Description of Ph.D. project in EXSS for Oct 2022 Entry

Project title:	Understanding charge dynamics in molecular solar energy conversion systems				
Principal Supervisor:	Jenny Nelson		Project No:		JN1
Email:	Jenny.nelson@imperial.ac.uk			Telephone	47851

Solar energy conversion in molecular electronic materials proceeds via photoinduced charge transfer across an interface between electron donating and accepting species. A critical question is how large the energetic driving energy needs to be to enable efficient charge separation and photocurrent generation, and hence what limits energy conversion efficiency. The influence of the driving energy on the loss mechanism of charge recombination similarly needs to be understood. Whilst past studies suggested that a threshold of some tenths of an eV were needed for successful charge separation, several recent observations indicate that the threshold may be very much lower, enabling higher charge separation efficiency. One key unknown is the role of molecular vibrations in mediating the desired charge separation and the undesired recombination processes. The aim of this project is to understand how the chemical structure and arrangement of the molecules controls the dynamics of charge separation and recombination, using a combination of spectroscopic measurements and modelling, and paying special attention to molecular vibrations.

Key research questions are:

- Under what conditions is efficient charge separation achieved with a small nominal driving energy? (when) Can charge separation be achieved in single component materials?
- How does driving energy affect charge recombination in an operating device?
- Can these processes be controlled via chemical design targeting molecular vibrational modes?
- How should the system be modelled in order to properly reproduce the impact of molecular energetics on charge separation and recombination?

In this project we will combine spectroscopic measurements on series of materials of systematically varying structure, with computational modelling of the processes studied. We will study the relevant excited states using electro- and photoluminescence, temperature dependent and transient measurements. Via collaborators we will have access to state-of-the-art ultrafast measurement techniques and data. With the aid of computational modelling we aim to identify the molecular features (such as vibrational modes) associated with charge relaxation, separation and recombination in the studied systems, and thereby analyse the effects of chemical structure on device performance.

The work will provide insight into the fundamental processes controlling solar energy conversion in molecular materials and may ultimately benefit photovoltaic technology. The methods are relevant to charge separation in other photochemical (such as photosynthetic) systems.

[1] M. Azzouzi, J. Yan, T. Kirchartz, K. Liu, J. Wang, H. Wu, J. Nelson, *Phys. Rev. X* **2018**, *8*, 031055. https://journals.aps.org/prx/abstract/10.1103/PhysRevX.8.031055

[2] F. D. Eisner, M. Azzouzi, Z. Fei, X. Hou, T. D. Anthopoulos, T. J. S. Dennis, M. Heeney, J. Nelson, *J. Am. Chem. Soc.* **2019**, *141*, 6362. https://pubs.acs.org/doi/abs/10.1021/jacs.9b01465

[3] J. Yan, E. Rezasoltani, M. Azzouzi, F. Eisner, J. Nelson, *Nat. Commun.* **2021**, *12*, 3642. https://doi.org/10.1038/s41467-021-23975-3

[4] M. S. Vezie et al, ACS Energy Lett. 2019 https://pubs.acs.org/doi/abs/10.1021/acsenergylett.9b01368

Techniques, activities, and equipment used:

Computational: To model behaviour of systems under excitation, recently developed non-adiabatic excited-state molecular dynamics methods will be used, alongside density functional theory to access electronic structure and molecular dynamics to simulate molecular conformations and interface structures in experimental systems.

Experimental: Electro- and photo-luminescence, photocurrent spectroscopy, transient optoelectronic measurements, temperature dependent measurements

Via collaborators: Raman spectroscopy, including femtosecond stimulated Raman spectroscopy; Transient absorption spectroscopy on different time scales.

Locations of equipment / collaborators:

Dr Tracey Clarke, UCL (working with the new femtosecond stimulated Raman spectroscopy capability enabled by UCL's new Photon Science Hub and other advanced vibrational and absorption spectroscopic techniques spanning femtosecond to millisecond timescales)

Dr Artem Bakulin, Chemistry (developing sub-femtosecond spectroscopic techniques to probe excited state dynamics in molecular electronic materials)

Description of Ph.D. project in EXSS for Oct 2022 Entry

Project title:	Evaluating organic mixed-conductor materials for sustainable electrochemical energy storage			
Principal Supervisor:		Prof Jenny Nelson	Project No:	JN2
Email:		Jenny.nelson@imperial.ac.uk	Telephone	47851
Other supervisors:		Dr Piers Barnes		

Aims of the project: Realising the full potential of renewable electricity resources will require new, sustainable and high-performance materials for electrochemical energy storage in batteries and supercapacitors. Organic electron- and ion-transporting materials are interesting candidates for both electrodes and electrolytes because of their abundance, low cost, low toxicity, mechanical properties and the potential ease of both manufacture and recycling [1]. Conjugated polymers are particularly attractive for use as electrodes in battery or supercapacitor devices. They charge and discharge rapidly, are stable over many cycles and work with non-toxic salt-water electrolytes [2], although their specific capacity is still limited [3]. Their properties can be tuned via chemical design, once their operational mechanisms are properly understood. Polymer-based electrode materials would thus enable a completely new electrochemical storage technology of low environmental impact.

