2025 Australia-NZ Ultrafast Spectroscopy Symposium

Advances in Ultrafast Spectroscopy
13-14 February 2025

Acknowledgement of Country

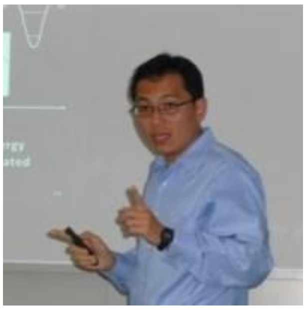
The University of Adelaide acknowledges that the Kaurna Peoples are the original custodians of the Adelaide Plains and the land on which the University's campuses at North Terrace, Waite and Roseworthy are built. We acknowledge and pay our respects to the Kaurna people, the traditional custodians whose ancestral lands we gather on. We acknowledge the deep feelings of attachment and relationship of the Kaurna people to country, and we respect and value their past, present and ongoing connection to the land and cultural beliefs.

WELCOME

Welcome from the 2025 Australia-NZ Ultrafast Spectroscopy Symposium Organising Committee

It is our great pleasure to welcome you to the 2025 Australia-NZ Ultrafast Spectroscopy Symposium. The aim of this two-day symposium is an opportunity for ultrafast spectroscopy researchers in New Zealand and Australia to connect, reconnect, and for students, postdocs, and senior researchers to present their work. The symposium will include keynote, invited and contributed talks covering both experimental and theoretical aspects of ultrafast spectroscopy.

Thank you so much for joining us: together we are delighted to welcome you to Adelaide and hope you enjoy the symposium!



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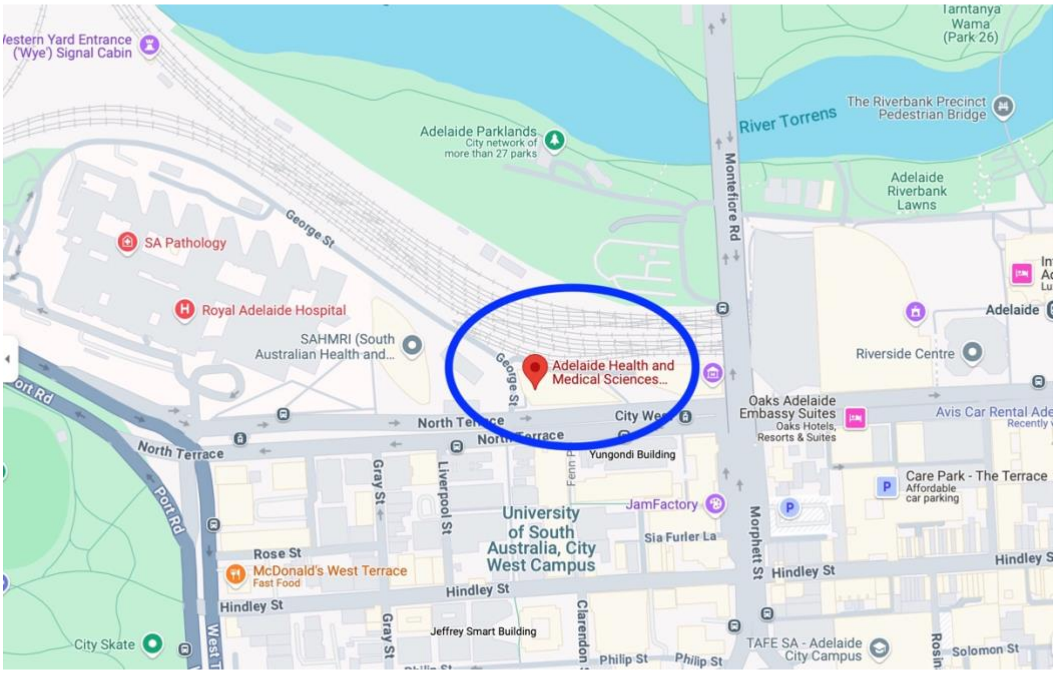
EVENT INFORMATION

Venue – Room 1059A/B, Adelaide Health and Medical Sciences (AHMS) Building, North Terrace, The University of Adelaide

The Adelaide Health and Medical Sciences (AHMS) Building is a part of The University of Adelaide's campus on North Terrace (blue highlight on map below, Google Maps). The AHMS Building is situated approximately 6 kilometres from the Adelaide Airport, and is accessible by bus, and rideshare. Paid parking is also available. The closest bus stop is Stop C1 on Currie Street, 2 blocks away from the AHMS Building. As you enter the AHMS Building, keep going straight until you see the lifts. Take the lift to go to Level 1 and Room 1059A/B is nearby.

