The ability of molecules to autonomously organise into well-defined nano- and microscale architectures while retaining the ability to recognize other molecules opens exciting opportunities in the production of new functionalised surfaces, nanoparticles, and responsive materials with applications in bioimaging, biosensing diagnostics and targeted drug delivery. The multidisciplinary approach needed for the development of these novel devices calls for events bringing together scientist from traditionally distant areas of science.

This one day workshop seeks gather together theoreticians and experimentalists from Physics to Life Sciences with the aim of exchanging knowledge and building up novel collaborations for synergistically solving real life challenges with self-assembly, molecular recognition, and their applications.

We further acknowledge the “Research and Innovation Staff Exchange (RISE) Project 645684 (Immuno-NanoDecoder)”
**Programme**

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*Evaluation and Optimisation of Interface Force Fields for Water on Gold Surfaces* |
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| 15:45 | Michael Rappolt (University of Leeds)  
*Lyotropic Liquid Crystalline Phases for the Formulation of Future Functional Foods* |
| 16:15 | Pietro Parisse (ELETTRA Synchrotron Light Source, Italy)  
*Biophysical analysis of Extracellular Vesicles* |
| 16:45 | Ryan A. Brady (University of Cambridge)  
*Crystallisation of Amphiphilic DNA C-stars* |
| 17:00 | Chandrashekhar Vishwanath Kulkarni (University of Central Lancashire)  
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| 17:15 | Closing remarks |

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Self-Assembly, Recognition, and Applications (SARA) 2017

14 December 2017, University of Lincoln Joseph Banks Laboratories, Lincoln, UK

https://www.iopconferences.org/iop/1179

Poster Programme

Alessandro Cecconello (Queen Mary University of London)
Programmed DNA Scaffolds for the Dictated Assembly of Left- or Right-Handed Plasmonic Au NP Helices Provide Insights to the Chiral Asymmetry Puzzle

Karen Johnston (University of Strathclyde)
Phase behaviour of self-assembled monolayers controlled by tuning physisorbed and chemisorbed states

Marjorie Ladd Parada (University of Leeds)
Impact of thermal pre-treatment on the crystallisation kinetics and polymorphic transformations of cocoa butter

Natalia Martsinovich (University of Sheffield)
Theoretical Modelling of Metal-Organic Frameworks for Exfoliation into Metal-Organic Nanosheets

Mukhil Raveendran (University of Leeds, don, London, UK)
DNA origami analysis using nanopipettes

Bart Vorselaars (University of Lincoln)
Self-organising diblock copolymers grafted to a sphere: a self-consistent field theory study
The workshop will be held at the Joseph Banks Laboratories, room JBL 3C01, University of Lincoln, Green Lane, Lincoln LN6 7TS.

The building is number 22 on the following map:
DNA-PAINT for resolving co-clustering properties of signalling proteins within intracellular nanodomains of the heart

Isuru Jayasinghe\textsuperscript{1,2}, Alexander H. Clowsley\textsuperscript{2}, Ruisheng Lin\textsuperscript{2}, Tobias Lutz\textsuperscript{2}, Carl Harrison\textsuperscript{2}, Ellen Green\textsuperscript{2}, David Baddeley\textsuperscript{3}, Lorenzo Di Michele\textsuperscript{4}, Christian Soeller\textsuperscript{2}

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\textsuperscript{4} Emmanuel College, University of Cambridge, Cambridge UK.