In this project we aim to improve the electrochemical storage properties of conjugated polymer-based electrodes by developing a deeper understanding of the interactions between ions, electrons and molecules and of the charging process at a microscopic level and using these ideas to develop new material designs. We will study the ion transport and charging characteristics, specific capacity, redox stability and microstructure of polymer electrodes based on different materials using electrochemical and spectroscopic methods; and seek to relate the observations to chemical structure and energetics. These results will help to identify suitable electrode materials and test them within simple battery devices. The student will have the opportunity to use computational methods (possibly including device models, electronic structure and molecular dynamics) to interpret experimental measurements.

The key questions are:

- What ultimately limits the capacity of a conjugated polymer electrode?
- How does chemical structure and influence operational stability?
- What are the chemical design rules for higher performing materials and device architectures?

The ultimate goal of this research is to contribute to global climate stabilisation efforts, by exploring the options for storage and use of renewable energy.

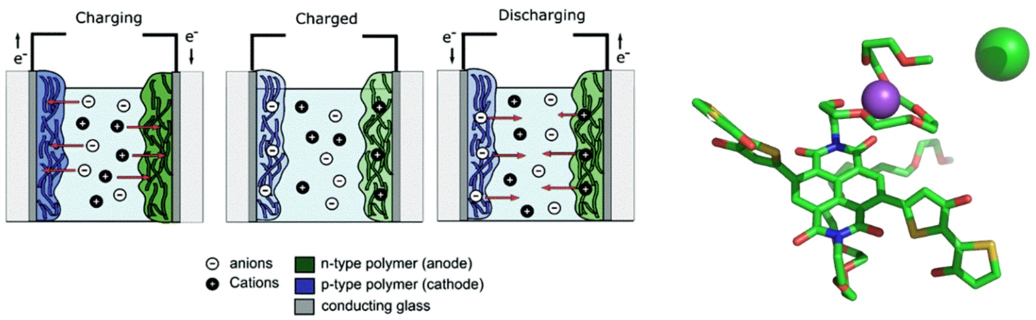


Figure 1: Left: schematic of the polymer battery concept; Right: molecular dynamics snapshot of ion-polymer interaction.

[1] S. T. M. Tan et al, ***Redox-Active Polymers Designed for the Circular Economy of Energy Storage Devices*** ACS Energy Lett. (2021) https://doi.org/10.1021/acsenergylett.1c01625

[2] Davide Moia et al., ***Design and evaluation of conjugated polymers with polar side chains as electrode materials for electrochemical energy storage in aqueous electrolytes***, Energy and Environmental Science, 2019, https://doi.org/10.1039/C8EE03518K

[3] A. A. Szumska et al., ***Reversible Electrochemical Charging of n-Type Conjugated Polymer Electrodes in Aqueous Electrolytes*** J. Am. Chem. Soc. (2021) https://doi.org/10.1021/jacs.1c06713

Techniques, activities, and equipment used:

Electrochemical, spectroscopic, optical and structural characterisation. Impedance spectroscopy, voltammetry, spectroelectrochemistry, quartz crystal microbalance, AFM and optical microscopy; device fabrication and testing.Simulation of ion dynamics using continuum models and atomistic molecular dynamics models. Some electronic structure calculation.Division of time between experimental and modelling work is flexible depending on student background and interests. The project would suit a student with a first degree in physics, chemistry, materials science or engineering and an interest in sustainable energy technology.

Locations of equipment / collaborators: The experimental facilities for this project are mainly located in EXSS labs (undercroft, clean room and level 9). Some spectroscopic or chemical characterisation measurements may be done at labs in Chemistry belonging to collaborators in the Imperial Centre for Processable Electronics. Occasional measurements may need to be done with or by collaborators in USA or elsewhere in Europe.