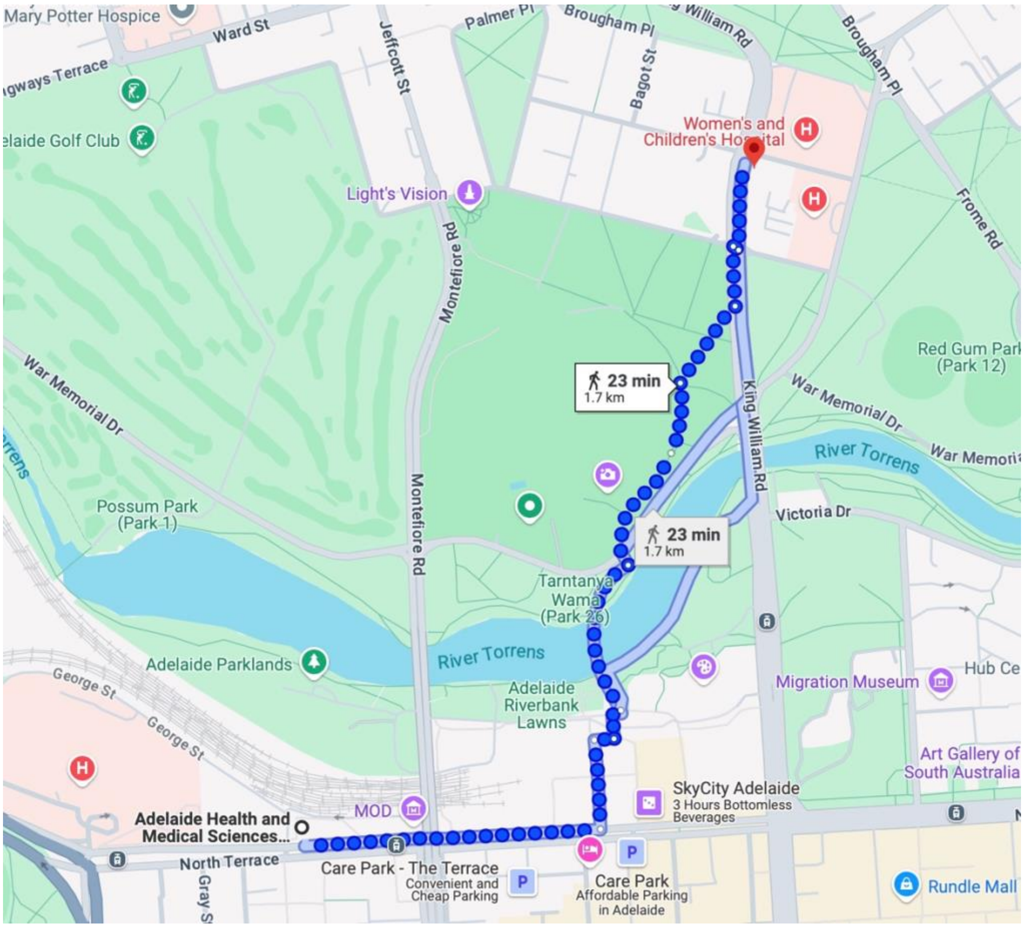


More information on the AHMS Building and other locations on the North Terrace Campus of The University of Adelaide.

AHMS Building

Conference Dinner – Cathedral Hotel 45 Kermode St, North Adelaide

The conference dinner will be held at the Cathedral Hotel at 45 Kermode St, North Adelaide on Thursday, 13 February at 6:00pm. It is a 23-minute walk from the Adelaide Health and Medical Sciences building (the conference venue). Please visit Google Maps for directions.



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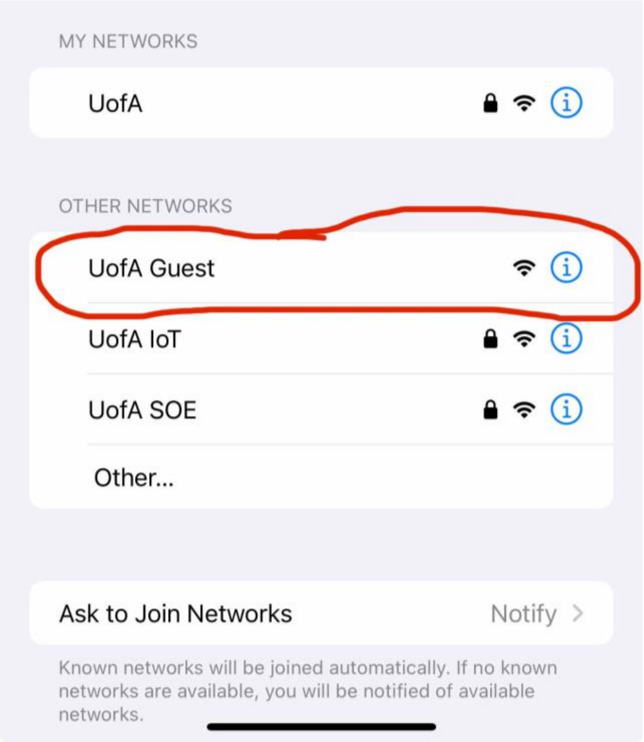

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KEYNOTE SPEAKER

Professor Stefan Maier
Monash University

INVITED SPEAKERS

Dr Alex Stuart
The University of Sydney

Prof Jeffrey Davis
Swinburne University of Technology

Dr Jack Muir
CSIRO

A/Prof Paul Shaw
The University of Queensland

A/Prof Evan Moore
The University of Queensland

Prof Timothy Schmidt
University of New South Wales

Dr Issac Etchells
The University of Queensland

Prof Yasuhiro Tachibana
RMIT University

Prof Trevor Smith
The University of Melbourne

CONTRIBUTED SPEAKERS

Simon Durst
University of Bayreuth

Zane Peterkovic
The University of Adelaide

Euca Brooks
The University of Queensland

Andrew Dolan
The University of Adelaide

Rees Hughes
The University of Melbourne

Zi Goh
The University of Adelaide

Harrison McAfee
The University of Adelaide

2025 Australia-New Zealand Ultrafast Spectroscopy Symposium

- **Duration of presentation: Keynote - 60 mins, Invited & Contributed- 30 mins.**
- **All times above include Q & A.**