Super-resolution microscopy has revealed protein organisation in nature at unprecedented detail and sensitivity. Proteins like ryanodine receptor calcium release channels (RyR) in muscle are organised intimately in clusters, making them difficult to be visualised individually and quantified with techniques such as dSTORM where resolution is yet routinely limited to \(~\text{20-30 nm}\). DNA-PAINT \cite{1} is a new and alternative technique which exploits the high specificity and the predictable binding kinetics of complementary oligonucleotides for localising protein targets like RyR at high spatial precision. Applying this technique to imaging RyRs in heart muscle cells at resolutions of \(~\text{10 nm}\), we observed irregular arrays of punctate labelling densities and significant gaps within the clusters, which challenged the long-standing idea that RyRs are assembled into crystalline arrays \cite{2}. A series of Monte Carlo simulations confirmed to us that this RyR cluster topography is the likely result of a spontaneous, directionally-unconstrained, cluster assembly process. The nanoscale gaps which we observed within the RyR arrays were occupied by an inhibitory protein junctophilin-2 (JPH2) \cite{3}, which we localised in multiplexed DNA-PAINT imaging. We adapted a target ‘counting’ algorithm, called qPAINT \cite{4}, which could indirectly estimate the number of labelled proteins within a specific nanodomain by calibrating the first-order binding kinetics of the DNA probes. This revealed unexpected cluster-to-cluster variations in the JPH2:RyR ratio between 0.5 and 3.5 as evidence of a new level of complexity in the nanoscale regulation of RyRs. Similar methods of achieving improved resolution for target detection and assessing protein co-clustering stoicheometry represent a leap forward in studying in situ protein interactions in Cell Biology.

References

Naked protein nanoparticles

Ario de Marco

Laboratory for Environmental and Life Sciences, University of Nova Gorica, Slovenia

For long time, biological nanoparticles have been mostly thought as mere vehicles to deliver active molecules. Their role was to protect the cargo during in vivo transport, to reach the target cells, and to facilitate the intracellular delivery of the drugs. In front of these capacities, it seemed affordable the strong investment to build nanoparticles with the optimal biophysical characteristics, and to accept the biological risk that they could represent. However, nanoparticles are still highly inefficient in terms of amount of correctly delivered drugs (1). This observation prompted to conceive new strategies and in the last years a new approach emerged: all the nanoparticle components should be functionally active and involved in the therapeutic activity (2). This perspective requires that complex structures can be built starting from self-assembling components which still conserve sufficient activity once aggregated into ordered nanoparticles. Fusion proteins can be designed for being sensitive to production conditions to generate nanoparticles of variable features (3). The available examples are encouraging, offer space for further development (Figure 1), and at the same time suggest the necessity of further optimization steps.

Figure 1. Self-assembling protein nanoparticle which uses basic nanobodies for targeted delivery and cell internalization

References

Unravelling Hidden Abilities Encoded in the DNA Structure

Alexei A. Kornyshev

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DNA molecule continues to surprise us with new abilities encoded in its structure. I will overview some of the latest findings of the theory and experiments related with the structural effects on DNA-DNA interactions is solutions, with a focus on (i) DNA condensation and DNA mesophases, and (ii) the ability of homologous genes to recognize each other from a distance prior homologous recombination. The latter will be the main point of the talk, as it refers to one of the great mysteries of molecular genetics: how two homologous genes find each other in the genomic haystack. I will emphasize new experiments performed at Imperial College and Harvard University, which bring new light on this fascinating ability. The talk will summarize activities of a large team of people, including groups at NIH (S. Leikin), Imperial [D.J. (O')Lee, J. Baldwin, J. Seddon, T. Abrecht, and AAK]; Harward (M. Prentiss, C. Danilowicz), University of Minnesota (A. Wynveen) and few others.

Some key references
Displaying functionality into biomaterials through self-assembly

Helena S. Azevedo

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Queen Mary University of London, London E1 4NS, UK

Molecular self-assembly is becoming increasingly important in the development of innovative biomaterials because it offers a bottom-up approach for fabricating them with high level of precision and complexity. A number of building blocks has been explored for developing self-assembling biomaterials, but peptides offer many important advantages. The possibility of displaying biomolecular functionalities (e.g. small peptide molecules) with spatiotemporal control for triggering (macro)molecular recognition events is an important goal in biomaterials engineering. Through rational design, our group is integrating interacting domains into peptides to drive their self-assembly onto 2D solid surfaces (templated self-assembly) or into soft biomaterials. In this communication, I will cover some of our work on 2D and 3D biomaterial platforms to generate self-assembling biomaterials customized for specific biomedical applications (skin wound healing, bone regeneration, cell culture).

