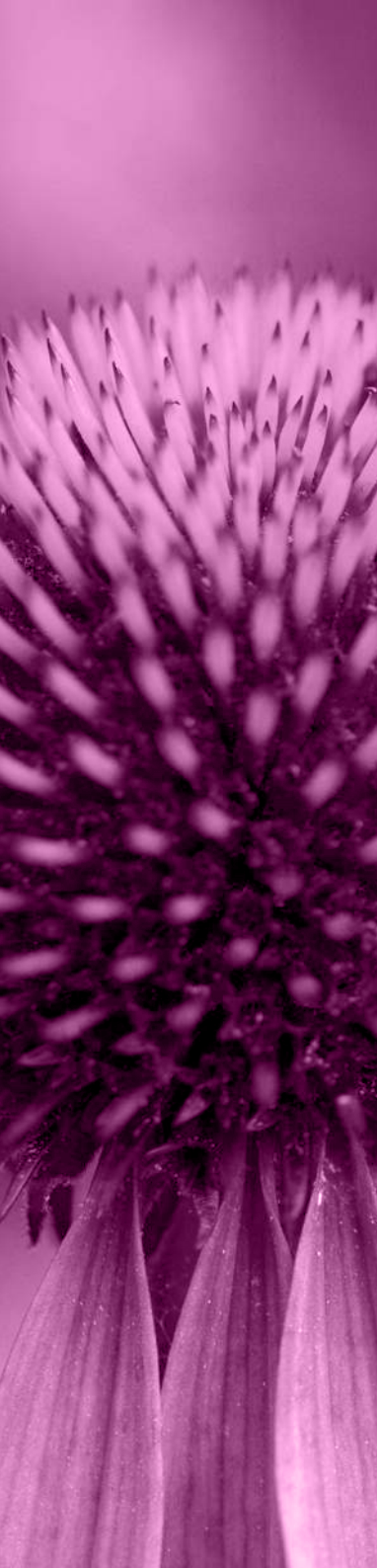




THE WINTON PROGRAMME FOR THE
Physics of Sustainability

ANNUAL REPORT

2022



Launched in 2011, the Winton Programme for the Physics of Sustainability was established through a £20 million donation from David Harding. This gift enables Departmental researchers to explore fundamental science research to develop the innovative technologies required for future renewable energy systems, resource-efficiency and low-power methods to meet the demands of a growing human population. Support of early-stage researchers through the Winton PhD Scholarship scheme, the Winton Advanced Research Fellows, and support of recently-appointed lecturers, is broadening the scope for fundamental advances in science to tackle the climate emergency. In this report students, fellows and lecturers from our Winton community will highlight some of their recent research breakthroughs.



REVIEW

Siân Dutton,
Winton Director

My Directorship of the Winton Programme began in 2020, following on from Richard Friend's tireless work in establishing the Winton Programme and championing its grand objective of supporting new science to address real and practical challenges in achieving a sustainable future.

When the Winton Programme for the Physics of Sustainability was established in 2011 through the donation of £20 million from David Harding, the aim was bold and timely: to support fundamental research that underpins new technologies that can impact how we manage our fragile ecosystems. Ten years later, the need to harness fundamental science to overcome depleting natural resources, increased pollution and climate change is more critical than ever.

Since 2011, the urgency to take action against climate change has become policy for the governments of many nations, currently enshrined in the 2019 amendment to the UK's Climate Change Act that requires net greenhouse gas emissions be reduced to zero by 2050. This goal necessarily requires an 85 % reduction in emissions by 2035, a milestone only thirteen years away.

After two years of a global pandemic, the goal of zero carbon emissions remains as formidable as it was ten years ago, but significant progress continues to be made: industrialisation at scale has brought wind and solar electricity costs to match those from fossil fuel generation, and electric car prices are ever-decreasing.

2020 saw the establishment of the University's climate change initiative, Cambridge Zero, which draws on the University's combined expertise in zero-carbon and sustainability research, education and engagement, to accelerate the global transition to a zero-carbon future. The Winton Programme engages strongly with Cambridge Zero to maximise the impact of the outstanding physics research undertaken within the Winton community.

This year we welcome back the much-loved annual Winton Symposium. Over the years this event has become established as the Department's annual forum for a broad and enthusiastic international community to come together to engage on topics of energy, sustainability and resource-efficiency. We are pleased to also support a further round of the tremendously successful Winton-Berkeley exchange, and we are currently reviewing applicants for the next cohort of Winton Scholars, to start their PhD studies in October 2022.

Over the past ten years, the Winton Programme has supported research in an extraordinarily broad range of topics, some with well-established connections to sustainability, and others seeding new exciting research areas, the impact of which will be revealed in the future. Our ten new 2021 Winton Scholars will all pursue their PhD projects on a wide range of topics, from sustainable drug design to new materials for rechargeable batteries, but with a common ambition: to explore basic science that can generate the new technologies and new industries needed to achieve the sustainability agenda and transform the world.

PROGRAMME UPDATE

Lata Sahonta, Programme Manager



Now in its eleventh year, the Winton Programme has provided a focus for sustainability research and technology, both in the Cavendish and across the wider University. During the two-year global coronavirus pandemic the Winton Programme continued to invest in new science and technology for a sustainable future.

Winton Pump Prime Funding has provided an innovative funding opportunity for Cambridge researchers pursuing exciting new research directions. Two such projects are outlined in this report.

The Winton-Berkeley Exchange scheme continued despite 2020's lockdown, and numerous exchange visits and sabbaticals were awarded to researchers hosted by both the University of Cambridge and University College Berkeley. Both institutions look forward to a new group of exchange visitors this summer to enhance their research activities

through collaborative projects with their host labs. Two 2021 exchanges from Cambridge researchers to UC Berkeley are summarised in this report.

The Winton Programme also continues to support recently-appointed lecturers through studentships, equipment facilities and travel funds, vital for research groups to explore new research phenomena. Hugo Bronstein from the Department of Chemistry describes recent Winton-supported research highlights from his group.

The recent easing of coronavirus restrictions has seen a revival of in-person seminars, discussions and social events in the Maxwell Centre, where the Winton Programme is officially based. Home to an interdisciplinary academic and industrial community, the Maxwell Centre is uniquely-placed for Winton researchers to engage with a range of sustainability activities including Cambridge Zero, the University's climate change initiative, and the University's

Energy Interdisciplinary Research Centre. Winton researchers can also make use of the Maxwell Centre's state-of-the-art equipment suites provided by the Henry Royce Institute for Advanced Materials and the Winton-funded Advanced Materials Characterisation Suite. The Maxwell Centre also enables Winton researchers to foster links with industry through a broad range of entrepreneurship initiatives.

March 2022 sees the welcome return of the Winton Symposium to the Cavendish with the theme of *Sustainable Futures*. Six high-profile speakers will be welcomed to the Pippard Lecture Theatre for talks and discussions on a range of sustainability topics, and will explore the future science and technology of a sustainable and resource-efficient future for all.

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Yorrick Boeije

Supervisor: Sam Stranks

Charge dynamics in optoelectronic materials



Pin Yu Chew

Supervisor: Rosana Collepardo-Guevera

Sustainable drug design: Investigating the evolution of antibiotic resistance through liquid-liquid phase separation



Cullen Chosy

Supervisor: Sam Stranks

Optical characterization of non-radiative losses at interfaces in perovskite solar cells



Rokas Elijosius

Supervisor: Alpha Lee

Design and optimisation of molecular catalysis for sustainable materials manufacturing



Mahmoud Elshenawy

Supervisors: Sian Dutton and Judith Driscoll

Resistive switching in amorphous ionic films



Lorenzo Peri

2022 NanoDTC MRes cohort



Nicholas Popiel

Supervisor: Suchitra Sebastian

Transport property measurements in high temperature copper oxide superconductors



Zhengkang Qu

Supervisors: Henning Sirringhaus and Akshay Rao

Spectroscopic mapping of local electronic/thermoelectric properties



Ayan Rakshit

2022 NanoDTC MRes cohort



Camilla Tacconis

Supervisor: Sian Dutton

Study of polyanionic materials as cathodes for Mg-ion rechargeable batteries

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Yorrick Boeije

Charge dynamics in optoelectronic materials

My research focusses on the interplay between vibrational dynamics, and exciton and charge transport at the femtosecond timescale in various optoelectronic materials, including the inorganic-organic hybrid lead halide perovskites. These materials have an ABX₃ crystal structure, in which the optoelectronic properties are primarily dictated by the inorganic BX₃- sublattice, with only an indirect role for the organic cation (A⁺).