Thursday 13th Feb, AHMS Bldg, Rm 1059A/B

8:30 – 9:15	**Registration**
9:15 – 9:30	**Welcome and opening remarks**

9:30 – 10:30	**Session 1** **Chair: Tak Kee, University of Adelaide**
Invited	Alex Stuart, University of Sydney *Transient absorption of organic semiconductor microcavities*
Contributed	Simon Durst, University of Bayreuth *Fluorescence-Detected Two-Dimensional Electronic Spectroscopy of a Single Molecule*

10:30 – 11:00	**Morning Tea/Coffee**

11:00 – 12:30	**Session 2** **Chair: Alex Stuart, University of Sydney**
Invited	Jeff Davis, Swinburne University *How Ultrafast Spectroscopy Can Help to Understand High-Temperature Superconductivity*
Contributed	Euca Brooks, University of Queensland *Photophysics of Y6 Crystals*
Contributed	Zane Peterkovic, University of Adelaide *Photon counting cameras for low-light biological imaging of autofluorescence using ultrafast lasers*

12:30 – 13:30	**Lunch**

13:30 – 15:15	**Session 3** **Chair: Euca Brooks, University of Queensland**
Invited	Paul Shaw, University of Queensland *Elucidating the roles of the acceptors in organic solar cells with a ternary blend*
Invited	Jack Muir, CSIRO *Butterfly Polaritons*
Contributed	Andrew Dolan, University of Adelaide *Photo-induced Singlet Dissociation in High Performance Organic Photovoltaics*
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15:15 – 15:45	**Afternoon Tea/coffee**

15:45 – 17:15	**Session 4** **Chair: Paul Shaw, University of Queensland**
Invited	Evan Moore, University of Queensland *Time Resolved Adventures in the Visible and Infra-Red*
Contributed	Rees Hughes, University of Melbourne *Origins of Trap Emission in CdSe Quantum Dots*
Contributed	Zi Goh, University of Adelaide *Size-Dependent Photocatalytic Activity of Organic Semiconducting Nanoparticles*

18:00 – 21:00	**Symposium Dinner, The Cathedral Hotel, North Adelaide** *For registrants who have RSVP'd.*

Friday 14th Feb, AHMS Bldg, Rm 1059A/B

8:30 – 9:30	**Registration**

9:30 – 10:30	**Session 5** **Chair: Tak Kee, University of Adelaide**
Keynote	Prof Stefan Maier, Monash University *Emergence and ultrafast optical modulation of optical resonances*

10:30 – 11:00	**Morning Tea/Coffee**

11:00 – 12:30	**Session 6** **Chair: Evan Moore, University of Queensland**
Invited	Timothy Schmidt, UNSW *Ultrafast Spectroscopy for Singlet Fission and Upconversion*
Contributed	Harrison McAfee, University of Adelaide *Evolving Hydrogen Gas Using Triplet Excitons of An Organic Photocatalyst*
Invited	Issac Etchells, University of Queensland *Probing Singlet Fission in Phenyl Diketopyrrolopyrrole Dimers with Transient Absorption Spectroscopy*

12:30 – 13:30	**Lunch**

13:30 – 14:30	**Session 7** **Chair: Issac Etchells, University of Queensland**
Invited	Yasuhiro Tachibana, RMIT *Charge carrier dynamics of photo-functional semiconductor films in correlation with their device performance*
Invited	Trevor Smith, University of Melbourne *Ultrafast Spectroscopy and Microscopy of Thin Film Materials*

14:30 – 14:45	**Symposium close and prize ceremony**
14:45 –	**Departure/Lab visit**

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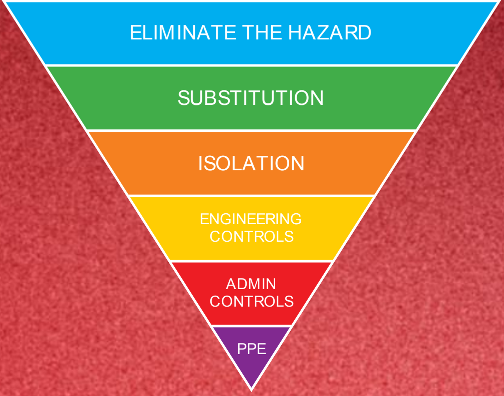

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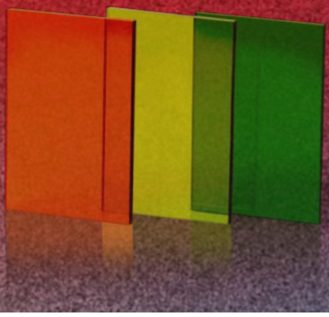

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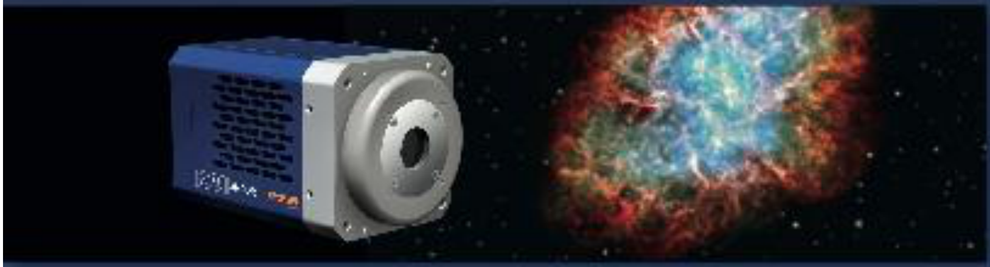

