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Effective Meta-Materials for High-Power Microwave Applications

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Summary

This Document summarises the work undertaken at the university of Huddersfield in 2018 under AFOSR Grant FA9550-17-1-0197. The document summaries the work towards realising artificial electromagnetic materials for application in Vacuum Electronic Devices with a focus towards high-power high-frequency applications and manufacture.

Key points:

- Artificial EM materials enable precise engineering of the dispersion relation of materials and bulk material properties, to specific user defined criteria.
- Artificial EM materials enable unique tailored interactions for VEDs
- Conventional fabrication approaches to artificial EM materials cannot achieve the throughput and resolution of sub-wavelengths geometries, with nm feature size, that are ideally required.
- DNA-Origami could offer a cheap, reliable, approach to the mass fabrication of meta-atoms with sub-20nm features.
- Additional funding looking to scale-up DNA-Origami based nanofabrication has been secured from DSTL.
- Based on the DNA-Origami work a patent has been filed, and we are in the process of looking to create a spin-out company.

Journal Papers:

- Boyd, T., Gratus, J., Kinsler, P., Letizia, R. & Seviour, R. "Mode Profile Shaping in Wire Media: Towards An Experimental Verification", Applied Sciences. 8, 8, 11, p. 1276 (2018)
- Seviour, R. & Xu, Y., "Multibeam Photonic Klystron", Submitted - IEEE. Transactions of Electron Devices.
- Butler, Seviour, Byrek & Elfick "High Throughput Manufacture of DNA Origami", Submitted Nature Methods

Conferences presentations:

- Seviour, R. & Hopper, A., "Metamaterials for MIMO", URSI GASS 2017 (Quebec), IEEE, p.4
- Seviour R., "Artificial materials for EM Amplification & Generation", URSI GASS 2017 (Quebec) (Invited)
- Foulkes, S. & Seviour, R., "High-Power Metamaterial RF Sources", National Vacuum Electronic Conference, 10th Sept 2018 (Oxford)
- Seviour, R. & Butler, S., "Bio-inspired Artificial Media", National Vacuum Electronic Conference, 10th Sept 2018 (Oxford)
- R Seviour & S Foulkes, "Electromagnetic Wave generation and Amplification mediated via Artificial Materials", META 2019 (Portugal), pp. 942 (Invited)
- S. Butler & R. Seviour, "Synthetic Biological Approaches for the Fabrication of Optical Metamaterials", META 2019 (Portugal), pp. 1644
- Foulkes, S. & Seviour, R., "Metamaterial based RF Sources", PPS 2019 (Orlando), 2P08

1.0 Background

In Vacuum Electronic Devices (VEDs) generation and amplification of RF is achieved via particle wave interaction, in many devices this interaction is mediated in via a material, where the functionality of the material manipulates the EM wave in a controlled fashion. In a simplistic form this behavior between wave and material is described via the constitutive relations:

$$\mathbf{D}(k, \omega) = \epsilon(k, \omega)\mathbf{E}(k, \omega)$$

$$\mathbf{B}(k, \omega) = \mu(k, \omega)\mathbf{H}(k, \omega)$$

Where the permittivity (ϵ) and the permeability (μ) are the complex averaged EM response functions of the material due to the interaction with the electric and magnetic components of the incident wave.

Electromagnetic artificial materials are man-made macroscopic composites with periodic cellular sub-wavelength functional geometries acting as meta-atoms. Where the properties of the material come from the geometry of the meta-atom. These meta-atoms must be much smaller than the EM wavelength of interest ($< \lambda/10$) to ensure the we can treat the material as homogenous, this enables us to use an effective media theory to describe the interactions between wave and material using the bulk abstracted parameters permittivity (ϵ) and permeability (μ) of eqn1.

The history of artificial materials dates back to the 19th century to the work of Rayleigh and Bose in the 1890s. Rayleigh proposed a system of small scatterers as an equivalent continuous medium, and Bose produced an artificial chiral material by twisting “jute” root. This work was extended in 1914 by Lindman who considered small wire helices embedded into a host medium to create an artificial Chiral material. Although the first practical applications did not appear until the 1940s with the pioneering work of Kock. Kock created Artificial Dielectrics from arrays of sub-wavelength metallic structures (spheres, rods and plates) with the aim to develop light weight RF lens.

Prior to 1965 several researchers have considered materials with simultaneous negative permittivity and permeability, a Double NeGative (DNG) materials, but the first systematic study of the general properties of a hypothetical DNG medium with a negative refractive index is attributed to the seminal 1967 paper by Veselago. The seminal paper by Pendry in 2000 marked a turning point in artificial materials and can be said to be the key driver for the tremendous increase in interest and research of artificial materials since the beginning of the 21st century. The key aspect of Pendry's paper was to reconsider the Veselago's work, proposing how to realise a DNG material.