References:
Harnessing Nature's ability to create membrane compartmentalisation

Barbara Ciani

Centre for Membrane Interactions and Dynamics & Krebs Institute
Department of Chemistry
University of Sheffield, UK

A biological cell can be thought of as a complex chemical reactor where vast numbers of interactions are simultaneously taking place. To prevent unwanted cross-talk and interference within the "noise" of all these concurrent chemical pathways, a cell compartmentalises these processes localizing different functions within individual membrane-bound structures (organelles). Confinement of chemical processes also allows a cell to maintain incompatible environments that are optimal for each organelle's function, which would not be possible within a single "pot".

If we are to mimic this complexity within synthetic "nanoreactors", we need to develop ways of mimicking cellular compartmentalisation within synthetic structures. We will show that it is possible to create multi-compartment architectures, in vitro, using a purified membrane remodelling protein complex. We will also show how this in vitro system allows us to learn what controls the membrane shaping action of these proteins and therefore regulate the encapsulation of cargo.
Lyotropic Liquid Crystalline Phases for the Formulation of Future Functional Foods

Michael Rappolt

School of Food Science and Nutrition, University of Leeds, UK

Flavonoids are of great importance concerning the health benefits of foods and food additives due to their potential anti-oxidative, anti-inflammatory and immune regulatory actions. It is eminent that flavonoid consumption reduces the risk of several chronic diseases, including cancer and diabetes. Interactions of flavonoids with lipids, via their adsorption at the membrane water interface or insertion into the lipid chain region, can alter the lipid bilayers features (e.g. thickness, fluctuations and fluidity), and thus, influence their therapeutic potentials. The overall aim is to understand the structure-function relationship of flavonoids in the biomembrane in order to design functional food with a higher bioavailability profile. First results on encapsulation studies on flavonoids using curved lyotropic liquid crystalline phases are presented.
Biophysical analysis of Extracellular Vesicles

P. Parisse\textsuperscript{1,2}, F. Perissinotto\textsuperscript{2,3}, E. Ambrosetti\textsuperscript{1,3}, K. Pachler\textsuperscript{4}, M. Gimona\textsuperscript{4}, H. Amenitsch\textsuperscript{5}, L. Vaccari\textsuperscript{2}, A.P. Beltrami\textsuperscript{6}, D. Cesselli\textsuperscript{6}, L. Casalis\textsuperscript{1,2}

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\textsuperscript{6} Department of Medical and Biological Sciences, University of Udine, Udine, Italy

Extracellular vesicles (EVs) are small vesicles ensuring transport of molecules between cells and throughout the body. EVs contain cell-type specific signatures and have been proposed as biomarkers in a variety of diseases. Their small size and biological and physical functions make them optimal candidates for therapeutic agents in immune therapy, vaccination, regenerative medicine, and drug delivery. Indeed there is no objective set of criteria available for designing synthetic EVs for a specific task in biomedicine. It is therefore urgent and critical to address these issues for EVs-based medicine to fulfill its promise. Here, we try to assess the phenotypic properties of EVs, through a multi-technique characterization based on FTIR spectroscopy, Atomic Force Microscopy (AFM) and Small Angle X-Ray Scattering (SAXS). This detailed analysis allows us to model, visualize and quantify EVs’ physical and chemical properties up to single vesicle level and it serves as a basis for the correlation of phenotypic parameters of EVs with their functional activity. The proposed work, part of the European Regional Development Fund Interreg V-A Italia - Austria 2014-2020 project EXOTHERA ITAT1036, could allow for the design of innovative strategies for their sorting and detection, and for the personalized nanomedicine in general.
Functional DNA assemblies for bioimaging and biomarker detection

Abimbola.F.Adedeji\textsuperscript{a,b,c}, Alex Stopar\textsuperscript{d,e,f}, Franco Pascucci\textsuperscript{d,e}, Francesco Ricci\textsuperscript{d}, Andrea Dardis\textsuperscript{c}, Miguel Soler\textsuperscript{a,g}, Sara Fortuna\textsuperscript{a,g}, Giacinto Scoles\textsuperscript{a,b}, Matteo Castronovo\textsuperscript{a,b,c,d,h}

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\textsuperscript{h} School of Food Science and Nutrition, University of Leeds, UK.