Transient absorption of organic semiconductor microcavities

Alexandra N. Stuart,[1] Yahui Tang,[1,2] Timothy van der Lann,[2] Andrew Musser,[3] Girish Lakhwani[1]

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[2]*CSIRO Linfield, NSW, Australia*
[1]*Department of Chemistry and Chemical Biology, Cornell University, NY, USA*

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Embedding molecules in optical microcavities is an emerging method of controlling excited-state processes of optoelectronic materials without synthetic modification. This has potential benefit in organic photovoltaics, where the strong light-matter coupling achieved in optical cavities could be used to assist charge formation and mitigate loss pathways. Transient absorption (TA) spectroscopy is often used to gain insight on the photophysics of organic photovoltaic systems and would be an ideal technique for determining the effect of strong-coupling on exciton dynamics. However, conducting TA spectroscopy on optical microcavities results in several challenges, such as excitonic signals being masked and large artefacts arising from changes in the cavity structure. Here I present some of the recent progress we have made in analysing the TA spectroscopy of organic photovoltaic blends inside an optical microcavity. Specifically, we study blend films of PTB7-Th:$PC_{71}BM$ cavities, which have previously been found to exhibit enhanced performance in photovoltaic devices. We use transfer matrix simulations to reproduce the cavity TA spectra, and show that, when artefacts are accounted for, there is very little difference between cavity and non-cavity photophysics.

Fluorescence-Detected Two-Dimensional Electronic Spectroscopy of a Single Molecule

Sanchayeeta Jana[1], Simon Durst[1], Markus Lippitz[1]

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Single-molecule fluorescence spectroscopy is a powerful method that avoids ensemble averaging, but its temporal resolution is limited by the fluorescence lifetime to nanoseconds at most. At the ensemble level, two-dimensional spectroscopy provides insight into ultrafast femtosecond processes, such as energy transfer and line broadening, even beyond the Fourier limit, by correlating pump and probe spectra. In this talk, I will present our method of combining these two techniques and demonstrate coherent 2D spectroscopy of individual dibenzoterrylene (DBT) molecules at room temperature. We excite the molecule in a confocal microscope with a phase-modulated train of femtosecond pulses and detect the emitted fluorescence with single-photon counting detectors. Using a phase-sensitive detection scheme, we were able to measure the nonlinear 2D spectra of most of the DBT molecules that we studied. Our method is applicable to a wide range of single emitters and opens new avenues for understanding energy transfer in single quantum objects on ultrafast time scales.

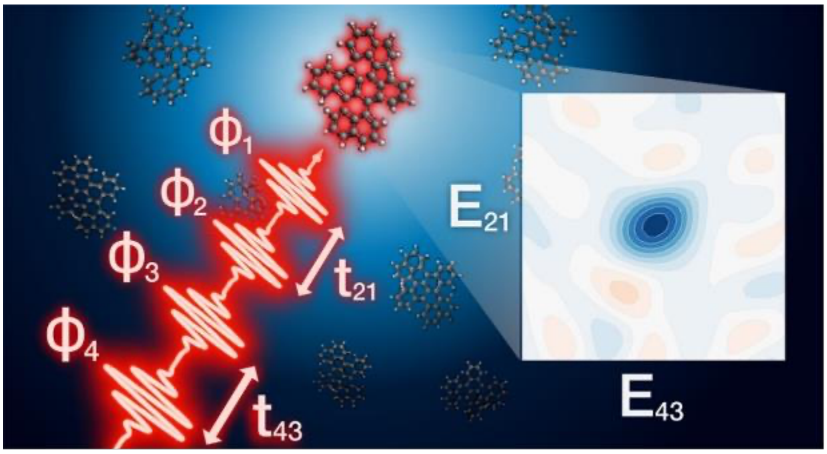

Figure 1. An illustration of our technique. A phase-modulated train of femtosecond pulses excites a single DBT molecule. By measuring the fluorescence it emits upon this excitation with phase-sensitive lock-in detection, we can extract its nonlinear 2D spectrum via Fourier transformation.

References

1. Sanchayeeta Jana, Simon Durst, and Markus Lippitz, *Nano Letters* 2024 *24* (40), 12576-12581 DOI: 10.1021/acs.nanolett.4c03559

How Ultrafast Spectroscopy Can Help to Understand High-Temperature Superconductivity

Rishabh Mishra[1], Jonathan Tollerud[1], Daniele Fausti[2], Claudio Giannetti[3], Jared Cole[4], Jeffrey Davis[1]

[1]*Optical Sciences Centre, Swinburne University of Technology, Australia*
[2]*Friedrich-Alexander-Universität Erlangen-Nürnberg, Germany*
[3]*Università Cattolica del Sacro Cuore, Brescia, Italy*
[4]*School of Science, RMIT University, Australia*

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High-temperature superconductors (HTSCs), particularly cuprates and iron-based materials, exhibit unconventional superconductivity driven by strong electron correlations and competing phases. Despite extensive research, the microscopic pairing mechanism remains elusive. By measuring the ultrafast (fs) dynamics of quasiparticles, Cooper pairs, and collective excitations in HTSCs it is possible to gain insights into the interactions among charge, spin, and lattice degrees of freedom. This in turn can reveal insights into the interplay between superconductivity, the pseudogap, and competing orders.