Around twenty years ago the word MeTaMaterial (MTM) entered the lexicon to refer to certain types of effective media. Even though a large number of peer-reviewed papers using the word metamaterial have been published an agreed definition of what a metamaterial is remains elusive. The origin of the word “meta” from the Greek “beyond” implies in some sense that “metamaterials” are a form of material beyond conventional materials. Sources suggest the term metamaterial was first coined by Rodger Walser in 1999 [3], who defined a metamaterial as; “...macroscopic composites having man-made, three-dimensional, periodic cellular architecture designed to produce an optimized combination, not available in nature, of two or more responses to specific excitation”, which in the case of a MTM is a simultaneous negative permittivity and permeability.

2.0 Metamaterial VEDs

The concept of utilizing MTMs in VEDs to mediate the interaction between wave and beam came rather later, with the first known publication by Seviour [1]. Whilst numerous papers and books have been published on the electromagnetic properties of metamaterials, until relatively recently all of the applications that have been described focused on low power devices. By manipulating the specific geometry of the unit-cells we can engineer the dispersion relation of the material to exhibit novel electromagnetic properties, such as engineering an arbitrary phase shift of an incident EM wave. Or in the case of a VED we can engineer the material to slow the velocity of an EM wave to match a particle beam, maximize the energy exchange between wave and beam, exhibiting either (or both) forward wave or backward wave interactions, see figure 2.1. This creates the possibility to imagine new and novel types of RF sources and amplifiers based around metamaterial mediated wave-beam interactions [1, 3].

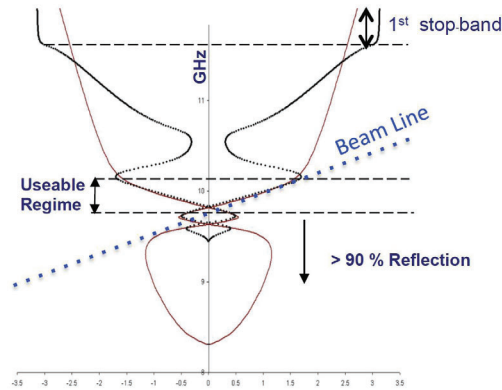


Figure 2.1: Engineered dispersion of a metamaterial loaded waveguide, designed with an extended electron beam interaction.

2.1 Beam interaction in loaded waveguide

To explore the potential of MTM based VED devices for RF generation we have designed a Complementary SRR (CSRR) based material, which has been loaded into a waveguide geometry, as shown in figure 2.2. Orientated such that the that an electron beam passes parallel to the CSRR. The material design has been optimized to act as a low-loss slow-wave structure. Figure 2.3 shows the refractive index of the CSSR based material in figure 2.2 determined via the Smith modified Nicolson Ross Wier (NRW) approach. Around 8.2 GHz in the refractive index the structure presents as a double positive media, with a very low imaginary component. The low imaginary component of the refractive index, relates to absorption, hence the material exhibits low loss.

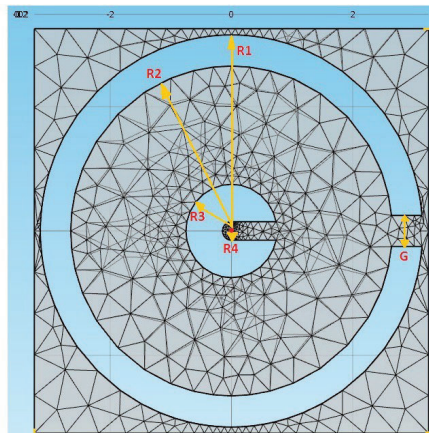


Figure 2.2: CSRR geometry based artificial EM material loaded into rectangular waveguide

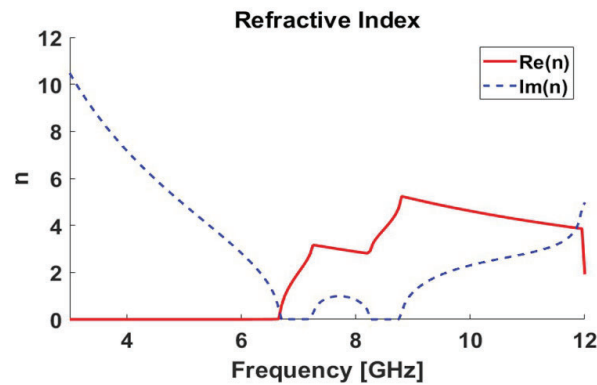


Figure 2.3: Refractive index of the CSRR based artificial EM material from fig 2.2.