Collaborators:

Professor Magda Titirici, Chemical Engineering

Professor Martin Heeney, Chemistry

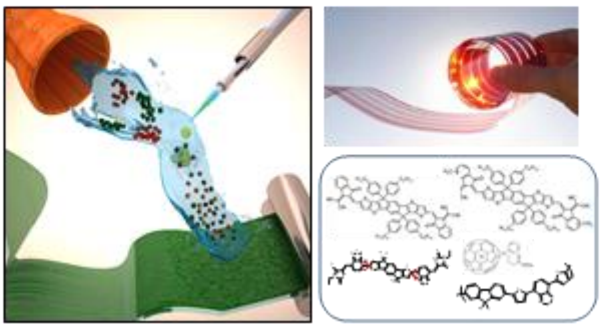
Dr Alex Giovannitti, Stanford University

Some industrial collaboration.

Description of Ph.D. project in EXSS for Oct 2022 Entry

Project title:	**Organic Solar Cells as Renewable Green Energy Source?**		
Principal Supervisor:	Prof. Ji-Seon Kim	Project No:	JSK1
Email:	ji-seon.kim@imperial.ac.uk	Telephone	ext.47597

Aims of the project: Organic semiconductors provide an exciting opportunity for large-area, light-wight, printable and flexible future electronics. In particular organic solar cells (OSCs) based on non-fullerene acceptors have made significant breakthrough in their device performance, now achieving a power conversion efficiency of ~18% for single junction devices, driven by the rapid development in their molecular design and device engineering in recent years. However, achieving long-term stability remains a key challenge to overcome for their commercialization. This is mainly due to the current lack of understanding of their degradation mechanisms as well as the design rules for enhancing their stability. There are many factors which can influence the photostability of OSC devices such as chemical/ electrochemical instability of materials, morphological instability of photoactive layers, and the interfaces between the photoactive and charge extraction layers. In this project, we will first investigate the photostability of organic solar cells using new electron donor and electron acceptor materials to explore these factors and identify which factors should be considered most importantly for photo-stable devices.



Schematics of printing flexible OPV devices (left), printed flexible OPV modules (top right), organic electron acceptors including NFAs (bottom right)

Beyond Si-based solar cells, finding niche applications is one of the potential ways for future of organic solar cells. Unlike opaque Si solar cells, colourful semi-transparent devices can be demonstrated using organic semiconductors – which can be applied to solar windows in buildings acting as renewable green energy source. Another key potential application is indoor light harvesting system. Unlike Si solar cells, thanks to well-matched light absorption properties of organic semiconductors to indoor lighting, organic solar cells show excellent device performance under indoor light – which can be applied to power supply system on Internet of Things (IoT). Based on our understanding of OSC devices and their unique advantages, we will also investigate potential applications of organic photovoltaic cells and identify important material/device parameters required for such applications.

References:

Clarke, A. J., Luke, J., Meitzner, R., Wu, J., Wang, Y., Lee, H. K. H., . . . Li, Z. (2021). Non-fullerene acceptor photostability and its impact on organic solar cell lifetime. CELL REPORTS PHYSICAL SCIENCE, 2(7), 22 pages. doi:10.1016/j.xcrp.2021.100498

- Labanti, C., Sung, M. J., Luke, J., Kwon, S., Kumar, R., Hong, J., . . . Kim, J. -S. (2021). Selenium-Substituted Non-Fullerene Acceptors: A Route to Superior Operational Stability for Organic Bulk Heterojunction Solar Cells. ACS NANO, 15(4), 7700-7712. doi:10.1021/acsnano.1c01345 accepted 23/3/2021
- Luke, J., Correa, L., Rodrigues, J., Martins, J., Daboczi, M., Bagnis, D., & Kim, J. -S. (2021). A Commercial Benchmark: Light-Soaking Free, Fully Scalable, Large-Area Organic Solar Cells for Low-Light Applications. ADVANCED ENERGY MATERIALS, 11(9), 10 pages. doi:10.1002/aenm.202003405
- Wang, Y., Lee, J., Hou, X., Labanti, C., Yan, J., Mazzolini, E., . . . Li, Z. (2020). Recent Progress and Challenges toward Highly Stable Nonfullerene Acceptor-Based Organic Solar Cells. ADVANCED ENERGY MATERIALS, 11(5), 41 pages. doi:10.1002/aenm.202003002
- Luke, J., Speller, E. M., Wadsworth, A., Wyatt, M. F., Dimitrov, S., Lee, H. K. H., . . . Kim, J. -S. (2019). Twist and Degrade-Impact of Molecular Structure on the Photostability of Nonfullerene Acceptors and Their Photovoltaic Blends. ADVANCED ENERGY MATERIALS, 9(15), 14 pages. doi:10.1002/aenm.201803755
- Wu, J., Lee, J., Chin, Y. -C., Yao, H., Cha, H., Luke, J., . . . Durrant, J. R. (2020). Exceptionally low charge trapping enables highly efficient organic bulk heterojunction solar cells. ENERGY & ENVIRONMENTAL SCIENCE, 13(8), 2422-2430. doi:10.1039/d0ee01338b

Techniques, activities, and equipment used:

OPV device fabrication and characterisation techniques (such as JV and EQE), as well as advanced spectroscopy techniques including confocal Raman spectroscopy, surface photovoltage spectroscopy (SPV), ambient photoemission spectroscopy (APS) will be used. This project will also involve theoretical simulations of molecules in terms of energy levels and molecular structures utilizing density functional theory.