Using Multi-Dimensional Coherent Spectroscopy, we have recently extended the realm of ultrafast measurements to include coherent dynamics. This approach provides several advantages and new insights not possible with traditional pump-probe measurements. In this talk I will focus on measurements that were able to selectively excite Bogoliubov quasiparticles (the fundamental excitation of Cooper pairs) at the nodal and antinodal regions of the reciprocal lattice, corresponding to the regions of the superconducting gap and pseudogap. These results show enhanced coherence for the excitations at the nodal region, and point to the likelihood that superconductivity is destroyed in the pseudogap phase not because of broken Cooper pairs, but due to the increased scattering and resultant lack of macroscopic coherence.

Photophysics of Y6 Crystals

Eucalyptus Brooks [1], Paul L. Burn [1], Paul E. Shaw [1]

[1] *Centre for Photonics & Electronics (COPE), School of Chemistry and Molecularly Biosciences, The University of Queensland, Queensland, Australia.*

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Recent studies have shown that it is possible to fabricate organic photovoltaic devices comprising only Y6 as the light absorbing material.[1,2] To best optimise these devices and aid in the design of future materials, it is essential to understand the charge generation pathways present. Quantum chemical calculations have indicated that the energetic landscape of Y6 crystals could potentially lead to intrinsic generation of charges and intermolecular charge transfer (iCT) states within crystalline domains.[3,4,5] Transient absorption spectra of Y6 films have identified a complex excited state landscape comprised of Frenkel-type excitons, polarons and iCT states.[6] However, as of yet there has been no direct evidence identifying the intermolecular interactions that contribute to the origin of these states, although there is evidence that the populations of these excited states are morphology dependent.[1,2,7] In this study, we present our findings from transient absorption measurements on Y6 crystals. We identify that each of the states present in the films are also present in similar proportions in the crystals, indicating that polarons and iCT states are indeed generated intrinsically in the crystalline phases of Y6. That is, the preliminary results suggest that charge-carrier generation occurs predominantly within the Y6 crystalline domains of films.

References

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Photon counting cameras for low-light biological imaging of autofluorescence using ultrafast lasers

Z. Peterkovic[1], A. Upadhya[1], C. Perrella[1], R.B. Gonzalez[1], A. Bajraktarevic[1], M. Lim[2], K. Dunning[2] and K. Dholakia[1]

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Hyperspectral imaging of autofluorescence has been demonstrated to provide biological information regarding the metabolic health of a sample, which is relevant, for example, in the assessment of embryo viability.[1] However, the weakly emissive nature of autofluorescent proteins and the photosensitivity of many biological samples makes capturing autofluorescence a challenge. While great improvements have been made with the inclusion of imaging geometries such as light sheet microscopy, two-photon illumination, and ultrafast femtosecond lasers to reduce photodose, [2] little attention has been paid to the role of low-light photon-counting EMCCD or sCMOS cameras in this field quantitative imaging.

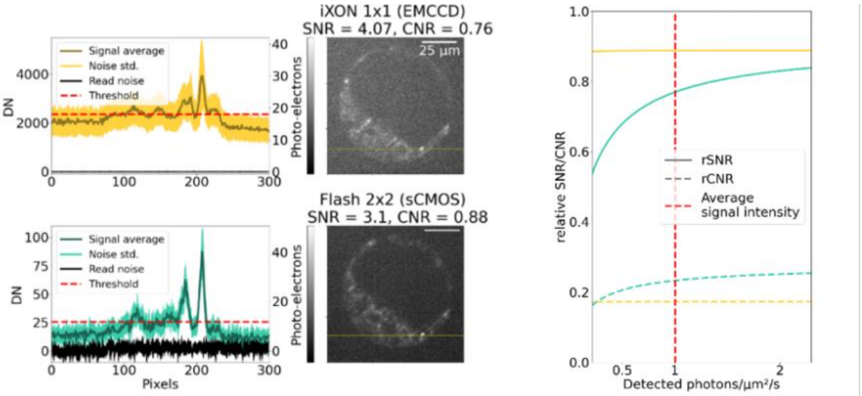

Figure 1. Noise trace comparisons comparing the autofluorescent signal captured by an EMCCD and sCMOS camera, respectively for a mouse embryo at blastocyst stage.