Incorporation of large organic spacers results in the formation of layered (2D) perovskite quantum wells. These structures show great promise for applications in both light-emitting and photovoltaic devices, due to their enhanced stability and chemical tunability with respect to their bulk (3D) counterparts. Organic spacers with extended conjugated systems may be electroactive, implying a contribution to the optoelectronic properties, paving the way for novel rational design strategies. The excited-state behaviour, and in particular the coupling between the inorganic and organic sublattices, should be elucidated to achieve this. Tracking

vibrational coherences (VC) through excitation with ultrashort laser pulses, called impulsive vibrational spectroscopy (IVS), is an excellent tool to study the coupling of charges and excitons with the lattice on an ultrafast (<100 ps) timescale. In this experiment, femtosecond laser pulses generate a configuration-space localized nuclear wavepacket, which evolves over time and manifests itself as VCs in transient absorption (TA) spectra. These VCs provide structural information about the excited state in real time through extraction of the Raman mode frequencies by Fourier transformation, which has the potential to demonstrate how electroactive spacers impact exciton and charge transport.

When combining the temporal resolution of TA with nanometer spatial accuracy, these exciton-vibration interactions may be linked to local heterogeneous features such as defects. In particular, iodine interstitials are known to form deep trap states in bulk perovskites, which hampers their power conversion efficiencies.

With spatially-resolved IVS, the role of defects and their intimate connection with vibrational dynamics in nonradiative decay may be understood, which is key to minimizing this unfavorable carrier decay channel.

Pin Yu Chew

Sustainable drug design: Investigating the evolution of antibiotic resistance through liquid-liquid phase separation

The formation of biomolecular condensates in cells via liquid-liquid phase separation (LLPS) is an important mechanism which cells use to compartmentalise and organise the intracellular environment. A number of biomolecular condensates have been observed to display some degree of internal structuring, where immiscible phases with different compositions coexist within the same liquid droplet. Since such condensates are involved in regulating many biological processes, understanding the molecular driving forces for the assembly of complex multiphase condensates and the link between their structure and biological function is highly desirable.

My PhD research has mainly focused on investigating the formation of multiphase condensates using genetic algorithms and computer simulations. Recently, Lichtinger and co-workers have used genetic algorithms to evolve protein sequences towards either higher or lower upper critical solution temperatures. In our work, we extend this approach to investigate protein sequences that are

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able to form multiphase condensates. Accordingly, we couple a genetic algorithm to a residue-resolution coarse-grained model (developed in our group) to evolve a system that forms two liquid phases with a multilayered structure. Our co-evolution approach is able to maintain the multiphase organisation of the initial system, and allows for the design of protein sequences which, together with a predetermined sequence of interest, yield multiphase condensates. This approach therefore presents new opportunities for designing multilayered condensates, probing more closely the underlying physicochemical factors that lead to their formation and ultimately deciphering the missing links to their function inside cells.

This approach can be further extended to study the effect of additional components. Since many biologically-relevant multiphase condensates in cells (such as the nucleolus and stress granules) contain, or are stabilised by, RNA as a component, the co-evolution approach can be used to investigate the effect of RNA systematically. Comparison of the co-evolution of the same system with and without RNA present could reveal the role of RNA in the formation of such condensates. Another possibility would be to study the

effect of post-translational modifications (PTMs) on the phase behaviour of multicomponent systems, which would require us to develop a method to model such PTMs in the current coarse-grained model that we are using.

Other projects we plan on working on include investigating antibiotic resistance due to the Pf4 pro-phage in *P. aeruginosa*. A few experimental studies have shown that the rod-like Pf4 phage phase separates and assembles into liquid-crystalline droplets, which confers protection against antibiotics such as aminoglycosides by physically blocking the antibiotics from entering the bacterial cell. This mechanism of antibiotic resistance involves LLPS in prokaryotic systems, and computational studies on the phase behaviour of the Pf4 coat protein may give insight into potential ways to overcome such antibiotic resistance.

Cullen Chosy

Characterization of non-radiative losses at interfaces in perovskite solar cells

Metal halide perovskites are semiconductor materials with great potential for use in low-cost, high-performance solar cells. The past decade has seen perovskite solar cells rise to achieve power conversion efficiencies exceeding 25%, rapidly approaching the record for conventional silicon cells. More importantly, perovskites are relatively tolerant of crystalline defects such as missing or interstitial ions, allowing high-quality films to be deposited by simple methods. While conventional semiconductors such as silicon or gallium arsenide require extensive processing to form pure single crystals, perovskites can be rapidly deposited from a precursor solution onto a substrate as one might print a newspaper, enabling high-throughput processing at low cost. The remaining challenge for commercialisation of perovskite solar cells is to validate their long-term stability. One of the most important avenues towards improving device stability is by overcoming performance loss and degradation mechanisms initiated specifically at the interfaces.

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My work aims to characterize the interfacial chemistry and nonradiative recombination losses between perovskites and contact layers by applying a suite of complementary imaging techniques including photoluminescence microscopy as well as nanoscale X-ray diffraction (XRD) and X-ray fluorescence (XRF). Only through a holistic understanding of interfaces as well as bulk materials can nonradiative losses be effectively mitigated.

An exciting frontier for photoluminescence characterization of perovskite solar cells is the optical determination of local current-voltage (JV) curves. The open-circuit voltage of a solar cell is closely linked to its radiative efficiency, allowing voltage losses to be assessed through purely optical measurements. Specifically, we can fit the steady-state photoluminescence spectrum to extract the quasi-Fermi level splitting, which represents an upper limit on the open-circuit voltage. Mapping the quasi-Fermi level splitting using photoluminescence microscopy then reveals the spatial distribution of voltage losses in a device. Recent studies have further measured photoluminescence as a function of applied bias or excitation intensity to give curves that mimic a JV

curve. What distinguishes such optically implied “pseudo” JV curves is that they can be measured locally and thus correlated to structural or chemical features from nano-XRD or nano-XRF experiments. For example, nano-XRF measurements at the Diamond Light Source have provided maps of the halide distribution in a perovskite film, which can then explain shifts in the shape or intensity of local photoluminescence spectra. I plan to continue in these approaches to explain why certain perovskite/contact layer pairings outperform others, and then rationally design improvements.

Rokas Elijosius

Design and optimisation of molecular catalysis for sustainable materials manufacturing

My PhD project involves materials design through investigating biases in data-driven approaches, physics-inspired machine learning, and performing molecular simulations.

My research focuses on the optimisation of chemical processes. The chemical industry accounts for 15 % of global primary oil demand and 9 % of gas, making it the largest industrial energy

consumer. Many chemical processes require aggressive heating or rare catalysts to achieve high yields, thus requiring huge energy demands, resources and materials costs. More efficient catalysis could mitigate these issues.

Throughout my PhD, I aim to model various industrially relevant reactions with physics-inspired machine learning. A machine learning model can screen thousands of conditions in a fraction of the time, energy and cost compared to more traditional computational approaches. However, data-driven approaches require large amounts of data. A core assumption of many machine learning approaches is that seen and unseen data follow the same distribution. This is very rarely the case in historical data, thus, a core part of my research is finding ways to debias data.