Self-assembled DNA nanostructures are inherently amenable to novel applications in the areas of the life sciences and molecular medicine, such as biosensing and therapeutics\textsuperscript{1}.

In the first part of my talk, I will present a novel method, whose development is undergoing in our laboratories, for analysing multiple biomarkers in tissues and cultured cells, in monochromatic regime using an intelligent fluorescent label. Our prototype is comprised of an antibody inked to DNA nanostructures that contains a DNA triplex motif that can reversibly switch its fluorescence signal in response to pH, hybridization and enzymatic reactions, thus allowing imaging biomarkers one-by-one, in a layer-by-layer fashion. Likewise, we have tested our prototype in \(\alpha\)-tubulin cells.

The second part of my talk I will present a lithography application of atomic force microscopy (AFM) to assemble highly selective cyclic-peptides on a solid support. We formed confined nanopatches as prototypical screening arrays for biomarkers that are over-expressed in ovarian cancer\textsuperscript{2} i.e. Beta-2-microglobulin (\(\beta2m\)). Specifically, we applied AFM ‘nanografting’ and ‘DNA-directed immobilization’ (DDI) to spatially-orient DNA-cyclic peptides in the nanopatches to binds to the solvent-exposed binding site on \(\beta2m\). Next, we designed three, 10 amino-acids long, cyclic peptides that bind to two distinct sites on \(\beta2m\) (collaboration with Dr Sara Fortuna, University of Trieste, Italy). Each peptide was cross-linked with distinct 22 bps DNA using NHS-ester-primary amine/lysine conjugation chemistry. Using the approaches, we construct DNA-peptide nanoarrays with constant surface coverage. We then tested for \(\beta2m\) recognition by AFM, measuring the topographic height of the nanopatch before and after \(\beta2m\) recognition. We interpreted nanopatch height increase as positive binding signal associated with successful recognition of \(\beta2m\) on the surface\textsuperscript{3}. In this way, we obtained a binding curve that fitted the Hill’s equation. Our results show that three peptides have binding affinities towards \(\beta2m\) in the \(\mu\)-molar range, with one leading with 7 \(\mu\)M sensitivity.

References

DNA origami based platforms for characterising enzymatic reactions at the single molecule level

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Our understanding of enzymatic interactions is typically derived from averaged ensemble studies, including biochemical methods, static structural reconstructions, and indirectly reported kinetics such as FRET based studies. These approaches are typically limited in either their temporal or spatial resolutions and are insensitive to the subtle variations between individual biomolecular interactions which are arguably most revealing with regards to biological function.

Bionanotechnology offers novel experimental platforms for answering fundamental biological questions. Here, DNA origamis which form frame-like reference structures capable of hosting individual enzymatic reactions are exploited to enable studies at the single molecule level. When combined with high-speed AFM (HS-AFM) this approach enables the study of enzymatic function in real-time and with high spatial resolution whilst providing in situ references for characterising specific quantities such as interaction mode, geometry and enzymatic rate.

These aspects are demonstrated here for the DNA homologous recombination enzyme Recombinase A (RecA) (figure 1). The use of a DNA origami nanostructure in these experiments enables the specific interaction geometry, mode of interaction – including the presence of 1D facilitated diffusion – and sensitivity to sequence micro-homology to be quantified at the single molecule level for this nucleoprotein complex.

