The angle of curvature of spiral CNT is achieved by the techniques presented in [12].

DNA origami is the folding of ssDNA creating nanoscale shapes. First proposed by Nadrian Seeman [18] and later developed by Paul Rothemund [19], the proposed technique exploits the ssDNA binding characteristics and artificially synthesized small strands to form different nanoscale shapes. Many shapes have been designed and synthesized by using Rothemund's method; for example, spiral, triangular, smileys face, or bowtie. Rothemunds methodology involves heating and then cooling a long DNA virus phage and staple strand in a buffer solution. During the cooling phase, the short staple strands manipulate the shape of the longer single strand by pulling it into the desired form [12]. Outcome shape could be analyzed using Atomic Force Microscopes (AFM).

The real challenge in exploiting DNA-based structures, remains to be the fabrication of nanoscale devices with good electrical properties and complex geometries. Poor electrical characters of DNA prevents its use in nano-electronics. Variety of methods including doping, chemical modification, coating with conductive polymers, nanofiber, deposition of nanoparticles [20-23] have been proposed to enhance the electrical conductivity of DNA molecule. Carbon based structures such as graphene and nanotubes (CNT) have received lot of attention due to their excellent structural, mechanical and controllable electrical properties. Depending upon the arrangement of carbon atoms in the honeycomb structure, CNT bandgap varies to be that of semiconductors, or metals with nearly ballistic conduction [24]. DNA wrapped CNT was first reported in 2003 [25]. However, the arrangement of individual SWNTs into complex nanoscale geometries remained a challenge. An

interesting approach to bind nanotubes to a DNA origami structure was reported by Erik Winfree and coworkers [26]. The group demonstrated that DNA origami can be used to introduce two-dimensional geometry to the self-assembly of SWNT structures. They further showed that DNA constructs can be transferred from solution to SiO without losing electrical characteristics of SWNT.

In the literature, a variety of methods have been proposed to bind ssDNA molecule to SWNT, this paper does not specifically address the methods of synthesis and characterization of nanotubes. We focus on the development of spiral DNA origami structure and theoretical model of CNT based spiral structure. This paper is organized as follows: Section 2 briefly describes the design procedure and protocol for development of the origami structure. Section 3 discusses Graphene and CNT structure. Conductivity of conformal and spiral nanostructure is also discussed in section 4.

II. DNA ORIGAMI

Building a DNA origami structure starts by designing the desired shape in Cadnano software [27]. Cadnano makes the sequence designing procedure by allowing users to create junctions, called crossovers, between different DNA strands or at different locations of a same DNA strand. 2D and complex 3D origami structures can be formed by creating crossover between multiple layers of long ssDNA.

Creating an origami structure with nanoscale curvature's radius can be challenging since the software only consider the mechanical forces and torques due to the bending of DNA structures. The folding geometry of scaffold can be predicted by finite element based software, CANDO. It is worth mentioning that although finite element based predictive modeling software are extremely useful, most softwares have their limitations. While predicting the geometry of nanoscale structure, softwares are unable to predict the effect of Van der Waal's forces as well as Casimir forces. Intermolecular forces can have significant effect on the nanoscale designs especially if flexible ssDNA is used to design the nanoscale structure [28]. In complex structures, staples undergo multiple crossovers to connect several double helical domains. In addition to intermolecular forces the stiffness of such DNA origami structure also varies as more strands are bundled together to form complex structure [28].

A. Creating Spiral Structure

Nanoscale spiral structures are of interest due to their application in electrical, electronics and biomedical engineering [29, 30]. The focus of our research is to

develop Nantennas using configurable DNA structure as a template. Although chemical and electrical characteristics of DNA is limited in functionality, it can be used as scaffold structure in order to form functional material [31]. Spiral antenna, with infinite numbers of turns and optimal angles, possesses frequency independent radiation and impedance pattern. In practice, spiral antenna radiates from a region where the circumference of the spiral is equal to its wavelength, thus the spiral antenna frequency independent behavior is determined by its inner and outer radius of the spirals. The proposed 120(nm)×120(nm) spiral structure [figure 1] is designed in CADNano.