Coming into Cambridge for my undergraduate, I thought I would be a synthetic chemist. However, many long hours in the laboratory and some fantastic theoretical courses persuaded me to follow a more computational path. Still, I remain excited by organic chemistry – only now I can do chemistry from the comfort of my computer chair. On a more serious note, molecular

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materials impacts countless industries. In the context of climate change, developing more efficient materials that use less energy, resources and generate less waste can accelerate the global pathway to a sustainable future.

Mahmoud Elshenawy Resistive Switching in Amorphous Ionic Films

Resistive-random-access-memory (ReRAM) is a promising emerging solid-state memory technology. On a fundamental level, ReRAM leverages the ability of a material to reversibly change its internal resistance state, from high to low and vice versa, i.e. resistive switching. In traditional ReRAM, an initial high voltage-forming process creates defect-formed conductive filaments in the material. Resistive switching is hence induced by the formation and rupture of these filaments. The operation mechanism and forming process are, however, inherently stochastic and random, leading to challenges of poor control, uniformity, scaling and stability.

The development of non-filamentary ReRAM promises to overcome these issues. In these devices, resistive switching is instead achieved by altering the defect

profile at a Schottky interface, usually between the active switching material and the electrode. This modulates the Schottky barrier height and defines the high- and low-resistance states. The challenge lies in the materials, as this behaviour is shown in primarily complex oxides and perovskites (SrTiO_x, PrCaMnO_x, etc.), which are not suitable for CMOS scaling.

A suitable practical materials solution is key for the success of non-filamentary ReRAM. Amorphous oxides are particularly attractive for industry as they allow for Back-End-of-Line (BEOL)-compatible processing by sputtering at low temperature. Amorphous In-Ga-Zn oxide (a-InGaZnO_x or IGZO) is industrially established in TFTs for OLED displays, and is particularly attractive for ReRAM, thanks to its low oxygen vacancy migration barrier and highly tunable properties.

In my PhD project I am developing non-filamentary ReRAM devices using IGZO thin films grown by sputtering. Fabricated devices are characterised with electrical (I-V, pulse) and physical characterisation tools (XPS). I will explore the inherent links between growth conditions, device design and electrical performance in order to

develop optimum ReRAM devices. Ultimately, I intend to achieve a strong understanding of the inherent device physics, and leverage the mature IGZO platform to achieve circuit-level ReRAM implementations. This will open pathways into efficient hardware for neuromorphic computing, including artificial intelligence (AI) and deep learning (DL).

Nicholas Popiel Transport property measurements in high temperature copper oxide superconductors

My research focuses on unveiling signatures of strong electron correlations in the recently-discovered class of unconventional insulators. In these materials, the high electrical resistance suggests that the electrons are stationary; yet in stark contrast to this, quantum oscillations are seen in the magnetisation. These two diametrically-opposed observations are accompanied by a novel insulating phase in which the bulk of the material is that of a robust insulator, yet the surface appears to be a fair conductor. These robust surface states host many of the requisite symmetries and excitations to develop the next generation of quantum computers. The novel physics that emerge from strong electron correlations are not

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only interesting, but also may have the ability to reshape the future technological landscape!

Throughout my PhD I intend to identify new materials that may host these exotic insulating states, and systematically investigate what physical phenomena these states are accompanied by. Complementary magnetic, resistive, and other thermodynamic measurements will be conducted to determine the constituent parts of this class of unconventional insulators. Much of this work involves tuning the material with ultra-low milli-Kelvin temperatures and enormous magnetic field strengths. Research will likely be a collaborative effort spanning many national magnetic laboratories throughout the world.

Information technology has revolutionised the world, but it has been a while since silicon revolutionised information technology. A new world could soon emerge in which quantum computers reign supreme. To make this fanciful dream a reality, a suite of new materials with exotic physics must emerge. Finding materials which may meet these requirements is in part the goal of my PhD.

Zhengkang Qu

Spectroscopic mapping of local electronic and thermoelectric properties

Understanding the charge and heat transfer of organic materials down to small length scales is important to reveal the transport physics of these systems. However, different measurement methods may provide thermal conductivity values which vary greatly. I'm interested in using time-resolved microscopy to study organic thermoelectric materials, specifically interferometric scattering (iSCAT) microscopy, an exotic pump-probe technology used to visualise heat transport down to the nanoscale. By visualising the change of polarisability at different temperatures, we can then visualise the temperature distribution in the material at high temporal and spatial resolution.

Charge modulation microscopy is another powerful technique to investigate these properties with high spatial resolution. This will require setting up a new measurement system that speeds up the signal acquisition and improves the signal-to-noise ratio compared to our existing charge modulation spectroscopy setup. The mapping of transport phenomena will provide us with a more comprehensive idea about structure-property relationships in organic materials.

My research plan for 2022 is to measure the variation in the optical constant of thin films of the polymer DNTT at different temperatures with a UV-visible laser. This will be compared to similar temporally- and spatially-resolved thermal conductivity measurements of DNTT thin films using iSCAT, allowing a comparison of the accuracy of both methods. This will be used to optimise the charge modulation spectroscopy and microscopy setup, allowing me to electronically inject charge into a range of organic semiconductors and investigate their thermal and electronic transport properties.

In the meantime I am working on confirming the transient localisation phenomena in ion exchange-doped organic polymers by analysing the optical conductivity by FTIR. At room temperature, we have found that the optical conductivity at low frequencies is suppressed, as predicted by the transient localisation model. To get stronger evidence of transient localisation phenomena, I plan to measure the optical conductivity at different temperatures.

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Camilla Tacconis

Study of polyanionic materials as cathodes for Mg-ion rechargeable batteries

The increased interest in developing renewable energies to face the issues of climate change has been accompanied by the necessity to develop better performing energy storage devices. Commercially available batteries currently adopt lithium-ion technologies, based on the exchange of lithium ions. A growing concern in the industry regards both the safety of lithium-ion batteries as well as the limited resources of the material available. Researchers aim to develop batteries based on more abundant metals to produce safer batteries which present fewer risks of dangerous short-circuits. Many alternative candidates to lithium are being researched, amongst them divalent cations are particularly interesting for their high theoretical gravimetric capacities.

Magnesium presents an inexpensive, abundant and safe energy alternative, and appears to be a promising lithium substitute due to attractively high gravimetric and volumetric capacities, as well as reduced dendritic formation. Not only does it offer higher capacities than lithium, magnesium is also twenty-five times cheaper and about 2000 times more abundant. Although magnesium batteries appear as excellent candidates on paper, the know-how acquired over the years of Li-ion development is not always transferable. This results in the technology still being in the early stages of research and development, especially when it comes to the development of high-voltage cathodes.

My research focuses on a particular group of magnesium inorganic cathode materials: polyanions constructed from borates, phosphates, and silicates.

Pyro- and orthoborates are favourable candidates for Mg-ion cathodes due to the demonstrated light weight structure. I will be building on the study of polyanionic borates, addressing the issue of attaining high operating voltages and favourable diffusion rates at room temperature. In addition, I will extend the research to phosphate and sulphate polyanions as viable magnesium battery cathode materials. I investigate the possible synthesis routes in creating these cathode candidates and, once an optimal one has been selected, I will proceed with structural and electrochemical characterisation of the materials.