Using the real component of the refractive index of the media the dispersion relation for a waveguide loaded with the material was determined and is shown in figure 2.4. Figure 2.4. also show the electron beam line interaction with a wave for a 30keV and 20keV electron beam These beams are quite low voltage compared to most conventional VEDs, and require the artificial media to slow the wave down to about 20~30% the speed of light in vacuum to enable the synchronization condition (wave phase velocity \approx beam velocity) for energy transfer between wave and beam to occur. These interaction points are shown on the dispersion curve in figure 2.4 at the points where the beam lines cross the dispersion curve. The 20keV beam shows a forward wave interaction at 8.45 GHz, which from figure 2.3 we can see at 8.45 GHz is a point of low absorption. Coupled EM and thermal simulations using the FEM software Comsol indicate that in the loaded waveguide system of figure 2.2 the material can withstand a continuous forward wave averaged power of 1kW before damage to the subwavelength unit-cell geometries occurs. Of course, in the case of pulsed power we would expect the system to be capable of supported 10s of MWs.

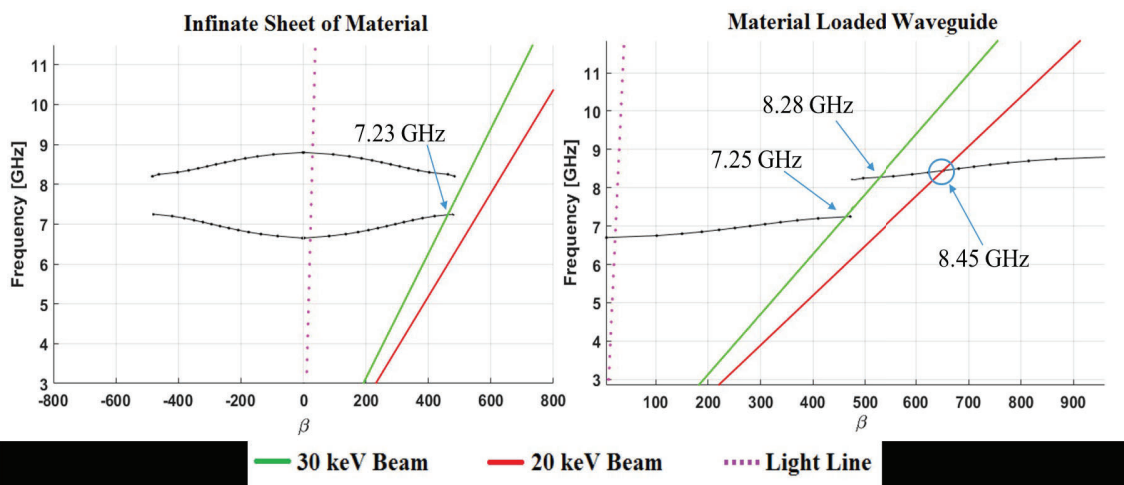


Figure 2.4: Dispersion relation for (left) the EM material from fig 2.2, (right) rectangular waveguide loaded with the material..

Using the 3D PIC code MAGIC the loaded waveguide system of figure 2.3 was modeled with a 20keV 0.5A electron beam emitted from the emitter, shown on the right-hand side, for 2.3ns. The monitoring ports on the left-hand side monitored the EM spectrum created by the interaction of the electron beam with the material. The results of the monitored EM

generated in the system are shown in figure 2.5, the graphs show the EM power measured at the port and the FFT showing the frequency spectrum of EM at the port, for the material loaded waveguide system (red) and the empty waveguide system (blue). As expected from the dispersion graphs of figure 2.4 the beam excites quite a pure EM spectrum at 8.45GHz in the loaded waveguide, whereas with the material removed we can see only broadband noise is observed at the port. Note the low power output is due to a non-optimised output coupling.