Fig. 1: Spiral DNA origami object design in CADNano

B. DNA Origami Protocol

DNA structures were synthesized using method proposed by Paul Rothmund [19]. Long ssDNA (M13MP18) is used with staples designed in cadnano. T

Fig. 2: DNA origami protocol

The DNA origami technique is a well understood and proven method to manipulate long ssDNA in order to form desired shapes.

The protocol is based on the DNA origami technique presented by [32] Shih et al. “Self-assembly of DNA into nanoscale three-dimensional shapes.”, Nature, 459, 414-418, (2009). The protocol, which is adopted for DNA origami purposes, is illustrated in the figure 2. Above protocol illustrate that 119ul of phage is diluted with 381ul distilled water to create 500ul solution. The origamis can be self-assembled by mixing 10ul of diluted M13mp18 virus phage with 10ul of diluted DNA scaffold. The solution is mixed with MgCl₂, Tris-HCl, EDTA and NaCl, buffer solution. The solution is cooled down slowly in a PCR thermocycler (Polymerase Chain Reaction) and origami shapes are imaged (Figure 3) using AFM.

Figure 3: DNA origami image

III. CNT REVIEW:

Carbon’s valance electrons can bond with other Carbon atoms covalently. Carbon has four valance electrons. Sharing these electrons in different ways allows carbon atom to form different allotropes with varying electrical properties.

Carbon has ability to create hybrid orbitals, bond across orbitals. Nanotubes are composed of sp^2 hybridization i.e. the bonding of one s-orbital with two p-orbital. Three of four valance electrons are bonded between s and p orbitals. The free electron, called π – electron give CNT its exceptional electrical properties and stimulated a surge of interest in the applications of CNT in engineering [33-36]. Despite recent advancement in nanoscale fabrication and reported success in using DNA as a template to guide the assembly [12, 37], designing CNT based nanoscale spiral structures remains a challenge. In this section, we will analyze CNT conductivity and further present the mathematical model of a spiral CNT base Nantenna.

Surface of CNT has a lattice honeycomb structure, which is exactly same as graphene. Conceptually, cutting CNT along the length and spreading it would result will be monolayer graphene sheet. Chiral vector (figure 4),

shows the circumference of CNT, when monolayer graphene sheet is spread.

Figure 4- $\vec{C}$ vector demonstration;

Since the foundation of CNT is Graphene, a reasonable approximation is to use graphene’s conductivity as CNT’s conductivity. Graphene, a mono-layered honeycomb form of Carbon atoms, has a two-dimensional structure with two sub-lattices (figure 5). Sublattices are separated with bullet points and circles.

Figure 5- Graphene honeycom lattice structure.

The unit cell is a rhombus shape with one atom from each sublattices. Unit cell is shown in figure 5

Figure 6- Graphene Unit Cell

As it is shown in (figure 6), a unit cell can be defined with two vectors, a_1 and a_2 :

$$\vec{a}_1 = \left(\frac{\sqrt{3}}{2}a_0, \frac{a_0}{2}\right) \quad (\text{III-1})$$

$$\vec{a}_2 = \left(\frac{\sqrt{3}}{2}a_0, -\frac{a_0}{2}\right) \quad (\text{III-2})$$

Where:

$$|a_1| = |a_2| = a_0 = \sqrt{3}b \quad (\text{III-3})$$

where “ b ” is the interatomic distance in graphene which is 0.142 (nm). We can show the chiral vector ($\vec{C}$) by using unit cell's basic vectors as:

$$\vec{C} = n\vec{a}_1 + m\vec{a}_2 \quad (\text{III-4})$$

Different n and m causes different edge types in the CNT's end, containing zigzag, armchair and chiral CNTs:

$$\begin{cases} \text{zigzag} & m = 0 \\ \text{armchair} & m = n \\ \text{chiral} & O.W. \end{cases} \quad (\text{III-5})$$

Also, m and n show whether the SWCNT is metallic or semiconductor. If $(n-m)/3$ is an integer value than CNT is metallic (highly conductive), otherwise it is semiconductor. Current density in metallic CNT can reach 4×10^9 A/cm² [38], which places CNTs among highly conductive materials. In contrast to one third of metallic zigzag CNTs, all armchair CNTs are metallic. In general approximately one third of CNTs are metallic [39].