Skills to be learned:Literature Survey (including device physics, molecular chemistry, and materials science)Scientific Research: Experimental (optoelectronic characterizations, photoemission spectroscopy, surface photovoltage spectroscopy, Raman spectroscopy); Theoretical (Density Functional Theory); Data processing (scripting), analysis, and interpretation.

- Professional skills: teamwork, project management, time management, and scientific presentation.

Locations of equipment / collaborators: Most equipment for device fabrication and characterisation is based at IC. The project will include collaborations with other academic (e.g. Imperial, Swansea, Oxford) and industrial (e.g. CSEM Brasil, KP-Tech, NPL) partners in OPV research. The project will be aligned with our recently awarded ATIP and GRL project to develop large area integrated organic solar cells for targeted applications (e.g. in-door photovoltaics).

Description of Ph.D. project in EXSS for Oct 2022 Entry

Project title:	**2D/3D Perovskites for Efficient Solar Cells**		
Principal Supervisor:	Prof. Ji-Seon Kim	Project No:	JSK2
Email:	ji-seon.kim@imperial.ac.uk	Telephone	ext.47597

Aims of the project:

Solution-processed three-dimensional (3D) perovskite solar cells have shown remarkable growth in recent years reaching a power conversion efficiency over 25% thanks to the excellent intrinsic properties of the perovskite photoactive layer (e.g. high absorption coefficient, excellent carrier mobility) and the growing scientific understanding leading to continuously improved morphology and interfaces to reduce losses in devices. One main factor still limiting the performance of and stability of perovskite solar cells is their high trap state density within the semiconductor bandgap. Therefore, gaining a fundamental understanding of these trap states in terms of their density and distribution, and finding a way to reduce these trap states will be crucial for further development of perovskite solar cells towards real commercialisation.

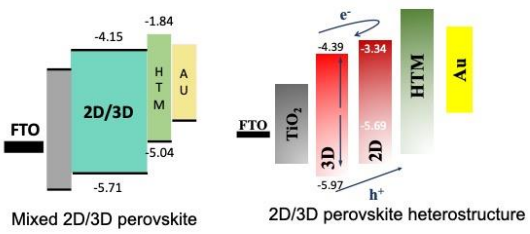


Figure 1. Schematic illustration of the dimensionally engineered 2D/3D perovskite thin film bandgap diagrams in mixed and heterostructure devices.

Recently, 2-dimensional (2D)/3D perovskite solar cells have attracted great research interest due to their potential to overcome the single-junction Shockley–Queisser theoretical efficiency limit, as well as to reduce the high trap state density and instability of 3D perovskite solar cells. In this project, we will first control the energy levels of perovskite layers via their dimensionality control (3D and 2D/3D) with varying composition and stoichiometry (applying different cations and anions in the 2D perovskite). We will then identify the nature of trap states in these dimensionality-controlled perovskite layers. The energy levels together with the trap state distribution will be investigated by using ambient photoemission spectroscopy (APS), which will be complemented by surface photovoltage (SPV) measurements to investigate the impact of cascaded energy levels and trap states on photogenerated charge carriers. Finally, the impact of dimensionality-controlled perovskites and their trap states on solar cell performance (efficiency and stability) will be investigated. The advanced spectroscopy techniques such as APS and SPV are relatively new and our expertise in this area will be crucial for the success of the project. The project will also include simulation of APS signals and/or one-dimensional drift-diffusion modelling of device performance and surface photovoltage.

References:

- G. Grancini et al., Nat. Rev. Mater. 4, 2019,
- S. Heo, G. Seo et al., Adv. Energy Mater. 31 (8), 2019
- C. T. Lin et al., Adv. Funct. Mater. 30, 2020
- Advanced Functional Materials, 30(25), 1-8. doi:10.1002/adfm.202001482
- ACS Applied Materials & Interfaces, 11(50) 46808-46817, doi:10.1021/acsami.9b16394
- ADVANCED SCIENCE, 5(11), 10 pages. doi:10.1002/advs.201801350

Techniques, activities, and equipment used:

Advanced spectroscopy techniques such as Ambient Photoemission Spectroscopy (APS) and Surface Photovoltage (SPV) Spectroscopy will be used to measure interfacial energetics and traps/defects related structural changes. Solar cell device fabrication and characterisation techniques (such as JV and EQE) will also be used.