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Jesús Arjona Martínez

Supervisor: Mete Atature

Quantum networks with novel colour centres in diamond



Julia Maristany

Supervisor: Rosana Collepardo-Guevara

Environmental sensing inside cells: deciphering sensory liquid-liquid phase separation to inspire the design of sustainable nanosensors



Megan Groom

Supervisors: Jeremy Baumberg, Tijmen Euser, Clare Grey

Operando fibre-Raman sensors for lithium ion battery chemistry



Lars van Turnhout

Supervisor: Akshay Rao

Photophysics and applications of organic semiconductor-lanthanide nanoparticle spin coupled systems



Dawn Kelly

Supervisor: Guiliana di Martino

Antiferromagnetic (AFM) materials for spintronic and data storage applications



Wenjin Zhu

Supervisor: Henning Sirringhaus

Correlation of structure and charge transport in uniaxially aligned copolymers containing alternating donor and acceptor groups

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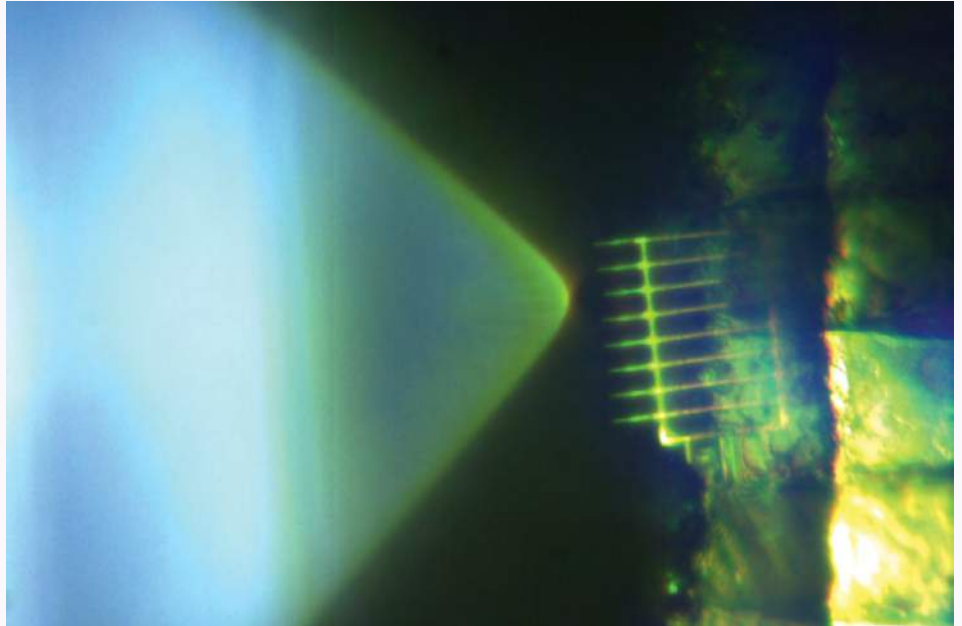
Jesus Arjona

Quantum networks with novel colour centres in diamond

Quantum networks are a platform for implementing quantum communication and distributed quantum computing protocols. By enabling quantum information to be shared over long distances, these quantum networks can benefit society by providing more efficient tools to solving a range of technological tasks.

Quantum computing allows parallel computing tasks to occur via *entanglement* and *superposition*, allowing quantum computers to be exponentially more efficient at solving mathematical problems such as integer factorisation and unsorted database searches. It is expected that the quantum approach will revolutionise the fields of cryptography and chemical simulation. Quantum communication is also important for applications in quantum key distribution, long baseline astronomical observations, or simply for sharing quantum information.

The goal of my PhD is to make a significant step in the development of practical, real-world realisation of quantum technologies. An idea tool to study quantum technologies is to use the nitrogen vacancy centre in diamond.



Above: A tapered fibre (left) collects light from a series of diamond waveguides deposited on the edge of a substrate (right), to improve collection efficiency over traditional macroscopic methods.

This is promising due to the ability of the vacancy centre to control spin, allowing easy manipulation of quantum information. At the current stage such systems have achieved a 3-node network with pairwise entanglement [1]. The practical value of this network is nonetheless limited by the low emission of coherent photons, only 3 %, which are needed to establish communication. In my PhD, we aim instead to explore

the use of a tin vacancy (SnV) as the increased symmetry due to the split-vacancy configuration in this system results in emission fractions of coherent photons of 50 %, and operation at 1.7K. Quantum control of the spin has recently been shown [2] and our goal is to make use of this to realise the building blocks of quantum networking, such as spin-photon entanglement.

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Key suggested developments in this road are:

1. Making use of fibre-coupled diamond waveguides [3] with implanted vacancies to maximize collection efficiencies. We expect this to significantly improve the rates of quantum communication protocols, as solid-state systems and in particular diamond offer a leading platform for integrated photonic structures.
2. Demonstrating the desired optical properties of the system. Recent work has been indicative of this [4], but key properties such as the indistinguishability of the emitted photons remain to be proven.
3. Exploring the dynamics of the system to characterise and improve the stability and repeatability of the devices, critical to achieving a future scalable system. This would follow recent developments on High Pressure High Temperature anneal of diamond [5], applied to nanostructures.

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Megan Groom

Operando fibre-Raman sensors for Li-ion battery chemistry

I am a Faraday Institution-affiliated Winton Scholar developing an embedded fibre-optic Raman chemical sensor for operando (in situ) monitoring of Li-ion batteries. The sensor is based on hollow-core optical fibres that avoid the large silica-Raman background signal that limits the use of conventional optical fibres in Raman spectroscopy. I am optimising and automating Raman analysis for both liquid electrolyte and electrode surfaces.

Preliminary results show that a micro-objective lens can enhance the sensitivity of a surface probe. To facilitate the development of these techniques, I have built a portable Raman set-up capable of liquid- and air- filled fibre Raman spectroscopy in forward and back scattered configurations. My future work includes further characterization of the

micro-objective surface probe, then I will study samples relevant for battery diagnostics and will explore using the portable liquid-based Raman sensor.

Dawn Kelly

Antiferromagnetic (AFM) materials for spintronic and data storage applications

One of the most promising contenders for ultralow-energy electronic devices are RRAMs and MRAMs, which deliver sustainably-scalable *neuromorphic* computing, potentially capable of reducing energy consumption in information and communications technologies by over 50%.

In particular, Antiferromagnetic (AFM) materials are emerging as highly promising materials for spintronic and data storage applications. The lack of stray field means that using magnetisation in AFMs to store information is a way to develop high density, non-volatile memories. However, the lack of stray field also means that it is not straightforward to read the magnetisation state of the material.

Current approaches to the mapping of domain structures in magnetic materials either rely on stray fields, require access to highly specialised facilities, or have limited spatial resolution. Development of reliable, scalable magnetic RAM would require detailed understanding of the

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mechanism of switching of the magnetic domains. Therefore a non-invasive, *in situ* technique to study the magnetic structure of AFMs is highly desirable. This project aims to establish an optical, tabletop readout method to access magnetism at the nanoscale, compatible for use with both ferromagnets and antiferromagnets.

The di Martino group adopts a novel approach to study optically-accessible memory devices by using nanoparticle-on-mirror geometries, to generate plasmon-enhanced ultra-concentration of light focused onto the switching material. This plasmonic hotspot allows us to collect optical signals from a nano-volume of the material of interest. In this project, movement of magnetic domains will be driven either via current-induced switching or direct application of magnetic fields, and optical signals will be collected during this switching. The aim here is to correlate changes in the dark-field scattering and/or Raman spectra with changes in the magnetic domain structure. Establishing such a framework to correlate optical signals with magnetic structure would allow us to read out information about magnetisation of a material on the nano-scale.

Julia Maristany

Environmental sensing inside cells: deciphering sensory liquid-liquid phase separation to inspire the design of sustainable nanosensors

In recent years, liquid-liquid phase separation has emerged as one of the key mechanisms used by eukaryotic cells to organise thousands of different components in the cytoplasm and nucleoplasm, via the formation of thermodynamically stable droplets we call condensates. Such biomolecular condensates are highly sensitive to changes in temperature, pH and biomolecule concentration, and therefore may serve as sensors for intra- and extracellular changes.