Circumference of CNT can be found by:

$$|c| = \sqrt{|na_1|^2 + |ma_2|^2 + 2|na_1ma_2|\cos\theta} \quad (\text{III-6})$$

Where:

$$\theta = 60^\circ \quad (\text{III-7})$$

$$|c| = \sqrt{|na_1|^2 + |ma_2|^2 + |na_1ma_2|} \quad (\text{III-8})$$

From equation (II-3):

$$|c| = a_0\sqrt{n^2 + m^2 + nm} \quad (\text{III-9})$$

So radius of CNT is:

$$r_{CNT} = \frac{|c|}{2\pi} = \frac{a_0}{2\pi}\sqrt{n^2 + m^2 + nm} \quad (\text{III-10})$$

IV. CNT CONDUCTIVITY:

Traditionally thermal and electronic conductivity is due to diffusive transportation of electrons in random directions to reach its destination. The sum of these movements correspond to a displacement vector. However, unlike at large scale, where electrons move in random direction, ballistic electrons transport at nanoscale in a straight path, with a good approximation [40, 41].

In other word, if we consider the common conductivity equation,

$$(\sigma = \left(\rho \frac{L}{A}\right)^{-1}) \quad (\text{IV-0})$$

since “ L ” tends to be zero at nanoscale, the conductivity would become infinity which is against the experimental results. Thus, to find the conductivity, we analyze the changes in the electrons' energy (electrons movements). Boltzmann transport equation (IV-1) is used for this purpose:

$$\frac{\partial f}{\partial t} + eE_z \frac{\partial f}{\partial p_z} + v_z \frac{\partial f}{\partial z} = \vartheta[f_0(p) - f] \quad (\text{IV-1})$$

Where “ e ” is the electron charge, “ E_z ” is the longitudinal component of the electric field at the nanotube surface, “ v_z ” is the electron's velocity, “ ϑ ” is the relaxation frequency and “ $f_0(p)$ ” is the Fermi-Dirac distribution equation which is a function of the electron's momentum. “ f ” is the electron's distribution function which can be shown as:

$$f = f(p, z, t) = f_0(p) + Re(\delta f e^{j\omega t}) \quad (\text{IV-2})$$

The Fermi-Dirac distribution relation can be written as:

$$f_0(p) = \left(1 + e^{\frac{E(p) - E_F}{k_B T}}\right)^{-1} \quad (\text{IV-3})$$

Where “ $E(p)$ ” is the energy-momentum relation; “ E_F ” is the Fermi energy; “ k_B ” is the Boltzmann constant and “ T ” is the nanotube's absolute temperature. The Fermi energy in graphene is zero [39].

Considering the longitudinal component of the electric field as:

$$E_z = Re(\hat{E}_z e^{j\omega t}) \quad (\text{IV-4})$$

Then we can write:

$$f = f_0 + \delta f \cos(\omega t) \quad (\text{IV-5})$$

$$E_z = \hat{E}_z \cos(\omega t) \quad (\text{IV-6})$$

Putting results in equation (IV-1) results in:

$$-\delta f \omega \sin(\omega t) + e \hat{E}_z \cos(\omega t) \frac{\partial f_0}{\partial p_z} = \vartheta [f_0(p) - f_0(p) - \delta f \cos(\omega t)] \quad (\text{IV-7})$$

$$\delta f = j \frac{\partial f_0}{\partial p_z} \frac{e \hat{E}_z}{\omega - j\vartheta} \quad (\text{IV-8})$$

The axial current density in two dimensions can be shown as:

$$J_z = \frac{2e}{(2\pi\hbar)^2} \sum_{p_\phi} \int_{\text{first Brillion zone}} v_z f dp_z \quad (\text{IV-9})$$

where

$$J_z = \text{Re}(\hat{J}_z e^{j\omega t}) \quad (\text{IV-10})$$

and

$$\hat{J}_z = \sigma_{zz}(\omega) \hat{E}_z \quad (\text{IV-11})$$

Using equation (IV-9) results in:

$$\sigma_{zz}(\omega) = j \frac{2e^2}{(2\pi\hbar)^2} \sum_{p_\phi} \int_{\text{first Brillion zone}} \frac{\partial f_0(p)}{\partial p_z} \frac{v_z}{(\omega - j\vartheta)} dp_z \quad (\text{IV-12})$$