Skills to be learned:

- Literature Survey (including device physics, physical chemistry, and materials science)
- Scientific Research: Experimental (optoelectronic characterizations, photoemission spectroscopy, surface photovoltage spectroscopy,); Theoretical (Density Functional Theory, one-dimensional drift-diffusion modelling); Data processing (scripting), analysis, and interpretation.
- Professional skills: teamwork, project management, time management, and scientific presentation.

Locations of equipment / collaborators:

Most equipment for device fabrication and characterisation is based at IC. The project will include collaborations with other academic (e.g. Imperial, Swansea, Oxford, QMUL) and industrial (e.g. CSEM Brasil, KP-Tech, NPL) partners. The project will be aligned with our recently awarded ATIP project to develop large area integrated solar cells for targeted applications.

Description of Ph.D. project in EXSS for Oct 2022 Entry

Project title:	**Nonlinear physics with extreme fields and Peta-Hertz Optoelectronics**				
Principal Supervisor:	Prof Rupert Oulton		Project No:	RFO1:	
Email:	r.oulton@imperial.ac.uk m.matthews@imperial.ac.uk		Telephone	x47576	
Other supervisors:	Dr. Mary Matthews (QOLS)				

Aims of the project:

When intense electromagnetic fields drive matter close to ionisation, Peta-Hz currents emerge generating extreme UV light and enabling resolution of physics on atto-second (10^{-18} s) timescales. To achieve the sufficiently intense optical fields, laboratory-based lasers are necessary, most of which are complicated bespoke systems: this is a barrier to wide-scale exploitation of this area of physics. This project explores extreme field opto-electronics essentially on a silicon chip. The primary excitement is the potential to break application barriers, eventually to enable portable and low-power implementations of extreme field physics. Moreover, the integrated opto-electronic setting enables the use of extremely high electrical fields in addition to optical fields. Uniquely, you will explore the mixing of both electrical and optical fields, both independently capable of ionising matter. This provides a unique opportunity to explore new physics.

The capabilities of such opto-electronic devices were demonstrated by our team with a publication in Science Magazine [*Science* **358** 1179 (2017)]. The technique has been honed over the past few years and is now ready to be deployed in this new area. The project will be co-supervised by Dr. Mary Matthews (Royal Society Fellow and Lecturer) who is an expert in extreme optical field and atto-second physics.

Techniques, activities, and equipment used

You will design and fabricate nanophotonic waveguide structures for optical experiments. The design process is conducted using Lumerical electromagnetic software. Nanofabrication will mostly involve electron beam lithography. You will test samples using our ultrafast laser laboratory. You will also have access to the ultrafast laboratories in QOLS to study the atto-second scale (Pet-Hz) responses of these devices.

Locations of equipment / collaborators

Nanofab lab, cleanroom, 1015 optics labs, QOLS group labs

Description of Ph.D. project in EXSS for Oct 2022 Entry

Project title:	**Hot Electron Science**		
Principal Supervisor:	Prof Rupert Oulton	Project No:	RFO2
Email:	s.maier@imperial.ac.uk r.oulton@imperial.ac.uk p.petrov@imperial.ac.uk	Telephone	Ext…47576
Other supervisors:	Peter Petrov and Stefan Maier		

Aims of the project:Optical nanostructures of metal or dielectric materials can support extremely large optical intensities of light. This is extremely useful for sensing applications – approaching single molecule level sensitivity. A side effect is the generation of heat due to absorption, which was thought to be a disadvantage but recently. The absorption processes generate "hot" electrons that can significantly influence physical and chemical processes near interfaces; not (only) as a result of the high electric fields, but also from the transfer of these energetic electrons to adjacent molecules or materials. This paradigm now allows photo-chemical reactors for water splitting and CO_2 reprocessing, for example.

The aim of this PhD project is to understand the physics and harness applications associated with such "hot" electron processes, induced by photo-absorption in designer nanostructures constructed from a new breed of materials. This will open up new paradigms in ultrafast control over nanoscale chemical reactions switchable with light, optically controlled catalysis, optical and electric processes in semiconductor devices induced by plasmonic hot-electrons, integration with two-dimensional materials such as graphene, as well as nanoscale metrology tools for temperature and field measurements.

Techniques, activities, and equipment

Electromagnetic simulations and design, Optical Spectroscopy, ultrafast spectroscopy, Chemical Analysis.

Locations of equipment / collaborators.

Collaboration with Stefan Maier's group at Ludwig Maximillian University, Munich and Peter Petrov's group in the Materials Department at Imperial. Extended placements in Munich are planned.