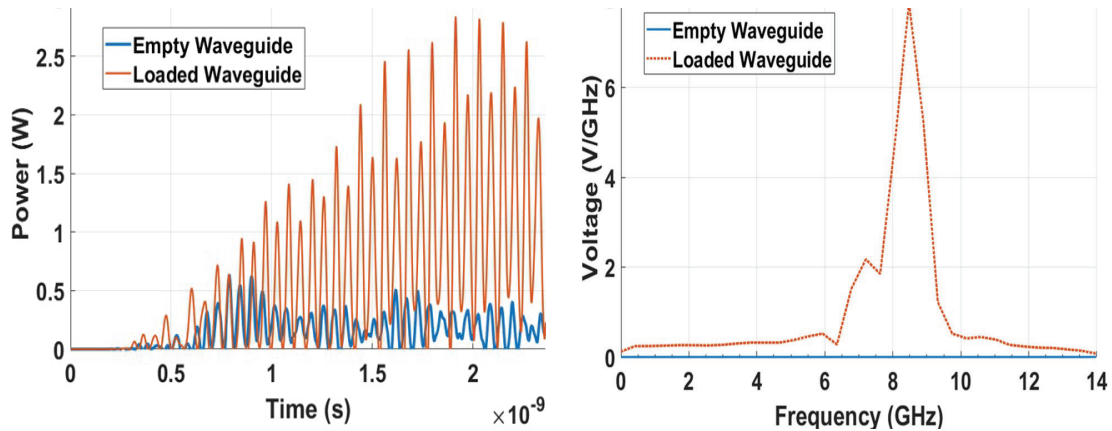


Figure 2.5: Power and EM spectrum measured at the port of the structure in figure 2.3.

2.2 Metamaterial VED

Having demonstrated the materials ability to mediate an energy exchange between wave and beam we move towards a realisable device configuration. Consisting of a cylindrical waveguide loaded with our artificial material formed from CSRRs. The device configuration is shown below in figure 2.6, the structure consists of 4cm long cylindrical waveguide loaded with the artificial material, joined to an expanding waveguide coupling system.

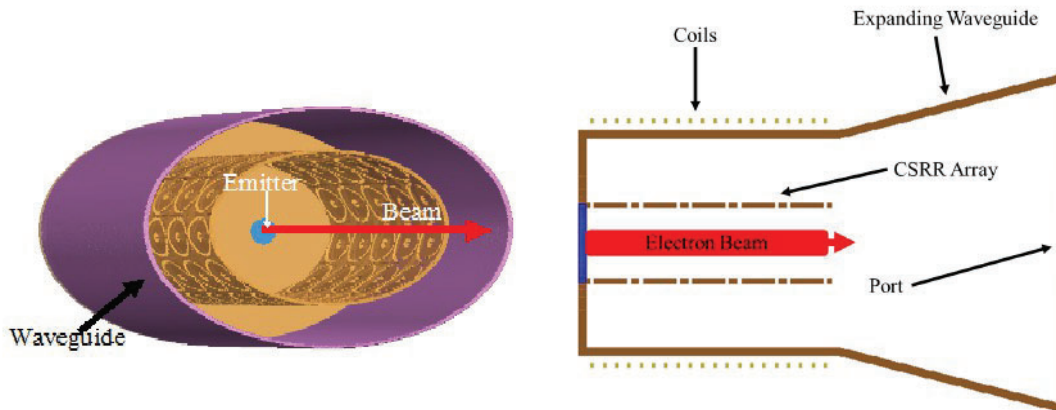


Figure 2.6: Current geometry under simulation.

Magic-PIC simulations of this configuration have been undertaken, we have considered several types of electron beams; 20 keV gyro-electron beam, 20 KeV DC Beam, 20 KeV Pulsed Beam, where beam currents of both 0.5mA and 200µA. As a check on the validity of the MAGIC sims Fig 2.7 shows the frequency spectrum generated by a DC beam through the cylindrical loaded waveguide. Using the refractive index determined by the NRW technique for the infinite block of material we simulated a volume of media loaded in to waveguide using comsol, with the refractive index determined via the NRW technique, using comsols FEM based eigenmode solver to identify modes of the system (i.e. no CSSRs were

modeled in the Comsol sims). As seen in fig 2.7 the Comsol FEM determined eigenmodes align with spectra in the frequency spectrum determined via the FDTD-PIC code Magic.

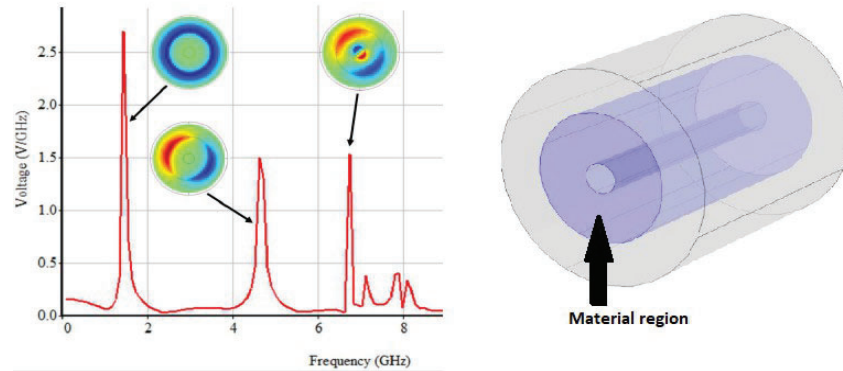


Figure 2.7: Frequency spectra and eigenmodes of the loaded waveguide.