This study tests several such cameras, comparing the effects of operating modes, sensor noise, and camera architecture on the quality of images captured (Figure 1). We also investigate post-processing denoising algorithms, and the unexpected relationship between architecture and denoising performance. We highlight the role of pixel size in determining quality and the relationship between spatial resolution and SNR, as well as the ability of new-generation photon-counting sCMOS cameras to offer low sensor noise without electron-multiplication excess noise, in contrast to EMCCDs.[3]

References

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Elucidating the roles of the acceptors in organic solar cells with a ternary blend

Hui Jin, Neil Mallo, Guanran Zhang, Oliver Lindsay, Ronan Chu, Mile Gao, Shaun McAnally, Isaac Etchells, Paul L. Burn, Ian R. Gentle, Paul E. Shaw

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Organic solar cells (OSCs) have attracted significant attention from both the academic and industrial sectors due to their attractive features of being lightweight and structurally conformable, along with their viability for high-volume low-embedded energy manufacturing. Improvements in power conversion efficiency (PCE) can be achieved by adding a third component into a host binary bulk heterojunction (BHJ) OSC, containing a donor and an acceptor, to form a ternary device. We have synthesised a fused ring non-fullerene acceptor (o-IDT-BT-DCV) for use with a PM6:Y6 host blend in ternary organic solar cells. The exciton dissociation and charge generation processes were investigated using femtosecond transient absorption spectroscopy (fs-TAS). The results showed that o-IDT-BT-DCV in the ternary blend was no longer behaving as an acceptor, as it was in a binary blend with PM6. Instead, it contributed to charge carrier generation through a sub-picosecond energy transfer process to Y6, which then generated charge through a photoinduced hole transfer mechanism with PM6 and/or spontaneous exciton dissociation within the Y6 phase.

Butterfly Polaritons

Jack Muir[1], Kieran Hymas[1], James Quach[1], Trevor Smith[2], James Hutchison[2]

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The natural world is teeming with creatures that have evolved to manipulate light in remarkable ways. In this work, we focus on the world's largest butterflies (the Birdwings) and aim to verify if they naturally host coherent light-matter coupling in the scales of their wings.[1] The composition of the scales is a 3D lattice of ~100 nm air spheres which results in an iridescent colour (structural colour) and can be modelled as a photonic crystal. Dispersed throughout the scales are unknown "blue absorbing pigments" which absorb around 470nm and fluoresce around 550nm. These two factors are key ingredients for polaritons: organic molecules with excitons and a quantised photon mode.[2]

In this work we conducted ultrafast measurements directly on the butterfly wings, utilising transient absorption to explore if polariton physics is revealed within the excited-state dynamics. Our preliminary measurements compare the dynamics of the scales with and without a refractive index matching oil. The oil acts to fill the airholes which removes the periodic structure, thus removing the photon mode. We aim to capture the dynamics with a theoretical Tavis–Cummings–Hubbard model, which will give an estimate of the coherent coupling strength.

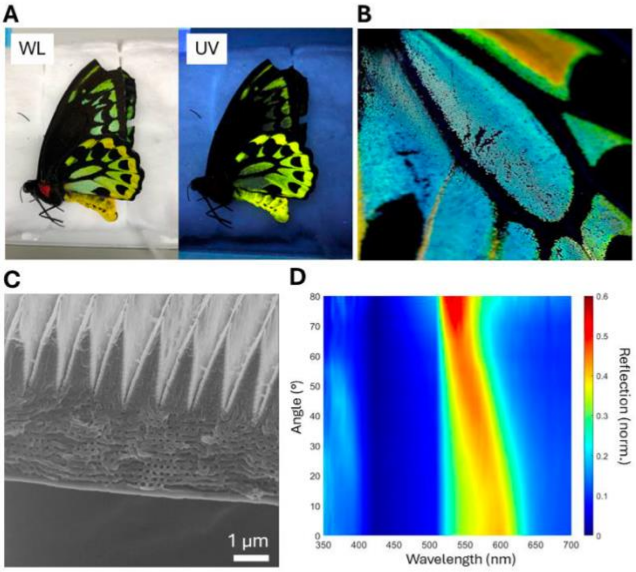

Figure 1. A Cairns Birdwing under white light and UV irradiation. **B** Image of scales. **C** STM image of the butterfly scale. **D** Angle resolved spectrum of scale with possible signature of an avoided crossing.

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Photo-induced Singlet Dissociation in High Performance Organic Photovoltaics

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Organic semiconductors have been an important class of photovoltaic materials. Current materials that may be competitive to commercial silicon solar cells come from a series of non-fullerene acceptor. These acceptors which include Y6 and L8-BO, have recently dis- played power conversion efficiencies approaching 20%, with many improvements being due to improvements in solid state packing. L8-BO is a molecular electron acceptor with its backbone structure identical to that of Y6 only altering side chains which influence solid state packing leading it to be the highest performing of the Y series acceptors. In this work, we show spectroscopic evidence of exciton to free charge generation in neat L8-BO films, similar to its analogue Y6, using the technique of pump-push-probe transient absorption spectroscopy. Furthermore, absorption signatures of charge transfer states transitioning to free charge within the neat solid-state material are presented. The comparison of singlet exciton lifetime and population in neat L8-BO, binary bulk heterojunction blends and ternary blend films reveals insight into the mechanism of achieving near 20% efficiency organic photovoltaics (OPVs). Comparing the results of the neat, binary and ternary blend films of L8-BO showed an enhancement of singlet dissociation with the addition of the ternary component. This work clarifies the spectroscopic nature of exciton and charge transfer states in the highest performing Y series acceptor, L8-BO, offering insight into how intrinsic charge generation materials may be optimally used in future OPVs.

Time Resolved Adventures in the Visible and Infra-Red

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Transient Absorption (TA) spectroscopy (or flash photolysis) has proven to be a useful pump-probe technique for investigating processes that occur on the femtosecond to microsecond timescale. Using a monochromatic laser pump pulse, synchronized with a broadband white light probe in the visible region, the time evolution of excited state populations can be monitored by tracking their Ground State Bleach (GSB) and/or Excited State Absorption (ESA).