Imperial's Physics Department Cleanroom, Glovebox, Nanofabrication facility and Laser Facilities.

Description of Ph.D. project in EXSS for Oct 2022 Entry

Project title:	**Nanophotonic based single photon sources**		
Principal Supervisor:	Prof. Riccardo Sapienza	Project No:	RS1
Email:	r.sapienza@imperial.ac.uk	Telephone	49577
Other supervisors:	Prof Jenny Nelson		

Aims of the project:

Electron transfer to an individual quantum dot promotes the formation of charged excitons with enhanced recombination pathways and reduced lifetimes. Excitons are central for the development of very efficient quantum dot lasing, and very bright and tunable single photon sources from single quantum dot LED. We have just observed a 210-fold increase of the emission rate from a CdSe/CdS quantum dot under bias in an electrochemical cell (10.1126/sciadv.abb1821).

Now we want to push this result further and reach a deterministic control over the charge state and emission properties for classical and quantum communication technologies. You will measure the photons emitted by individual quantum dot, and develop a single quantum dot LED with tunable optical properties.

You will join a team encompassing our group focussed on spectroscopy, Prof Moreel's group in UGent expert on quantum dot synthesis, and Prof. Di Stasio's in IIT, expert on particle deposition.

The work is mostly experimental, with some theoretical work to model the electromagnetic interaction around the quantum dot (using commercial finite-difference time-domain codes) and the charge dynamics (approximate analytical methods).

Techniques, activities, and equipment used.You will use custom-built single-emitter microscope in a state-of-the-art nanophotonic laboratory.

Locations of equipment / collaborators.

B1016A, in collaboration with Prof. Iwan Moreels in Gent University (quantum dot fabrication) and Prof Francesco Di Stasio in IIT (quantum dot deposition).

Description of Ph.D. project in EXSS for Oct 2022 Entry

Project title:	**Photon pair generation from nanoscale dielectrics**		
Principal Supervisor:	Prof. Riccardo Sapienza	Project No:	RS3
Email:	r.sapienza@imperial.ac.uk	Telephone	49577
Other supervisors:	Prof. Stefan Maier		

Aims of the project:

You will develop bright nanoscale quantum sources of light through nanoscale photonic systems and nano-antennas. The project, joint between Prof. Stefan Maier and Prof. Riccardo Sapienza, builds on the latest advances in nanophotonics, plasmonics and single-molecule spectroscopy, which gives powerful tools to control light-matter interaction at the nanoscale. Nanophotonic antennas can boost by many orders of magnitude the light emitted by sources couple to them, e.g. see one of our latest work https://www.nature.com/articles/s41467-021-26262-3.

We are seeking an enthusiastic PhD student to undertake experimental research. The project involves design, nanofabrication and optical studies of nanostructures and metamaterials. The successful candidate should have a degree in physics, or material science. Independent thinking and multidisciplinary attitude are sought.

Techniques, activities, and equipment used.

You will use custom-built single-molecule confocal microscope, and an ultrafast spectroscopy setup.

Locations of equipment / collaborators.

B1016A

Description of Ph.D. project in EXSS for Oct 2022 Entry

Project title:	**Rewritable Magnetic Nanostructures**				
Principal Supervisor:	Dr Will Branford		Project No:		WRB1
Email:	w.branford@imperial.ac.uk			Telephone	46674

Aims of the project: The functional magnetism group has a strong interest in the magnetic properties of nanostructured materials and devices. We have recently developed a method of writing any magnetic pattern we choose into magnetic nanostructured arrays that are usually called Artificial Spin Ice using a magnetic force microscope.[1,2] The aim of this project will be to fabricate artificial spin Ice structures and to use the writing technique to explore the possibilities for two new types of computation. One of these, known as a neural network, is a massively parallel computation based on the collective response of the whole network. The other, known as magnonics[3], relies on manipulating spin waves (magnons) within the nanostructures.[4] Ferromagnetic resonance, or FMR, is a standard tool used for probing spin waves and spin dynamics in ferromagnetic materials. FMR arises from the precessional motion of the magnetization of a ferromagnetic material in an external magnetic field.

[1] Controlling the network properties from different starting configurations.

[2] Measuring magnetic dynamics in different geometries.

[1] J. C. Gartside *et al. Sci Rep-Uk* **6**, 32864, (2016).
[2] J. C. Gartside *et al. Nat Nanotechnol* **13**, 53, (2018).
[3] D. Grundler. *Nature Physics* **11**, 438-441, (2015).
[4] T. Dion *et al. Phys. Rev. B* **100**, 054433, (2019).