This final configuration was modeled in MAGIC using a 100W electron beam, output power and frequency spectrum is shown below in figure 2.8, achieving 70W peak power output with a good frequency spectrum. Future and current work is underway to manufacture, build and test the device.

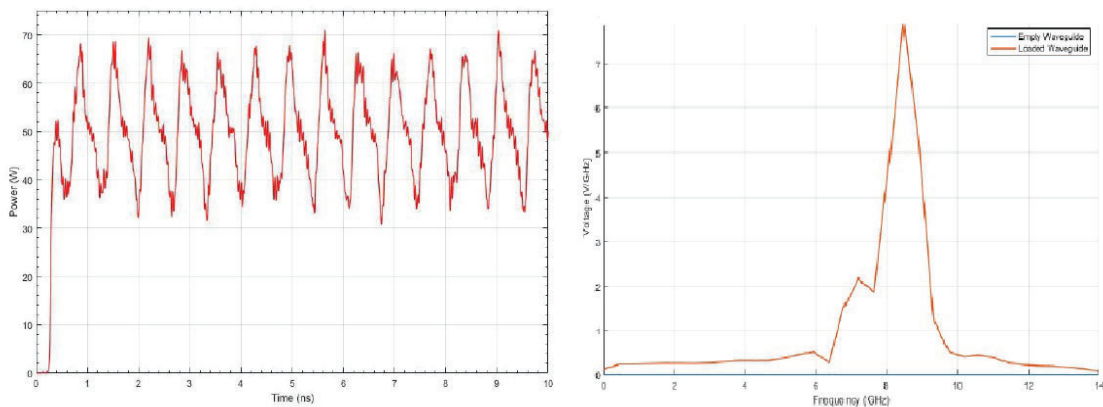


Figure 2.8: Current geometry under simulation.

2.4 Spatially dispersive materials

Recently we have started to expand our study of artificial media to include spatially dispersive media [5], we have a PhD student starting November who will pick up this work. The artificial materials we have considered so far are frequency dispersive, with no know work having been undertaken of the application of spatially dispersive media to VEDs or RF generation. Spatial dispersion is where the materials bulk parameters such as permittivity or conductivity have dependence on wavevector.

Our initial work has examined periodic arrays of dielectric wires of varying radius, we have found that this system behaves as a spatially dispersive wire medium that can support shaped longitudinal electric fields. This field profiling is achieved by varying the radius of the wires in a carefully calibrated way. Figure 2.6 shows the longitudinal electric field, present the electric field profile shaping, this enables us to create extended regions with a “flat” electric field.

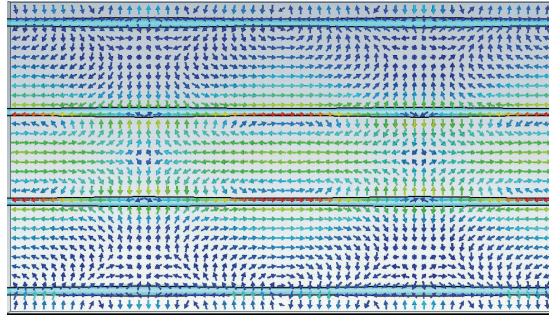


Figure 2.9 Electric field profile of our longitudinal mode as present in a 4x4 array of wires of varying radii, taken from [5]

Such field profiling has a variety of potential uses. The concept has been implemented by means of harmonic synthesis, and suggested in the context of nonlinearity-induced carrier shocking. The approach could find uses, such as fields with locally high gradients but without a large peak-minimising nonlinear effects, or fields with pronounced peaks and low amplitude elsewhere to improve signal to noise ratios

We intend to explore application of this field profile shaping for electron bunch shaping and enabling increased beam interaction regions in VEDs for RF generation and amplification. Initial results of our work can be found in ref [5].

2.4 High-Power Artificial Materials [Motivation for the DNA Origami approach]

The application of artificial materials (including metamaterials) in VEDs presents a challenging environment for the materials. Conventional realisations of materials fabricated from sub- 0.1λ geometries of SRRs or CSRRs, as discussed in ref [2] even at relatively low power can suffer from catastrophic failure. High power CSSR based materials can be design [4], fabricated from $\sim 0.16\lambda$ geometries, capable of withstanding MW peak power levels, although these structures offer a very narrow band of operation [4]. Recent work has shown that by utilising sub- 0.01λ geometries enables artificial materials to be fabricated that exhibit a very broad bandwidth response (multiple decades). In addition, previous work indicates that the smaller the meta-atoms the lower the losses compared to larger meta-atoms. Although of course these smaller dimensions of the meta-atoms necessities the manufacture of an extremely large number of meta-atoms.