The physical parameters dictating the stability and composition of many-component biomolecular condensates, and how they react to intra and extra cellular changes, are not fully understood. My PhD project focuses on developing minimal coarse-grained models of proteins and nucleic acids to simulate mixtures of thousands of interacting multivalent proteins. These can be used to determine key factors that dictate the range of stability (e.g. temperature, pH, salt concentration) of different biomolecular condensates. These models will allow us to understand how LLPS is regulated in intracellular mixtures, and therefore to

decipher and eventually control how a cell organises its contents in space and time.

Understanding, and controlling, these cellular mechanisms is a key step towards the design of a wide array of new technologies, for example, sustainable nanosensors. Findings in this area of study could revolutionise the way we construct biological nanosensors, making those designs more efficient, accurate and sustainable. In addition to the potential technological impact, the development of minimal coarse-grained computational models involving multiple parameters and large amounts of data is invaluable as a reliable tool for numerical simulations in cases that otherwise would be computationally prohibitive. The need to develop models with these capabilities will only continue to increase, and a cost-efficient, energy-efficient and sustainable solution is highly desirable to make much better use of computational and energetic resources.

Lars van Turnhout

Photophysics and applications of organic semiconductor-lanthanide nanoparticle spin coupled systems

The ability to control triplet excitons is of paramount importance in and beyond the field of optoelectronics. As molecular triplet excitons are dark states, triplets

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can generally not be accessed directly nor harvested luminescently. Conventionally, triplet excitons are controlled via heavy-metal induced spin-orbit coupling or tuning of the singlet-triplet energy gap, both of which put heavy constraints on the system design.

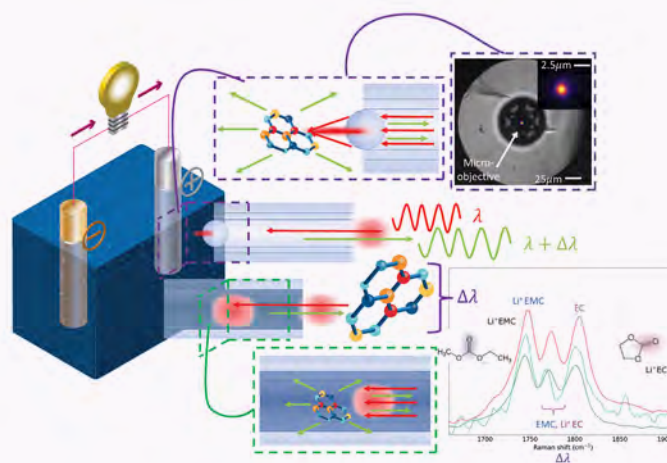
Recently, our group demonstrated triplet exciton control in organic molecules coupled to inorganic lanthanide-doped nanoparticles via a novel spin-exchange coupling mechanism, allowing for triplet exciton control from both the ground and excited state. My work explores the effect of molecular design of the organic molecules on the reported spin-exchange coupling.

By coupling three different anthracene carboxylic acid (ACCA) ligands to various lanthanide-doped nanoparticles, we demonstrated ligand-orientation dependent intersystem crossing (ISC) accelerations in the presence of lanthanides with unpaired 4f electrons. The ISC accelerations were not observed when the organic molecules were coupled to lanthanide ions without any unpaired 4f electrons, thereby showing that the observed effects cannot be attributed to heavy-metal induced spin-orbit coupling. Furthermore, we found ligand orientation-dependent triplet lifetime enhancements of up to three orders of magnitude upon coordination of the organic molecules to

the lanthanide-doped nanoparticles, giving rise to millisecond triplet lifetimes. These lifetime enhancements were observed upon coordination to any lanthanide-doped nanoparticle, not exclusively for ones that have unpaired 4f electrons.

Additionally, we have shown that energy transfer from the organics into the nanoparticles can successfully occur, and the resulting lanthanide emission could be related to the orientation of the surface-anchored ACCA ligands. Current measurements are looking in more detail at the distance dependence of the spin-

exchange coupling and energy transfer mechanism. Furthermore, using TADF molecules, the importance of the singlet-triplet gap in triplet control via this spin-exchange coupling process is investigated. Overall, these results provide new insights into a novel way of controlling triplet excitons in organic molecules and deepen our understanding of the relationship between molecular structure and photophysical properties of organic-lanthanide systems, which is essential for many optoelectronic and biomedical applications that utilise these systems.



Above: Kinetics of triplet generation and decay in pristine (1)-ACCA, ACCA-NaGdF₄, and ACCA-NaYF₄, measured using pump-probe spectroscopy. The results illustrate the enhanced triplet lifetimes upon coordination to NaLnF₄ nanoparticles, and show the accelerated triplet generation and higher triplet yield in the presence of lanthanides with unpaired 4f electrons.

ALUMNI REPORTS

Raj Pandya



I completed my PhD between 2016 and 2021, jointly supervised by Akshay Rao and Ulrich Keyser, and was supported by the Winton Program and EPSRC. My work focussed on developing and applying new optical methods to study light-matter interactions in organic and inorganic nanosystems for energy harvesting, energy storage and sensing applications.

Highlights included injecting quantum dots into frog embryos to visualise the depolarisation of newt neuronal membranes [*Nano Lett.* 2019, 19, 12, 8539–8549], performing experiments at magnetic fields beyond 30 Tesla to elucidate the spin fine structure of colloidal semiconductors [*ACS Nano* 2019, 13, 9, 10140–10153] and tracking how phonon modes influence a wide range of chemical reactions [*J. Am. Chem. Soc.* 2018, 140, 43, 14097–14111].

One of the major challenges my PhD sought to overcome was developing techniques to study extremely fast processes, such as energy transport, at extremely small length-scales. To overcome the spatio-temporal bottleneck, we developed a new technique called

super-resolution femtosecond transient absorption microscopy (see Figure), which tracks excitation (heat, charges, spins etc.) migration with sub-10 fs time resolution and sub-10 nm spatial precision. We applied this technique to biochemically- and technologically-relevant organic semiconductor materials, to show that long-range energy transport can be achieved at room temperature through a new light-matter coupling mechanism [*Nat. Comm.* 12, 6519 (2021)]. This allows energy to be transported to distances of ~300 nm and at velocities of $\sim 5 \times 10^6$ m s⁻¹, before being harvested. Placed in context, the energy migration lengths are three times larger than any of the current best semiconductors and two orders of magnitude faster.

After my PhD I did one year as an EPSRC Doctoral Prize Fellow before taking up a Junior Research Fellowship at Clare College in April 2021. I currently spend my time between Cambridge and the École Normale Supérieure in Paris where I am working with Sylvain Gigan and Hilton Barbosa de Aguiar, as well as the group of Alexis Grimaud and Jean-Marie Tarascon (Collège de France), to develop and apply methods for operando imaging

in highly scattering and absorbing electrochemical systems. Specifically, we have been using high-speed Raman microscopy and optical tomography to track the vibrational modes that drive intercalation in Li-ion battery particles, the fatigue mechanisms and timescales in energy storage devices, and the role of different electrolytes in state-of-the-art electro-catalysts.

Much of this work has been enabled (and continues to be enabled) by the Winton Program, and specifically the collaborative community it fostered. I continue to collaborate and work with many of the colleagues associated with the Winton Programme both in the UK, France and further afield. Beyond science it has also provided me with great friendships and memories which I do, and will continue to, cherish.

ALUMNI REPORTS

Hannah Stern



Originally from New Zealand, I came to the UK to pursue a PhD as a Winton Scholar in the 2013/2014 cohort. During my PhD, supervised by Prof Sir Richard Friend, I investigated the ultrafast dynamics of molecular semiconductors towards boosting solar cell efficiencies. My research was particularly motivated by understanding the mechanism of *singlet exciton fission*, an ultrafast process that generates two triplet excitons from one photogenerated singlet exciton, on ultrafast timescales. The Winton Scholarship enabled me the freedom to follow interesting research directions that arose during my PhD, and to share the results of my research at several international conferences. During my time as PhD student I also co-founded Cavendish Inspiring Women with another Winton Scholar, Dr Sarah Morgan.