Figure 1. Determination of the mode of interaction of RecA nucleoprotein filaments (NPF) with double stranded DNA (dsDNA) during location of sequence homology. a) A schematic diagram depicting a DNA nanostructure containing a reaction (R) and control (C) dsDNA strand upon which RecA can interact. A polarity marker is included in order to discern the orientation of the nanostructure (black triangle). b) A HS-AFM micrograph of RecA NPFs locating homology within a DNA nanostructure. c) The motions of an NPF along the dsDNA can be plotted with reference to the nanostructure dimensions enabling distinctive interaction modes to be quantified.
Controlled Assembly of Multifunctional Nanohybrids for Single-Molecule Investigations

Mark Freeley, a Harley Worthy, b Antonio Attanzio, a Alessandro Cecconello, a Ben Bowen, b J Emyr MacDonald, c D Dafydd Jones, b Matteo Palma, a

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There has been a great interest in developing strategies to assemble functional materials with nanoscale accuracy towards the fabrication of platforms for single-molecule investigations. Single-molecule investigations are key to elucidate chemical and biochemical processes that can be averaged in ensemble measurements, and to optimize the performance of optoelectronic systems.

Here we present a universal approach for the generation of multifunctional nanomaterials that employ molecular building blocks assembled on carbon nanotube (CNTs) electrodes. We will demonstrate single-molecule control in the formation of nanohybrids via the in-solution assembly of classes of molecular materials (inorganic and biological with promising attributes) to DNA wrapped CNTs.

In particular, we report the site-specific coupling of single proteins to individual carbon nanotubes (CNTs) in solution and with single-molecule control. Green Fluorescent Protein (GFP) was engineered to contain a genetically encoded azide group and then bound to CNT ends in different orientations using click chemistry: in close proximity or at longer distances from the GFP’s chromophore. Atomic force microscopy and fluorescence analysis in solution and on surfaces at the single-protein level confirmed the importance of bioengineering optimal protein attachment sites to achieve direct protein-nanotube communication and bridging.

Additionally, individual quantum dots (QDs) were tethered to the ends of individual CNTs with single-molecule control. Here, DNA was used as a linker, where the distance between the CNT and QD could be modulated by using DNA sequences of varying lengths. Fluorescence measurements showed that the QDs’ emission was quenched as the particle approached the end of the CNT, an indication of a higher degree of coupling between the two nanomoieties. Notably, we produced not only static but also dynamic organic-inorganic heterostructures using a G-quadruplex DNA aptamer linker. Upon addition of K+ to the system, the G-quadruplex is formed bringing the QD closer to the end of the CNT. This is reversed upon addition of cryptand 222. In effect, this allows the reversible modulation of coupling between the QD and CNT through environmental stimulation.

By and large we demonstrate a novel approach for the assembly of single molecules on CNTs with a high degree of spatial control. The heterostructures show promise in the bottom-up fabrication of nanoelectronic devices for applications in chemical or biological sensing, as well as for light harvesting and optoelectronic devices.
Pore translocation of knotted DNA rings

Antonio Suma*, Cristian Micheletti*

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Biopolymers, such as DNA, can be long enough to become spontaneously knotted. This can have detrimental effects on their functionality in biological contexts, and in single-molecule manipulation experiments too. A relevant example is the translocation of DNA through biological or solid-state nanopores, which can become hindered by the presence of knots. We report here on a first systematic theoretical and computational investigation of such translocation for knotted DNA chains [1], giving insight into the conformation aspects of the process. Moreover, we show how simulations can be used to advance the interpretation and design of future experiments aimed at probing the spontaneous knotting of biopolymers.

Evaluation and Optimisation of Interface Force Fields for Water on Gold Surfaces

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The structure and dynamics of water at gold surfaces are important for a variety of applications, including lab on a chip and electrowetting. Classical molecular dynamics (MD) simulations are frequently used to investigate systems with water–gold interfaces, such as biomacromolecules in gold nanoparticle dispersions, but the accuracy of the simulations depends on the suitability of the force field. Density functional theory (DFT) calculations of a water molecule on gold were used as a benchmark to assess force field accuracy. It was found that Lennard-Jones potentials did not reproduce the DFT water–gold configurational energy landscape, whereas the softer Morse and Buckingham potentials allowed for a more accurate representation. MD simulations with different force fields exhibited rather different structural and dynamic properties of water on a gold surface. This emphasizes the need for experimental data and further effort on the validation of a realistic force field for water–gold interactions.
**In silico** design of binders for the molecular recognition of protein targets