2.5 Fabrication Approaches

Existing manufacturing approaches to meta-atom fabrication can be categorised into one of three methods; lithographic, microscopy and epitaxial. Lithographic techniques, based around ultraviolet and laser interference, offer high throughput with $>15\text{nm}$ resolution. The resolution can be improved by use of a focused ion or electron beam the throughput is significantly reduced. If we contrast the lithography's resolution to Scanning Tunneling Microscopy (STM) or Atomic Force Microscopy (AFM), where effectively it is possible to manipulate individual atoms, the throughput drops even more significantly. These techniques are also limited to the production of only 2D artificial materials. In comparison, the epitaxial techniques are capable of 3D fabrication, but have a low resolution ($>40\text{nm}$). Although all these conventional approaches require a very high level of investment and infrastructure.

3.0 Bio-Manufacture

For fully realised homogeneous metamaterials to become commercially viable the ability to mass produce at low cost, high yield, with high fidelity and a feature size of less than 20 nanometres must be developed. To date metamaterial fabrication has been conducted in a top down manner, where these approaches have been characterised as lacking the ability to effectively produce metamaterials [4]. In addition, current fabrication approaches are also effectively limited to producing 2D (bianisotropic) metamaterials, what we ultimately require

to realise the full potential of metamaterials, is a full 3D media, where individual, sub-100nm meta-atoms exhibit long range order.

To overcome the limitations of conventional fabrication approaches [4] we are currently investigating the use of biological systems as foundries for the mass production of metamaterials and identification of synthetic biological nanofabrication techniques. Utilising modular DNA origami functionalised with gold nanoparticles to create individual, sub wavelength, meta atoms, grouped into 2D, and 3D lattices with long range order.

DNA Origami offers a very high resolution, at a greatly reduced cost to the other techniques, at a very high throughput in comparison to even the high output lithographic methods. While DNA origami is not without its limitations: Functionalisation, where the other techniques directly manipulate or introduce materials DNA Origami provides a scaffold to which functional moieties are then attached, and aggregation, where the other techniques create meta atoms in period directly DNA origami produces individual meta atoms which must then be navigated and fixed into lattice. However it is certainly possible to interpret these limitations as advantages over the other techniques, where the other techniques rely on the use of certain materials any functional moieties which can be modified to interact with the DNA construct can be used to fabricate devices, and aggregation, where the other techniques are limited to 2D design (Or a crystalline structure), individual DNA constructs can be position and manipulated in solution, potentially, to create 3 dimensional lattices and super structures.

3.1 DNA Origami

This process involves the folding of a long single-stranded loop of DNA (the scaffold) aided by multiple short oligonucleotide strands (staples). Careful design of hundreds of unique staples allows us to programmable fold the scaffold into bespoke geometries. Each staple binds the longer scaffold strand in two discrete positions, “stapling” sections of the “scaffold” together. The synchronous action of hundreds of staples drives the self-assembly of a user-defined nanoscale shape (see Figure 3.1). Over the last decade DNA origami has matured such that the above feature size, complexity and fidelity are routinely achieved. Where it should be noted that this manufacturing approach can be readily scaled-up for production, the molecules needed are cheaply produced in bulk and the whole operation uses water as a solvent.

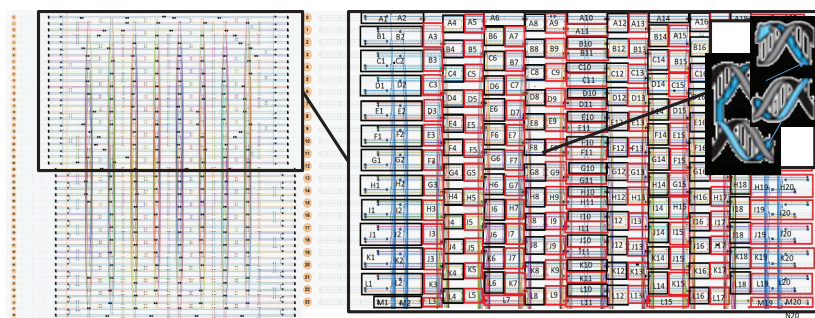


Figure 3.1: CadNano simulation of the DNA scaffold sequence with staples to form the DNA “breadboard”

The folded scaffolds can then be functionalised for the task at hand. It is trivial to decorate the scaffolds using the addressable nature of their DNA sequence to yield precise attachment of user-specified material (e.g. Au, Si, etc) to the structure. Using state-of-the-art DNA origami techniques to build scaffolds enables exquisite control of the nanoscale architecture of the MTM subwavelength artificial atoms. Metamaterials can be rationally designed, their geometrical and physical material properties tuned via adjustments to the DNA fabrication protocols. As the DNA is essentially acting “just” as a scaffold to enable the correct position and alignment for the metallic/silicon nanoparticles, after this has occurred

we no longer require the DNA scaffold, hence the DNA can degrade without effecting the performance of the material.