Techniques, activities, and equipment used: PhD projects in the group will typically involve a mix of sample preparation, structural characterization, magnetic and transport measurements and micromagnetic simulations. Cleanroom sample processing includes deposition of metal films and lithography (optical, e-beam and focused ion beam). Structural studies involve imaging by optical, electron and magnetic microscopy and x-ray diffraction. Magnetic measurement techniques include FMR spectroscopy, vibrating sample magnetometry and magneto-optic Kerr effect (MOKE) spectroscopy. Electrical transport studies include magnetoresistance and Hall effect measurements. These measurements can be performed over a wide range of temperatures and magnetic fields.

Locations of equipment / collaborators: All in the Blackett Lab. Fabrication in the EXSS nanofabrication lab and cleanroom. Other measurements in the functional magnetism laboratory (B815).

Description of Ph.D. project in EXSS for Oct 2022 Entry

Project title:	Design and Characterization of bespoke magnetic nanomaterials for neuromorphic and reservoir computing hardware			
Principal Supervisor:	Dr Will Branford	Project No:		WRB2
Email:	w.branford@imperial.ac.uk		Telephone	46674

Aims of the project:

The ballooning energy cost of computation is unsustainable, with more than 20% of global energy production forecast to be used for IT by 2030. Currently more the 75% of the world's data is stored magnetically and a key part of the problem is the 'von Neumann bottleneck' where ten times more energy is expended shuttling information between the processors and the hard drive as is used in the computation itself. A way to remove this bottleneck is to collocate the data storage and logic processing functions in the same device. This is how the brain works and so it is often referred to as neuromorphic computing. Software machine learning algorithms have multiple 'hidden layers' where the precise states are not known, but the weights of the different hidden elements are trained to achieve the best results. The idea of a physical reservoir is that these hidden layers are replaced by a physical system, which can do a lot of the work, and so simplify the computation, enabling more efficient machine learning hardware. Reservoir computing is particularly suitable for machine learning algorithms using dynamical (physical and software) systems that have non-linear response functions with some memory effects.

Several physical systems to perform reservoir computing have been proposed and demonstrated. Some of them exploit magnetic states of materials which have non-linear response functions to perform reservoir computing. The artificial spin ice system has excellent physical memory characteristics for reservoir computation, including strong non-linearity and complexity and a controllably fading memory. In this project, we will explore further optimisation of the nanostructured arrays to achieve the desired physical memory characteristics and further develop the reservoir computation protocols. Benchmarking tests with be performed for increasingly complex machine learning tasks, and performance will be measure against task-agnostic metrics for reservoir computing.

In our ongoing collaboration we have identified specific dimensions of ferromagnetic nanoislands that are bistable as either uniformly magnetized macrospins or as vortices. In a dense array of these nanoislands the particles are coupled such that macrospins promote neighbouring macrospins and vortices promote neighbouring vortices. We can perform the same operating magnetic field cycle 100 times, and each time there are slightly more vortices than the time before. The gradual learning and reinforcement of a task over many repetitions is exactly how our brains work and completely different from conventional computation. Additionally, we have found GHz spin spectroscopy to be a

very powerful tool for magnetic readout of the states. The latest findings in this system can be found in the paper linked here (https://arxiv.org/pdf/2107.08941.pdf). The potential of this for reservoir computing is entirely unknown and unexplored, which is the main aim of this proposed project. Specific objectives include: (1) identify which physical properties give the best reservoir computer performance by researching task agnostic metrics (2) test which nanostructures yield the desired physical characteristics? (3) benchmark performance against chosen metrics (4) compare to different systems and computational architectures.
Techniques, activities, and equipment used: PhD projects in the group will typically involve a mix of sample preparation, structural characterization, magnetic measurements and simulations. Cleanroom sample processing includes deposition of metal films and lithography (optical, e-beam and focused ion beam). Structural studies involve imaging by optical, electron and magnetic microscopy and x-ray diffraction. Magnetic measurement techniques include FMR spectroscopy, vibrating sample magnetometry and magneto-optic Kerr effect (MOKE) spectroscopy. Electrical transport studies include magnetoresistance and Hall effect measurements. In this particular project there will be a strong emphasis on taking utilising outputs from FMR spectroscopy data as the reservoir. Training on the reservoir outputs will be done with software machine learning (linear regression) methods.
Locations of equipment / collaborators: All in the Blackett Lab. Fabrication in the EXSS nanofabrication lab and cleanroom. Other measurements in the functional magnetism laboratory (B815).