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The design and characterization of protein binders is a widespread approach for important applications in nanomedicine and nanoeengineering, such as the molecular recognition of biomarkers, the oriented immobilization of proteins, and the inhibition or modulation of enzyme activity [1]. Antibodies have become the most popular binders due to their high stability and the exceptional selectivity they can reach [2]. However, the common engineering tools based on the maturation and *in vitro* selection of binder candidates are in general expensive, time consuming, and present difficulties to specify the target binding site to which the antibody will bind to [3]. Therefore, different methods and binders have been proposed as alternatives. One of the most promising approaches are the computational protocols for the design of small- and medium-size amino-acid based binders, as they offer minimum costs and optimal speed [4]. Here, we present a novel stochastic evolutionary algorithm for the design of binders that interact specifically with an *a priori* selected epitope [5]. Along these years, we have optimised peptides and single domain antibodies of camelid origin (also known as nanobodies) to bind proteins with different structural properties: β-2-microglobulin, lysozyme and HER-2. Our engineered probes have shown capable of recognising their target in solution, and have been further employed for protein oriented immobilisation.

![Diagram](https://www.iopconferences.org/iop/1179)

**Figure 1.**
(a) Optimisation kernel of the design algorithm. 
(b) Evolution of the scoring energies of peptide/β-2-microglobulin complexes during a typical run.

**References**
Crystallisation of Amphiphilic DNA C-stars

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Periodically structured nanoscale frameworks are critical for a host of applications including plasmonics, energy storage, molecular filtration, and sensing. A promising route for the reliable formation of such crystalline materials is through the development of building units which self-assemble in a pre-defined manner. Due to the highly specific nature of Watson-Crick base pairing, DNA has become a material of choice for the production of such units. However, due to the intrinsic physical properties of double stranded DNA the design space for successful architectures is very limited, and only a handful of all-DNA motifs able to crystallise in 3D have been demonstrated.1,2

We have overcome this long standing problem through a novel approach which combines the design freedom and nanoscale structural control offered by DNA nanotechnology with the robustness of hydrophobic interactions to produce single crystals with lattice parameters of over 30 nm, and overall crystallite sizes of over 40 µm.3 Here, the building blocks, which we term C-stars, are branched DNA junctions made amphiphilic through the addition of cholesterol (Figure 1).

In stark contrast to approaches based on all-DNA units, this methodology has proven to be robust to substantial changes in design. By changing the number of arms of the branched DNA molecule, the crystal structure of the resulting material can be altered (Figure 2.a). Additionally, C-stars offer the opportunity to precisely control both the size of the unit cell and the porosity of the resulting crystal by simply changing the arm length of the DNA moieties (Figure 2.b). Exploiting the controllable mesh size in combination with the amphiphilic properties of the material allows us to fine-tune the partitioning of macromolecules within the crystal, demonstrating the potential for these materials to be used as smart capsules for the encapsulation of specific targets.4 Due to the robustness and generality of this approach, C-stars promise to revolutionise the field of structural DNA nanotechnology and enable long awaited applications in a wide range of fields.

Figure 1: Amphiphilic DNA C-stars self assemble into single crystals through Watson-Crick and hydrophobic interactions

Figure 2: SAXS powder diffraction patterns (insets) and radially averaged intensity profiles (plots) demonstrate that crystal structure and lattice parameter, a, are tuneable by the shape (a) or size (b) of the DNA motif respectively

Towards Improving the Biocompatibility of Carbon Nanomaterials using Lipid Self-Assemblies

Chandrashekhar V Kulkarni1*, Vinod Kumar Vishwapathi1,2, Zeinab Moinuddin1, Yash Agarwal1,3, Pravin Kendrekar4

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Carbon nanotubes (CNT) and fullerenes (C₆₀) are exciting nanomaterials formed entirely of carbon. Their elegant structures and resulting properties are highly promising for many advanced and emerging applications. However, biotechnological and biomedical applications of these carbon nanomaterials suffer from inherent toxicity and corresponding lack of biocompatibility. Several researchers have tried to functionalize these materials in order to disperse them in water as well as to improve their biocompatibility.