Over the past few years, we have used this technique to investigate the sensitised lanthanide luminescence process using for a variety of organic and organometallic 4*f* metal complexes, and results from some of these studies will be discussed.

More recently, Time Resolved Infra-Red (TRIR) spectroscopy has also emerged as a powerful experimental technique. Vibrational transitions are typically much narrower (10-20 cm^{-1}) when compared to electronic transitions, are often well-separated, and can usually be associated with a characteristic functional group or bond. Hence, the time resolved evolution of these peaks can be useful for providing detailed structural information for photophysical processes and/or chemical reactions after photoexcitation.

Thanks to funding support from an ARC Linkage project (LE210100124), we have built a TRIR instrument at the UQ-PULSE labs in Brisbane, which is able to measure data on both the femtosecond and microsecond timescales. Herein, we will also report our initial results and provide a comparison of the vis-TA and TRIR spectra for a well-known metal complex.

Origins of Trap Emission in CdSe Quantum Dots

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Colloidal quantum dots (QDs) are widely utilized due to their size-dependent optoelectronic properties. Among both mature and emerging materials, trap states affect efficiencies by removing charge carriers and dominate the optical properties of core-only materials, causing broad, redshifted photoluminescence. The structural and chemical identity of these trap states remain unclear beyond their origin on the surface due a lack of direct evidence, obscured by the averaging effects of ensemble-level studies.

Here, we present our findings from the first single molecule study on the dynamics and kinetics of trap emission from individual colloidal CdSe-based quantum dots. We observe that individual QDs can exhibit both band-edge and trap-state photoluminescence over time, with non-characteristic timescales beyond a simple blinking model. Trap emission from individual emitters appears slow and monoexponential, indicating a single, long-lived trapped charge carrier configuration is responsible for trap emission. Band-edge emission is biexponential with a long lifetime tail matching that of the trap emission from the same QD, indicating that delayed emission arises from the same configuration responsible for trap state emission through a quasi equilibrium. This presents the first direct evidence of a shallow-trap model resulting in trap-state emission.

Next, we present initial findings on the fate and origin of these long-lived trap states using transient absorption spectroscopy. We track trapped charge carriers state formation (< ps) to recombination (>µs) using optical and electronic delays. We find that trapped charges persist on QDs for several microseconds in both core-only and core-shell QDs, despite differences in surface passivation and trap emission intensity. Further inducing surface defects on core-only QDs appears to accelerate this recovery, implying a competing pathway between rapidly quenching surface trap states and long-lived trap configurations that result in trap emission. These findings corroborate our previous results of trap emission originating from long-lived trap states, and emphasize the role of the surface on trying to study trap emission.

Size-Dependent Photocatalytic Activity of Organic Semiconducting Nanoparticles

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Organic semiconducting nanoparticles (NPs) have increasingly been used for photocatalytic hydrogen (H_2) generation due to their strong and broad visible light absorption, efficient charge generation, and high charge mobility. The morphology of organic semiconductors is crucial to their photophysical behaviour, and thus the performance of organic semiconductor-based devices. Unlike the organic photovoltaic (OPV) field that it draws from, the use of organic semiconducting NPs for photocatalysis is still relatively new. It is theorised that smaller NP sizes increase photocatalytic performance, as photo- generated excited states are more likely to diffuse to the surface to drive redox reactions before recombining. However, this reasoning only applies if the morphology of the semiconductor does not change significantly as NP size changes. Thus, investigating this factor can help inform further development of organic semiconducting NPs for photocatalytic H_2 production.

We report the size-dependent photocatalytic behaviour of neat Y6 NPs. Three samples of neat Y6 NPs with different diameters are prepared using the reprecipitation method and tested under photocatalytic conditions. The large NPs perform slightly better than the medium NPs, and the small NPs perform much worse than the other two. This trend is attributed to an increase in crystallinity as NP size increases, which leads to an increase in excited state mobility.

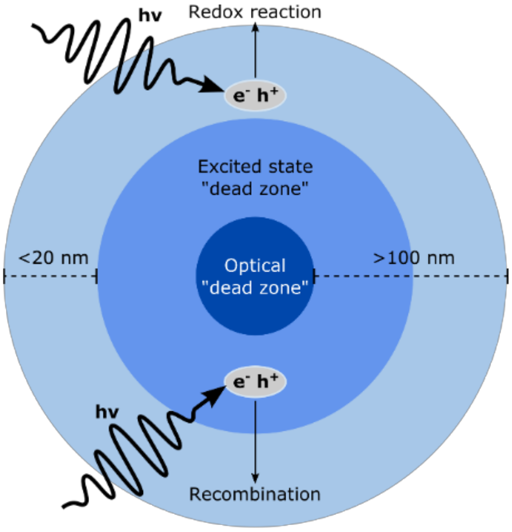

Figure 1. Schematic depiction of a nanoparticulate photocatalyst illustrating the optical and exciton "dead zones". [1]

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Emergence and ultrafast optical modulation of optical resonances

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The main part of my talk will focus on the emergence and modulation of optical resonances via ultrafast laser pulses. Specifically, I will show how permittivity-induced quasi-bound states in the continuum can be generated via optical pumping,[1] leading to spectrally sharp designer optical resonances. Judicious design of metasurface unit cells then allows for the emergence of ultrafast and sharp optical resonances. I will further demonstrate how quasi-bound states in the continuum can be induced via spatially shaped laser pulses. The second part of the talk will focus on our recent work on time-varying metasurfaces. [2, 3]