There are 3 variable parameters when considering fabrication of DNA origami:

- (1) Concentration of scaffolds to staples, seems to be largely arbitrary as long as a significant ratio of scaffold to staples is maintained.
- (2) Ionic concentration, is independent of other factors, such as stoichiometry and thermal cycles, and success can be visualised directly or observed by differential analysis of different samples moving through a gel electrophoresis. Correctly folded samples are likely to have a much tighter conformation and will pass through the gel quicker than less folded, and compact, samples.
- (3) The temperature that a DNA origami fabrication solution undergoes during thermal cycling/annealing is somewhat more debated than the others. There are essentially 2 contrasting theories in this matter. There is the possibility that isothermal annealing is effective and it is only the maintenance of a solution at its own, specific, fabrication temperature that dictates the effectiveness and efficiency of its folding.

There is also the suggestion that different DNA shapes, and different staple strands, display different specific folding pathways. Changing some staples even slightly can lead to significant interruption to these pathways. There also exists that possibility, especially when considering the different dynamics of different regions of a with functionalization points that flow freely until bound, that there is a combination of both dynamics at work. To ensure that my design is well folded (Obviously discounting direct AFM observation) a folding protocol which encompasses all temperatures, shown to be effective at folding similar, and then a further temperature buffer region, is used.

By the very nature of DNA replication, the probability of errors during mass fabrication of a given geometry are very small. Our approach, applying DNA origami to the long-standing issue of mass fabrication of optical MTMs is completely novel and scalable, enabling high-fidelity mass-production of stable metallic/semiconducting sub-wavelength nanostructures with sub-5nm resolution. Using this technique, the team have already demonstrated the ability to produce over 100 billion DNA sub-50nm origami geometries per cm^2 , see the AFM images of Figure 3.2 where the DNA origami structures are placed on a mica slide for imaging in an AFM.

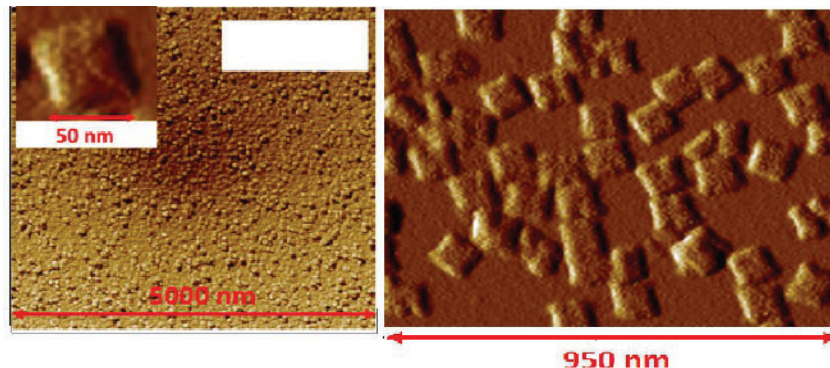


Figure 3.2: AFM image of the folded DNA "breadboard", inset zoomed in AFM of individual board.

The delivery of this project builds from our existing DNA "breadboard" technology, formed from folding the scaffold (bacteriophage M13MP18) into a 50nm X 50nm square of 5nm thickness as shown in Figure 3.2. The non-repetitive sequence of bases within this DNA yields a breadboard in which a unique position reference label exists for any point on the

breadboard. Each position can be addressed via a specific staple sequence, with no risk of mis-targeting. This enables us to pattern gold nanospheres (diameter 10nm) on to the surface of our breadboard forming a split ring resonator (SRR). The SRR has an individual engineered response to the electromagnetic field to produce the desired magnetic/electric susceptibility. The periodic construction of a tiled array of many SRRs is such that the electromagnetic wave interacts as if these were homogeneous materials.

3.3 DNA Functionalisation

As mentioned in the DNA Origami section the ease and reliability of DNA based fabrication comes from the specificity of Watson-Crick base pairing rules. Each of the designs we have examined have been constructed solely from DNA. DNA, while useful for construction/scaffolding, is electromagnetically inert and only exists stably in the comparatively mild environments dictated by physiological condition with few minor exceptions. This presents the need to “functionalise” the DNA by attaching material that will exhibit an EM response, such as Gold nanoparticles. For a material to be readily incorporated into DNA Origami based design, in a site-specific manner, it must first be complemented by its own unique DNA sequence.