Recently, we have established various protocols for decorating the lipids onto carbon nanomaterials and/or to encapsulate them within the lipid self-assemblies. Lipids, employed in this work, usually form non-lamellar (bicontinuous cubic) self-assemblies when mixed with excess water. These nanostructures can be further dispersed into oil-in-water emulsions retaining the lipid self-assemblies in the cores of the nanoparticles regarded as ‘cubosomes’.

![Image](image.png)

Single-walled CNTs, Multi-walled CNTS, highly-aligned CNT scaffolds (forests) and fullerene (C₆₀) were interacted with lipid self-assemblies formed from pure and commercial lipids including 1,2-Dioleoyl-sn-glycero-3-phosphoethanolamine (DOPE), 1,2-dioleoyl-sn-glycero-3-phosphocholine (DOPC), monoolein (MO) and dimodan U/J (DU). Details of the fabrication and characterization of these novel hybrid nanomaterials will be discussed in the presentation.

Programmed DNA Scaffolds for the Dictated Assembly of Left- or Right-Handed Plasmonic Au NP Helices Provide Insights to the Chiral Asymmetry Puzzle

Alessandro Cecconello, † Jason S. Kahn, † Chun-Hua Lu, † Larousse Khosravi Khorashad, ‡ Alexander O. Govorov, ‡ and Itamar Willner.*

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Within the broad interest of assembling chiral left- and right-handed helices of plasmonic nanoparticles (NPs), we introduce the DNA-guided organization of left- or right-handed plasmonic Au NPs on DNA scaffolds. The method involves the self-assembly of stacked 12 DNA quasi-rings interlinked by 30 staple-strands. By the functionalization of one group of staple units with programmed tether-nucleic acid strands and additional staple elements with long nucleic acid chains, acting as promoter strands, the promoter-guided assembly of barrels modified with 12 left- or right-handed tethers is achieved. The subsequent hybridization of Au NPs functionalized with single nucleic acid tethers yields left- or right-handed structures of plasmonic NPs. The plasmonic NP structures reveal CD spectra at the plasmon absorbance, and the NPs are imaged by HR-TEM. Using geometrical considerations corresponding to the left- and right-handed helices of the Au NPs, the experimental CD spectra of the plasmonic Au NPs are modeled by theoretical calculations.¹

Phase behaviour of self-assembled monolayers controlled by tuning physisorbed and chemisorbed states

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The self-assembly of molecules on surfaces into 2D structures is important for the bottom-up fabrication of functional nanomaterials, and the self-assembled structure depends on the interplay between molecule-molecule interactions and molecule-surface interactions. Halogenated benzene derivatives on platinum have been shown to have two distinct adsorption states: a physisorbed state and a chemisorbed state [1-3], and the interplay between the two can be expected to have a profound effect on the self-assembly and phase behaviour of these systems [4]. We developed a lattice model that explicitly includes both adsorption states, with representative interactions parameterised using density functional theory calculations. This model was used in Monte Carlo simulations to investigate pattern formation of hexahalogenated benzene molecules on the platinum surface [5]. Molecules that prefer the physisorbed state were found to self-assemble with ease, depending on the interactions between physisorbed molecules. In contrast, molecules that preferentially chemisorb tend to get arrested in disordered phases. However, changing the interactions between chemisorbed and physisorbed molecules affects the phase behaviour. We propose functionalising molecules in order to tune their adsorption states, as an innovative way to control monolayer structure, leading to a promising avenue for directed assembly of novel 2D structures.