Where the summation in the first Brillion zone of SWCNT and is represented by $|P_\phi|$ [42]:

$$\begin{cases} |P_\phi| = \frac{s}{m} \frac{2\pi\hbar}{\sqrt{3}a_0}, & s = 1, 2, 3, \dots, m \\ |P_z| < \frac{4\pi\hbar}{3a_0} - \frac{1}{\sqrt{3}} P_\phi \end{cases} \quad (\text{IV-13})$$

In the first Brillion zone, “s” accounts for the quantized momentum in the circumferential direction [43] and “m” is from the Chiral vector (equation III-4). For solving equation IV-12, we first need to find the energy-momentum relation in graphene using equation IV-3, which requires analysis of the energy band using Schrödinger time independent equation.

A. Energy Band:

Experiments showed Newtonian Mechanics is inaccurate at atomic distances. Schrödinger presented an equation for both microscopic and macroscopic

universes called the “wave equation”. The equation is of two types, time-dependent and time-independent. Since electrons have fixed total energy, we should use the time independent Schrödinger equation to analyze CNT. From time-independent Schrödinger’s equation, energy in an electron is presented as follows:

$$E\psi = \frac{-\hbar}{2m} \nabla^2 \psi + U(\vec{r})\psi \quad (\text{V-1})$$

We can rewrite it as:

$$E\psi = \left\{ \frac{-\hbar}{2m} \nabla^2 + U(\vec{r}) \right\} \psi \quad (\text{V-2})$$

“ ψ ” is called the “wave function” which is the function of space coordinates and time. “ ψ ” shows the location of electron, “ m ” is the mass of the electron in vacuum, “ $\hbar$ ” is the reduced plank constant and “ $U(\vec{r})$ ” is the potential energy. The equation is only true for vacuum condition. In solid objects, like Graphene, an electron behaves almost as if it is in vacuum with the Effective Mass Equation (EME) m_{eff} . Replacing the electron mass with EME in the equation (V-1) results in:

$$E\psi = \left\{ \frac{-\hbar}{2m_{\text{eff}}} \nabla^2 + U(\vec{r}) \right\} \psi \quad (\text{V-3})$$

Although Schrödinger solved this equation for the Hydrogen atom analytically, there are very few practical examples that can be solved with this approach. Most are solved using numerical solutions turning the differential equation into matrix equation. Since Graphene has two-dimensional structure, elements in the equation will be in a two by two matrixes [44]. By presenting the differential operator as H_{op} we receive

$$H_{\text{op}} = \frac{-\hbar}{2m} \nabla^2 + U(\vec{r}) \quad (\text{V-4})$$

and can simplify the equation to:

$$E\{\psi\} = [H]\{\psi\} \quad (\text{V-5})$$

here, ψ_n is presented as:

$$\{\psi_n\} = \{\psi_\phi\} e^{i\vec{k} \cdot \vec{r}_n} \quad (\text{V-6})$$

Where “ r_n ” is the position vector of an atom. The differential equation can be written as:

$$E(\vec{k})\{\psi_\phi\} = \sum_m [H_{nm}] e^{i\vec{k}(\vec{r}_m - \vec{r}_n)} \{\psi_\phi\} \quad (\text{V-7})$$

Since ψ_ϕ is a matrix, it cannot be cancelled from both sides of the equation. In this case, we find the energy’s eigenvalue by finding the eigenvalue of $[h(\vec{k})]$ where:

$$[h(\vec{k})] = \sum_m [H_{nm}] e^{i\vec{k}(\vec{r}_m - \vec{r}_n)} \quad (\text{V-8})$$

To present $[h(\vec{k})]$, we use the nearest neighbor model [45], resulting in:

$$h(\vec{k}) = \begin{bmatrix} \varepsilon & h_0^* \\ h_0 & \varepsilon \end{bmatrix} \quad (\text{V-9})$$