The adsorption of thiol molecules to gold is a spontaneous reaction and forms a strong bond. Essentially the process is limited by the rate at which negative point charges on the gold, which are vital in maintaining the nature of the gold colloid, and negative point charges on the DNA are overcome and a thiol binds to a gold surface. If positive monovalent salts, such as sodium chloride, are used to counter point charges at a rate faster than thiols can bind, and provide stabilisation/protection to the gold nanoparticles, then the solution will aggregate. This enables us to pattern gold nanospheres on to the surface of our breadboard forming a split ring resonator (SRR). The SRR has an individual engineered response to the electromagnetic field to produce the desired magnetic/electric susceptibility. The periodic construction of a tiled array of many SRRs is such that the electromagnetic wave interacts as if these were homogeneous materials.

3.4 Current Progress - DNA fabrication

As shown in figure 3.2 we have demonstrated the ability to use DNA origami to mass produce with high-fidelity, structures with features of 2nm size. each run producing over 150 billion bread boards ready for functionalisation. The unique sequencing on these bread boards has enabled us to design via FEM techniques a series of unique thiolated DNA staples, where the thiol group forms a sulphur bond to gold nanoparticle, enable precise functionalisation of the DNA bread board.

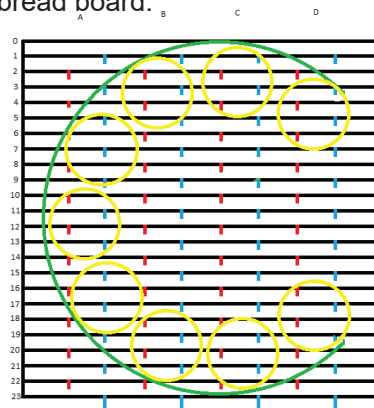


Figure 3.3: FEM simulation of the breadboard with the predicted binding sites of our gold nanoparticle

Figure 3.3 show the basic design of our DNA-Origami Split Ring Resonator, with a diameter of 50 nm. This simulation was used as the basis for our DNA breadboards shown in figure

3.2. Using these breadboards, we have carried out the functionalisation process, figure 3.4 below shows the AFM image of the functionalised breadboard with a series of 10nm diameter gold nanoparticles. As can be seen these form a rudimentary split ring resonator. The image presented is slightly noisy, due to the limits of the AFM, with approximate height variation between 1-17 nm from the mica surface. From Figure 3.3 the SRR structures appear slightly distorted, appearing more as “U” shaped than as the expected “C” shape. This appears to be due to the 10nm diameter gold spheres “lifting” the 5nm thick breadboards up at the edge. This appears to be caused partly by the relatively large 10nm diameter gold spheres compared to the 50nm by 50nm boards, and by the thiolated DNA tethers. To address this issue we are now using 3nm diameter gold spheres with a reduced number of tethers (lower left in fig 3.4). The smaller size and reduced tethers we expect to reduce the possibility of distorting the breadboard during the functionalisation process. Also we have produced long range order, the current breadboard design has a series of Poly-T’s encoded along the four edges of the breadboard, setup to take a unique “locking” staple that join adjacent boards together forming a 2D lattice of SRRs, as shown in figure 3.4, lower right.

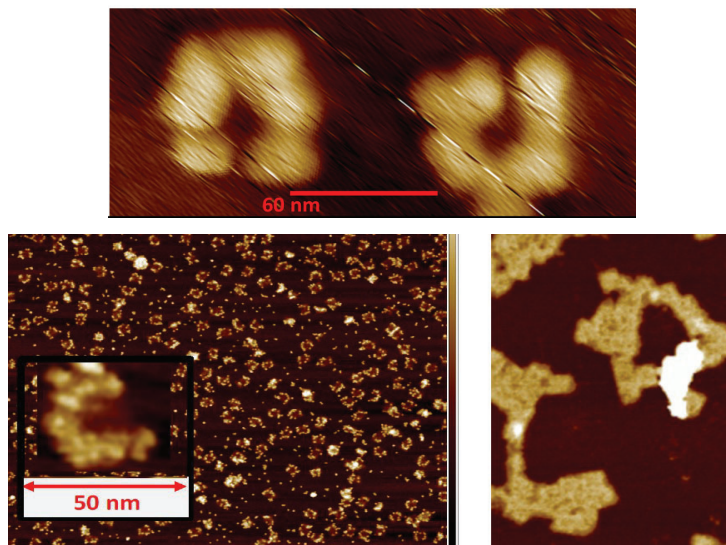


Figure 3.4: AFM image of SRR gold functionalised breadboards and long range order.

3.5 Future Progress - DNA fabrication

Short Term: Fabricate SRR film, and assess performance and interaction with EM waves, demonstrate optical metamaterial. Mid term: Recruit PhD student to look at using DNA-Origami to fabricate nano-electronic devices on to our DNA-breadboard design.

We are also looking to establish a company in the area of DNA origami nanomanufacture.

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