Following my graduation in 2017, I was awarded a Junior Research Fellowship at Trinity College, Cambridge. I have used the fellowship to explore new research directions from my PhD; in the initial years of my fellowships I

developed a collaboration between the Department of Chemistry and the Department of Engineering to study the photophysics of localised, single photon emitting defects in wide bandgap two-dimensional materials that are gaining in interest for use in quantum applications. As part of this project, I undertook a three-month Winton Exchange to University of California, Berkeley, where I was hosted by the group of Professor Naomi Ginsberg. This exchange gave me the opportunity to explore new techniques not available in Cambridge at the time, and broaden the collaboration. During this time I also co-founded a Cambridge University spin-off company, HexagonFab, which uses two-dimensional materials to build biosensors.

Most recently, I have been exploring the spin properties of room temperature single photon emitters in two-dimensional materials in the group of Mete Atatüre. Via optically detected magnetic resonance (ODMR) experiments, my team has been the

first to identify single spins in a two-dimensional material that are optically-addressable at room temperature. This result opens possibilities for using defects in two-dimensional materials for room-temperature quantum devices. In particular, I am looking forward to developing this spin-photon interface further, with the goal of building a room-temperature platform for future quantum communication, sensing and quantum network applications.

BERKELEY EXCHANGE

Virgil Andrei

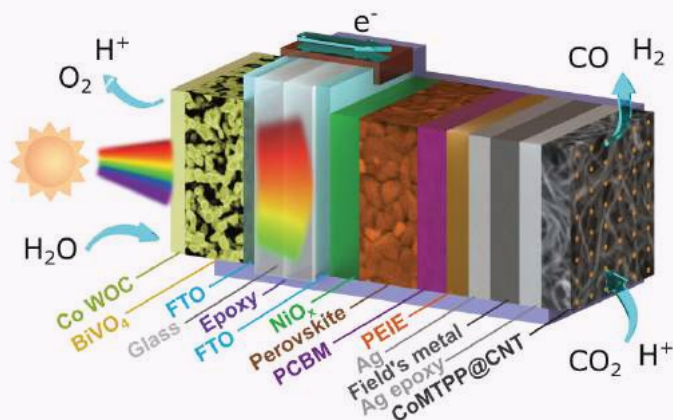
Winton-Berkeley Exchange Visitor 2022



My work focuses on developing artificial leaves, integrated panels which use sunlight to convert small building blocks into valuable solar fuels. Such tandem photo-electrochemical (PEC) devices combine a BiVO_4 photoanode for water oxidation with a perovskite photocathode for proton or CO_2 reduction. Recently, we have demonstrated that by rationally designing the device architecture, we can sustain H_2 evolution and selective CO production over several days.^[1,2] A significant increase in the moisture stability can be attained by introducing hydrophobic encapsulants and charge selective layers,^[2] whereas the selectivity can be finely controlled with an appropriate choice of electrocatalysts.^[3,4]

During my six months exchange visit to Berkeley, I will explore the (photo) electrochemical CO_2 reduction to higher value products in the group of Professor Peidong Yang. The first part of the study will focus on synthesising, characterising and optimising the performance of electrocatalyst inks towards desired multicarbon products.

In the next stage, the nanoparticle electrocatalysts will be interfaced to PEC devices for selective CO_2 reduction. This work will enable us to explore a variety



of (nanostructured) light absorbers and device configurations towards a more effective use of the solar spectrum. The project will strengthen existing research efforts in the Yang group at UC Berkeley and the Reisner group at Cambridge, and establish new collaborations, benefitting from the expertise and facilities available within the University of California, Berkeley, the Lawrence Berkeley National Laboratory, the Cavendish Laboratory and the Yusuf Hamied Department of Chemistry. The nanoparticle inks will provide a facile, scalable alternative to existing catalyst deposition techniques, which will support our ongoing efforts in Cambridge to scale-up PEC systems

towards practical applications. The overall findings on light-driven CO_2 reduction would be applicable to a wide range of photoelectrochemical systems, with the ultimate goal of contributing towards a circular carbon economy via solar fuel synthesis.

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BERKELEY EXCHANGE

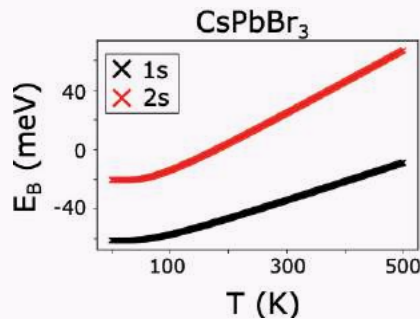
Antonios Alvertis

Winton-Berkeley Exchange Visitor 2021–2022



Exciton properties are critical to the optoelectronic response of materials, making their theoretical and computational description important to future technological devices. Many-body perturbation theory (MBPT), and specifically the *ab initio* GW-Bethe-Salpeter (BSE) equation approach, is a highly accurate method for predicting exciton properties that has been successfully applied in diverse systems.

However, recent work has shown that electronic effects alone are not enough to provide an accurate description of exciton physics. As previously shown by my co-authors and I, the effect of phonons can lead to significant renormalisation of exciton peak positions at finite temperatures^[1]. Moreover, the Neaton group at Berkeley recently showed that phonons can significantly alter electron-hole interactions in optoelectronic materials^[2]. In this project, which brings together the expertise of the Rao and Neaton groups, I am working on an extension of the GW-BSE method to account for these phonon effects as necessary. A first-principles approach based on MBPT is used, building on the framework introduced in prior work^[2] in order to rigorously implement phonon screening effects into the *ab initio* BSE



$$R_{SS'}^{ph}(\Omega) = - \sum_{cvk'c'v'k'v} A_{cvk}^{S*} g_{cc'v}(k', q) g_{vv'v}^*(k', q) A_{c'v'k'}^{S'} \left[\frac{1}{\Omega - (E_{ck} - E_{v'k'}) - \omega_{qv} + i\eta} + \frac{1}{\Omega - (E_{c'k'} - E_{vk}) - \omega_{qv} + i\eta} \right]$$

method. The method is applied to a variety of semiconductors, quantifying the role of phonon screening on exciton binding energies. Through this approach, temperature-dependent exciton properties that can be studied experimentally become accessible to computational modelling, and preliminary comparisons between the modelling and experiment show good agreement for lead-halide perovskite materials.

[1] Alvertis, A. M., Pandya, R., Muscarella, L. A., Sawhney, N., Nguyen, M., Ehrler, B., Rao, A., Friend, R. H., Chin, A. W., & Monserrat, B. (2020). Impact of exciton delocalization on exciton-vibration interactions in organic semiconductors. *Physical Review B - Condensed Matter and Materials Physics*, 102, 081122(R).