Description of Ph.D. project in EXSS for Oct 2022 Entry

Project title:	**Plasmonic control of Magnetic Metamaterials**		
Principal Supervisor:	Dr Will Branford	Project No:	WRB3
Email:	w.branford@imperial.ac.uk	Telephone	46674
Other supervisors:	Prof. Rupert Oulton Email: r.oulton@imperial.ac.uk		

Aims of the project: The functional magnetism group has a strong interest in the magnetic properties of nanostructured materials and devices. We have recently developed a method of writing any magnetic pattern we choose into magnetic nanostructured arrays that are usually called Artificial Spin Ice using a magnetic force microscope.[1,2] The aim of this project will be to fabricate artificial spin Ice structures, use the writing technique define starting states and plasmonic heating induce controlled relaxation of the magnetic structure. Structures of this type are interesting for neuromorphic computing hardware. Neuromorphic computing is a massively parallel computation based on the collective response of the whole network from a defined starting point. The nanostructures will be of a size such that the magnetic structure is static and can be written in ambient conditions, and then can be selectively heated by interaction with laser light to induce directed magnetic relaxation, which will be used as the computation process.

[1] Controlling the network properties from different starting configurations.

[2] Exploring the evolution of the magnetic structure under laser illumination

[1] J. C. Gartside *et al. Sci Rep-Uk* **6**, 32864, (2016).
[2] J. C. Gartside *et al. Nat Nanotechnol* **13**, 53, (2018).
[3] M. Pancaldi *et al. Nanoscale* **11**, 7656-7666, (2019).
[4] J. H. Jensen *et al.* in *The 2019 Conference on Artificial Life.* 15-22.

Techniques, activities, and equipment used: PhD projects in the group will typically involve a mix of sample preparation, structural characterization, magnetic and transport measurements and micromagnetic simulations. Cleanroom sample processing includes deposition of metal films and lithography (optical, e-beam). Structural studies involve imaging by optical, electron and magnetic microscopy and x-ray diffraction. Magnetic measurement techniques include FMR spectroscopy, vibrating sample magnetometry and magneto-optic Kerr effect (MOKE) spectroscopy. Electrical transport studies include magnetoresistance and Hall effect measurements. These measurements can be performed over a wide range of temperatures and magnetic fields.

Locations of equipment / collaborators: All in the Blackett Lab. Fabrication in the EXSS nanofabrication lab and cleanroom. Other measurements in the functional magnetism laboratory (B815).

Description of Ph.D. project in EXSS for Oct 2022 Entry

Project title:	**Neuromorphic computation on physical neural networks**		
Principal Supervisor:	Dr Will Branford	Project No:	WRB4
Email:	w.branford@imperial.ac.uk	Telephone	46674
Other supervisors:	Prof. Riccardo Sapienza		

Aims of the project: Neuromorphic computing, beyond the traditional van Neumann architectures of our personal computers, is poised to revolutionise the way we perform calculations, inspired by how the human brain works, and with better performances at a much lower energy cost.

You will implement neural networks on two different physical systems; arrays of magnetic particles and of nanoscale lasers, which are controlled by laser illumination. Both systems feature an array of coupled elements, where inputs can be encoded as a time sequence and the output measured as a frequency spectrum. The characteristic magnetic resonance, or lasing frequencies, have a non-linear dependence on the input sequence. In the nanomagnetic arrays it has already been shown that the frequency spectrum can provide numerous trainable outputs and so usefully act as physical reservoir in a neuromorphic computation (https://arxiv.org/pdf/2107.08941.pdf). The goal of the project is to transfer the same methodology to show useful reservoir computation from the random laser array structures and to compare the performance of the two systems for different neuromorphic computation tasks, such as time-series predictions, handwriting and language recognition.

The project, joint between Prof. Riccardo Sapienza and Dr Will Branford, builds on the latest advances in nanophotonics and nanoscale magnetism, which gives powerful tools to control the state of matter optically and at the nanoscale.
We are seeking an enthusiastic PhD student to undertake experimental research. The project involves design, nanofabrication and spectroscopy studies. The successful candidate should have a degree in physics, material science or engineering. Independent thinking and multidisciplinary attitude are sought.

Techniques, activities, and equipment used.

In this particular project there will be a strong emphasis on taking utilising outputs from ferromagnetic resonance (FMR) and ultrafast optical spectroscopy data as the reservoir. Training on the reservoir outputs will be done with software machine learning (linear regression) methods. Cleanroom sample processing includes deposition of metal films and lithography (optical, and e-beam). Structural studies involve imaging by optical, electron and magnetic microscopy and x-ray diffraction. Optical measurements will use custom-built lasing microscope and an ultrafast spectroscopy setup, Magnetic measurement techniques include FMR spectroscopy, vibrating sample magnetometry and magneto-optic Kerr effect (MOKE) spectroscopy.

Locations of equipment / collaborators. All in Blackett. B706, B815, nanofabrication laboratory.