“ h_0 ” can be written as:

$$|h_0| = -\gamma \sqrt{1 + 4 \cos^2\left(\frac{a_0}{2} k_y\right) + 4 \cos\left(\frac{\sqrt{3}}{2} a_0 k_x\right) \cos\left(\frac{a_0}{2} k_y\right)} \quad (\text{V-10})$$

The eigenvalue of matrix $[h(\vec{k})]$ is:

$$\varepsilon \pm |h_0| \quad (\text{V-11})$$

Where derived from experiments " ε " can be obtained and equals to zero [46]. Thus, the energy momentum relation for Graphene can be written as:

$$E(k) = -\gamma \sqrt{1 + 4 \cos^2\left(\frac{a_0}{2} k_y\right) + 4 \cos\left(\frac{\sqrt{3}}{2} a_0 k_x\right) \cos\left(\frac{a_0}{2} k_y\right)} \quad (\text{V-12})$$

Wherein " γ " is the overlap energy equal to 2.7e.V[46, 47]

From equations (IV-3), (IV-12), (IV-13) and (V-12) and considering a metallic CNT ($m-n=3q$, $q = 0,1,2,\dots$), the conductivity equation could be simplified as below [46]:

$$\sigma_{zz}(\omega) \cong j \frac{2\sqrt{3}e^2\gamma}{\pi\hbar^2(\sqrt{m^2+mn+n^2})(\omega+j\theta)} \quad (\text{V-13})$$

Consequently, for an armchair CNT, where $m = n$, we can find the conductivity as [46]:

$$\sigma_{zz}(\omega) \cong j \frac{2e^2\gamma}{\pi\hbar^2 m(\omega+j\theta)} \quad (\text{V-14})$$

B. Conductivity of CNT-DNA Origami Scaffold

Growing understanding of how DNA binds with SWNTs have led to several studies into the applications of synthesized DNA as templates for nano-electronic circuits [26, 48]. The synthesized DNA origami structure presented in section II can be used as a scaffold of highly conductive CNTs. The cross section and the dimension of the spiral structure presented in section II is given in figure 7. DNA origami is composed of five single strands of DNA that are linked together via crossovers. For the mathematical model, the width of CNT is approximately 10.82nm and as the radius of DNA is approximately 2nm.

Figure 7- Cross-section of DNA Origami Structure Assuming an armchair, for a CNT with the radius of 2nm, the value of m can be calculated from equation (III-10).

$$r_{CNT} = \frac{|c|}{2\pi} = \frac{a_0}{2\pi} \sqrt{n^2 + m^2 + nm}$$

$$m = 79.79 \cong 80$$

Thus the absolute value of conductivity for desired CNT is:

$$|\sigma_{zz}(\omega)| = \frac{1.2562 \times 10^{27}}{\sqrt{\omega^2 + \theta^2}}$$

That is 2.9975×10^{11} (s) for 450 (nm) and 3.2195×10^{11} (s) for 482 (nm) wavelengths.

V. APPLICATIONS:

In the following section, we discuss several applications of DNA origami based nanostructures. Programmable self-assembled DNA nanostructures can be used to create nanoscale highly asymmetric structures. Applications of such DNA origami range from a static scaffold for a two-dimensional nanoscale structure to a complex 3-D machine that is actively involved in drug delivery and complex biological reactions. DNA origami based scaffold to direct the growth of nanowires and interconnect are also investigated by various research groups. Our group focused on the development of DNA origami based nanoscale spiral antennas. Such antennas resonate at nanometer wavelength frequency; and have applications from point to point communications to fabrication of highly efficient broad spectrum solar cells.

VI. CONCLUSION:

In this paper, we present a DNA based nanoscale spiral structure. We further present a detailed protocol to develop such structure using origami. Techniques to fabricate DNA based structures with good electrical conductivity was briefly discussed in section III. Detailed mathematical models for CNT conductivity and DNA scaffold based CNT nan antenna has been presented in this paper. Our DNA structure has been synthesized

and an AFM images of synthesized DNA based spiral structure has been created at Brookhaven National Laboratory. The results show that such antenna can be used in biomedical as well as bio-electronics applications.

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