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PUMP PRIME

Eileen Nugent

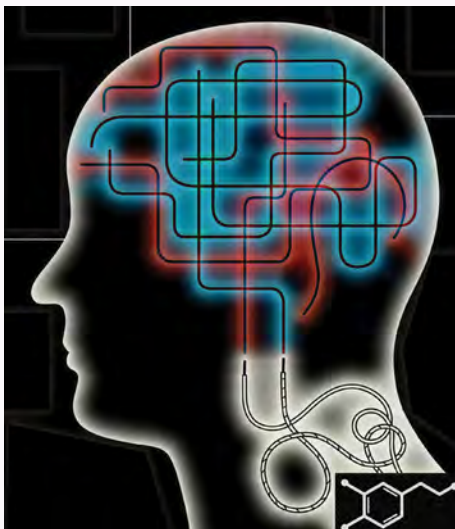
Winton Pump Prime Funding Awardee 2019



Sustainable Minds

This project combines theoretical and experimental work to understand how our mental activity is powered in the face of variable life stressors. The theoretical work offers a new perspective on the physical basis of mental health. In order to be our normal 'self' we must pay the typical energy costs of our mental activity. We draw this energy directly from ion-gradient based energy stores in networks of neurons. Mood is our sense of the energy reserve we hold to deal with fluctuations in the demand or supply of this energy. Anxiety is our sense of how often we are breaching our safety margin for this energy reserve. Failure to tightly regulate this energy reserve can result its depletion and mental illness i.e. depression, anxiety. Dysregulation can drive mental instability i.e. bipolar disorder, epilepsy etc. but may also permit periods of exceptionally high mental ability above that which we can typically achieve. Looser regulation, yielding periods of atypically high energy reserves, increases the power available for coordinated mental activity enabling us to solve more complex problems in short-lived manic states.

For mental stability, in addition to paying the costs of our current mental activity we must also predict that we can continue to do so in future. Our minds continuously



engage in a sub-conscious process of estimating their own energetic sustainability, balancing the supply of energy for mental activity - limited by our physiology - with what we predict our demands will be, as determined by our psychology and social environment. This is equivalent to continuously estimating the lifetime of the stochastic non-equilibrium process to determine the survival probability of this version of 'ourselves' in this environment. A rapid drop in the energy reserve available for mental activity can result in the loss of consciousness seen in epilepsy or, more

rarely, sudden death in epilepsy (SUDEP). If we maintain consciousness during such drops, the mind at a sub-conscious level will sense that it is dying, independent to what is happening in the rest of the body. This creates a conflict with what is known at a conscious level. If we are conscious at a point when the mind predicts no future for itself based on energetic grounds all hope will vanish. This is a point of maximum vulnerability to suicide and a medical emergency. By increasing or stabilising the energy reserve, a fraction of the total energy budget for mental activity, medicines can alter the probability of suicide.

I am working together with clinician Professor Paul Fletcher to publish this work. The experimental work will look at how molecules such as dopamine, adrenaline and calcium-handling proteins determine the power available to biological processes in living cells, and ultimately how such molecules can be assembled to form nanostructured organic energy materials *in vitro*.

The Winton pump-prime award has provided me with a vital gateway to external funding. The funds will enable me to do preliminary experimental work whilst I publish the theoretical work and apply for other funding opportunities.

PUMP PRIME

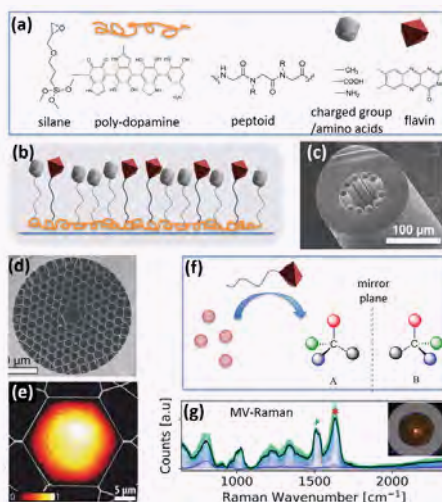
Tijmen Euser (Cavendish Laboratory, NanoPhotonics Group)
Ljiljana Fruk (Department of Chemical Engineering and Biotechnology)
Winton Pump Prime Awardees 2019



Immobilisation of enzyme-like biocatalysts in optofluidic microreactors

Natural enzymes are exceptionally well-suited to aid the manufacturing of high-value products. However, their applications have been limited to a few types of enzymes and only a handful of chemical processes they catalyse. Unfortunately, the development of *photocatalytic enzyme-like systems* that can efficiently catalyse reactions is held back by the lack of methods to explore and validate the enzyme activity.

In our Pump Prime project, we have developed new optofluidic techniques to study such reactions *in-situ*, using tiny reaction volumes. We have focused on flavin-based enzymes as inspiration to design new types of hybrid photocatalysts helpful in manufacturing processes, particularly the synthesis of novel dyes and pharmaceutical compounds [1,2]. Our work shows that hybrid compounds composed of flavin and polydopamine components can be used for the photo-triggered production of indigo dye, and such compounds are beneficial for light-triggered and in-dark reactions. To enable in-situ reaction monitoring of small volumes of the enzyme-based catalysts, we employ *optofluidic microreactors* based on *hollow-core photonic crystal fibres (HC-PCF)* [3]. HC-PCF microreactors strongly enhance light-matter interaction, enable



metre-long optical pathlengths, and allow to study of the kinetics of photo-triggered reactions on sub- μL liquid samples [3].

Our current research focuses on the surface functionalisation of flavin species onto the inner fibre walls, and we have recently attracted a Sensor CDT PhD student to further optimise this process. In addition, our pump-prime grant has helped us obtain a 1-year EPSRC Airguide Photonics Collaboration Grant funding, during which we have developed holographic mode excitation techniques [4] to probe the functionalised surfaces selectively. Finally,

the Winton programme has supported the development of new fibre-based fluorescence and Raman [5] methods that we are currently using to monitor reactions within the microreactors.

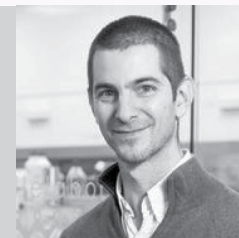
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5. A. S. Gentleman, E. Miele, *et al.*, *In-Situ Raman Spectroscopy of Reaction Products in Optofluidic Hollow-Core Fiber Microreactors*, *CLEO PR* (2020). https://doi.org/10.1364/CLEOPR.2020.C2H_2

Figure 1, above. (a-b) Silane- and polydopamine strategies are used to immobilise flavin catalysts within optofluidic microreactors. (b) Additional functional groups can improve the substrate orientation on the HC-PCF (c) surface. (d) kagomé-type HC-PCF with guided optical mode (e). (f) enantioselective enzyme-mimics. (g) example of Raman-based sensing in HC-PCF microreactors [5].

RESEARCH

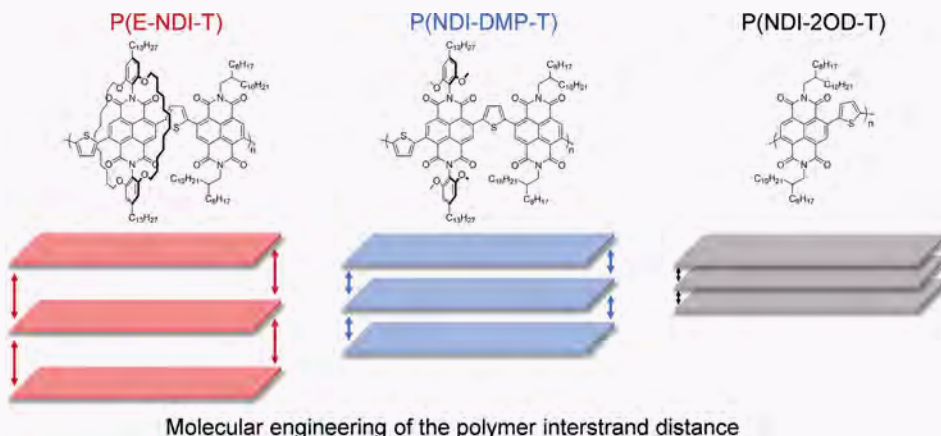
Hugo Bronstein, Yusuf Hamied Department of Chemistry



Novel conjugated materials for optoelectronic applications

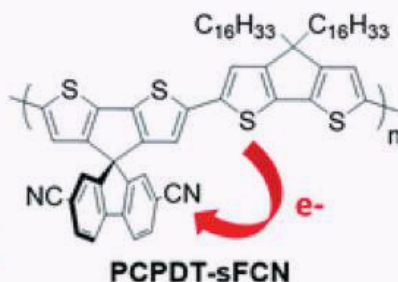
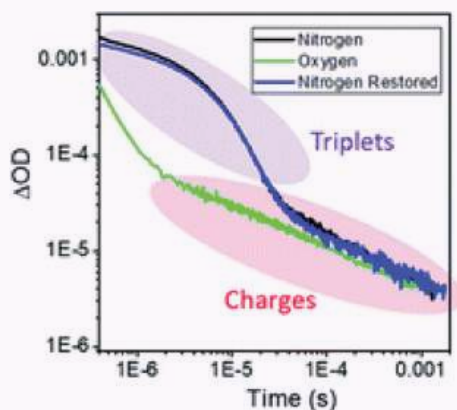
Conjugated polymers are an important class of chromophores for optoelectronic devices. Understanding and controlling their excited state properties, in particular, radiative and non-radiative recombination processes are among the greatest challenges that must be overcome.