Impact of thermal pre-treatment on the crystallisation kinetics and polymorphic transformations of cocoa butter

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A lack of consistency on the temperature used to melt cocoa butter prior to crystallisation studies has been made evident. Most commonly, temperatures of 50 and 60 °C are used, assuming that these are enough to start with a fully molten product; however, from compositional analysis it is well known that CB contains fully saturated triacylglycerols with melting points above 70 °C. Therefore, for fundamental studies that pursue an understanding of the crystallisation processes of CB, higher temperatures are needed.

Therefore, the aim of this project was to study the impact of thermally pre-treating CB, at three different temperatures (50, 80, and 100°C), on its crystallisation behaviour. Different techniques, such as X-Ray scattering and DSC, were used to evaluate the crystallisation kinetics, as well as to identify the polymorphs developed at different time-scales.
Theoretical Modelling of Metal-Organic Frameworks for Exfoliation into Metal-Organic Nanosheets

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Exfoliation (separation) of 3D metal-organic framework (MOF) structures into 2D metal-organic nanosheets (MONs) is highly desirable due to increased surface area of MONs and exposed labile coordination sites, allowing for uses in catalysis and sensors. We use computational modelling to evaluate the energy cost of separating MOFs into individual MONs layers. Following recent experimental work on MONs exfoliation,[1] we focus on MONs containing a pair of zinc or copper metal ions bound to terephthalate ligands in a paddlewheel structure, with a series of alkoxy side chains attached to the terephthalate ligands.

Benchmarking of computational methods shows that dispersion-corrected density functional theory (PBE+D) provides good accuracy, combined with DZVP and TZV2P basis sets for Zn-MOFs and Cu-MOFs, respectively. We find that the geometry of separated layers is slightly different from the same layers in their 3D MOF arrangement, and it subtly depends both on the nature of the central metal (Zn or Cu) and on the nature of the side-chain. Dispersion interactions between the interlayer organic chains of the MOFs are shown to be the main factor in the binding of the 2D layers into 3D structures. By varying the length of the organic side chains from pentoxy to methoxy groups, the binding energy of the 2D layers is reduced, suggesting that they will be more easily exfoliated. The propoxy (C₃H₇O) side-chain is shown to be the best of the chains tested: it provides weak but non-negligible interlayer binding in the MOFs and is expected to provide feasible exfoliation to form nanosheets.

References

DNA origami analysis using nanopipettes

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Controlled manipulation of single molecules at the nano-scale combined with a high-sensitivity feedback mechanism is a powerful approach to interrogate characteristic properties of the molecules which are often obscured when studied using traditional ensemble averaging techniques. Self-assembled DNA nanostructures (DNA origami) have the potential to be used for high-sensitivity biomolecular sensing. However, this requires precise detection and analysis of small changes in such structures at the single molecule level. Here we demonstrate that nanopipettes, a class of nanopores, are a promising approach capable of detecting variations between individual molecules.

We employ nanopipettes formed from glass capillaries with sub-100nm aperture as a tool to detect, deliver and analyze single DNA origami molecules. We demonstrate that the characteristic blockage current peak structure, equivalent charge and peak dwell time resulting from simple DNA origami translocating through the nanopore under an applied potential can be used to distinguish different DNA origami. The ability of nanopipettes to control the delivery and characterization of DNA origami molecules at the single molecule level demonstrates the potential of this approach for label free bio sensing applications with extremely high sensitivity.
Self-organising diblock copolymers grafted to a sphere: a self-consistent field theory study

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Polymer chains of which either half repel each other, characterized by \( \chi N \), have been extensively used as a way to pattern surfaces on the nanometre scale. The focus will be on a diblock copolymer film grafted to the surface of a spherical core, thereby creating patterned nanoparticles. This makes it feasible for promising applications such as environmentally-responsive surface patterns and drug delivery. Here self-consistent field theory \cite{vorselaars2011} will be used to explore the phase diagram for such systems. We developed an efficient algorithm based on the pseudo-spectral approach involving a spherical harmonics expansion in combination with a modified Crank-Nicolson method to solve the relevant partial differential equations. The particle exhibits various kinds of surface patches, when the fraction of non-grafted blocks, \( f \), is varied (see the figure).

\[ \chi N = 20 \]