We reported the synthesis and characterisation of a molecularly-encapsulated naphthalene diimide-based polymer, one of the most successfully-used motifs, and explored its structural and optical properties. The molecular encapsulation enables a detailed understanding of the effect of interpolymer interactions. We revealed that the non-encapsulated analogue P(NDI-2OD-T) undergoes aggregation-enhanced emission, an effect that is suppressed upon encapsulation due to an increasing π -interchain stacking distance. This suggests that decreasing π -stacking distances may be an attractive method to enhance the radiative properties of conjugated polymers in contrast to the current paradigm where it is viewed as a source of optical quenching.



Efficient charge photogeneration in conjugated polymers typically requires the presence of a second component to act as electron-acceptor. We reported a novel low band-gap conjugated polymer with a donor/orthogonal acceptor motif, referred to as PCPDT-sFCN. The role of the orthogonal acceptor is to spatially isolate the LUMO from the HOMO, allowing for negligible exchange energy between electrons in these orbitals and minimising the energy gap between singlet and triplet charge transfer states. We employed ultrafast and microsecond transient absorption spectroscopy to

demonstrate that, even in the absence of a separate electron-acceptor, PCPDT-sFCN showed efficient charge photogeneration in both pristine solution and film. This efficient charge generation is a result of an isoenergetic singlet/triplet charge transfer state equilibrium acting as a reservoir for charge carrier formation. Furthermore, clear evidence of enhanced triplet populations, which form in less than 1 ps, is observed. Using group theory, we show that this ultrafast triplet formation is due to highly efficient, quantum mechanically-allowed intersystem crossing between



the bright, initially photoexcited local singlet state and the triplet charge transfer state. Remarkably, the free charges that form via the charge transfer state are extraordinarily long-lived with millisecond lifetimes, possibly due to the stabilisation imparted by the spatial separation of PCPDT-sFCN's donor and orthogonal acceptor motifs. The efficient generation of long-lived charge carriers in a pristine polymer paves the way for single-material applications such as organic photovoltaics and photodetectors.

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RESEARCH

Dr Tom Bedford, Professor Judith Driscoll



Development of High-Temperature Superconducting Coated Conductor Tape Materials for Commercial High Field Fusion Magnets

Nuclear fusion can provide an abundant source of safe, reliable and clean energy. The 21st century has seen a renaissance in fusion research, with private companies emerging worldwide, each one working on a unique approach to the challenge of putting fusion power on the grid.

A promising approach under investigation by Tokamak Energy UK is the compact high-field spherical tokamak. These devices use powerful magnetic fields to confine a fusion plasma. Sufficiently high current densities are required to produce the strong fields required for fusion, at as high a temperature as possible in a compact system (see Figure 1[1]). The only suitable material for this purpose is a high-temperature superconductor (HTS), Rare-Earth Barium Copper Oxide or REBCO. The HTS material is in the form of coated conductor tapes which need to be wound into coils to form the magnet. The tapes are made using a highly complex process, and are formed of very thin layers of superconducting

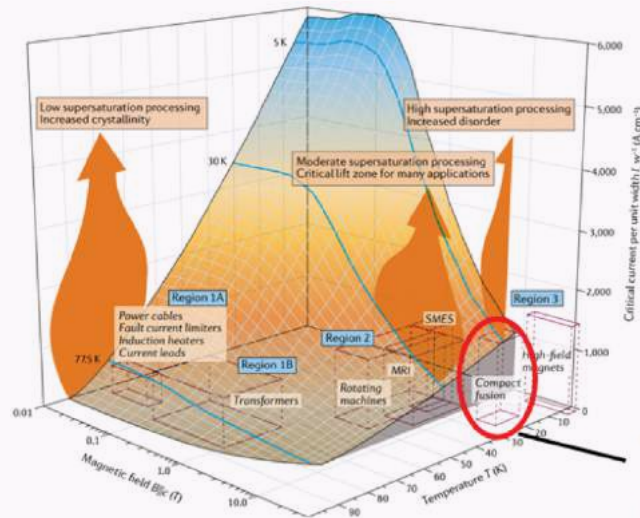


Figure 1: Temperature and Field Regimes for HTS applications¹

material deposited on metallic templates with numerous layers in between. The best performing tapes require expensive vapour deposition processes.

There are a few challenges in implementing HTS tapes in fusion magnets, namely the performance of the tapes, which is adequate but would improve the magnet performance if they

were improved further. Also the tape fabrication cost and the low current rate of world production present challenges. The performance of the tapes and production costs can be reduced using several methods which lead to higher current densities in the superconductor layer e.g. by adding second phase magnetic flux-pinning centres in the tapes. The degree of current density

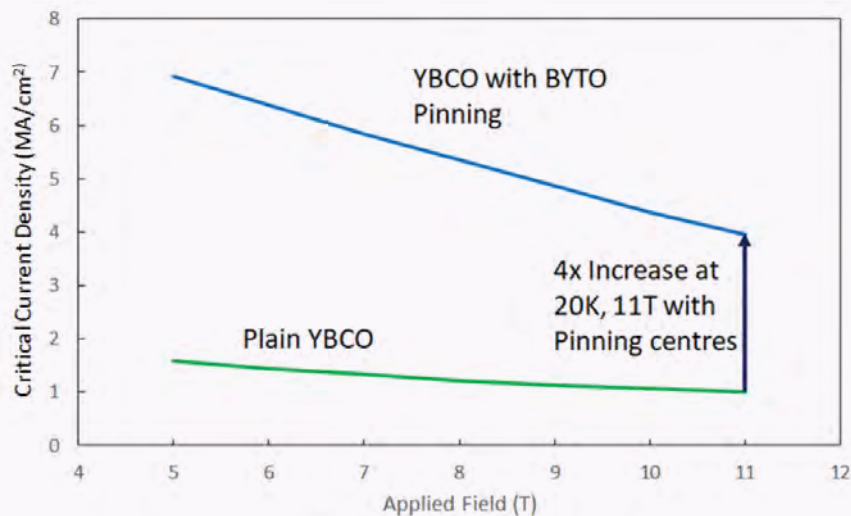


Figure 2: Jc enhancement from BYTO Pinning

enhancement depends on several factors including size, shape, density and composition. Our work sheds light on what pinning centres are optimum and how to create them. We use a highly controllable, very fine (few nanometre) addition of Ba₂YTaO₆ (BYTO) to the REBCO and achieve an impressive four-fold enhancement in current density at 20 K and 11 Tesla applied field (the highest field we are able to measure)

(Figure 2). The trend for even higher field improvements is clear.

We are currently optimising the fraction of BYTO additions to give high densities of pinning centres, suited to the high densities of magnetic flux lines which permeate the superconductor at high fields.

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<https://doi.org/10.1038/s41578-021-00290-3>

PEOPLE



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THE WINTON PROGRAMME FOR THE
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