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Ultrafast Spectroscopy for Singlet Fission and Upconversion

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For more than a decade, we at UNSW and previously at The University of Sydney have been fighting with molecules to force them to do difficult things. Motivated by the desire to access higher solar energy conversion efficiencies, we have been aided by the tools of ultrafast spectroscopy in untangling the energy transfer steps required to split photon energies in two, or join the energy of two photons together. This talk will be a deranged rambling diatribe where I hope to show you a bunch of different things that were aided by our recently deceased femtosecond laser, Roxanne.[1]

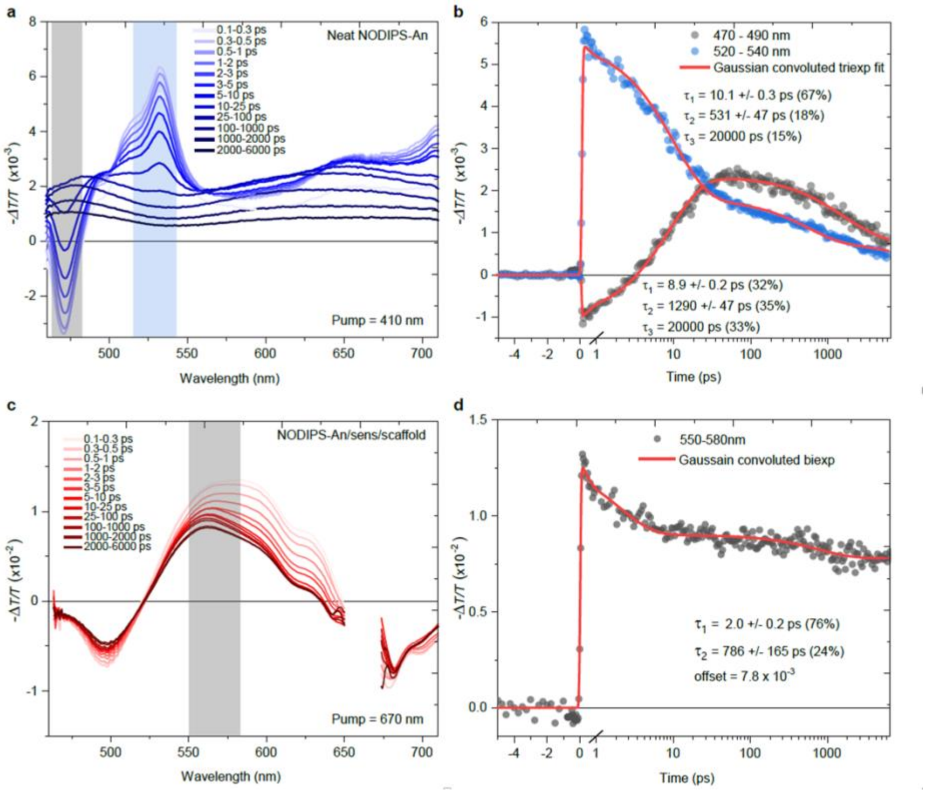

Figure 1. Transient absorption spectra of an upconversion system. **a, b)** Dynamics of NODIPS-anthracene. **c,d)** Dynamics of porphyrin sensitizer.

[1] She didn’t have to put out the red light, but she did. RIP. 2008-2023.

Evolving Hydrogen Gas Using Triplet Excitons of An Organic Photocatalyst

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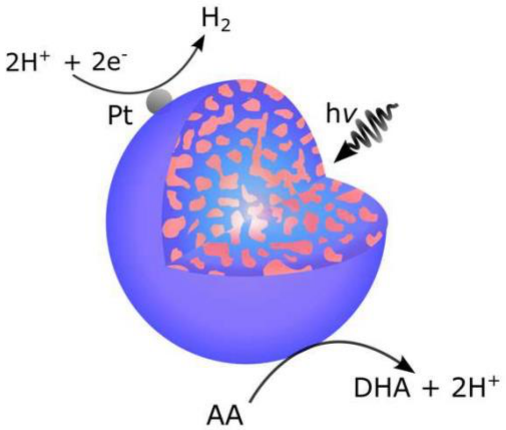
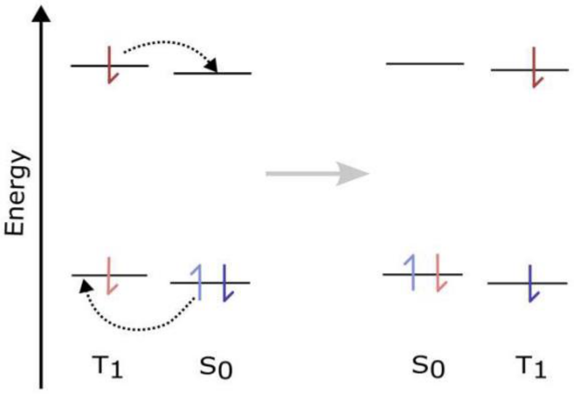
Hydrogen gas (H_2) is an important energy source that is attracting significant attention because it is a carbon-free fuel. However, the current methods of producing H_2, which are powered by fossil fuels, release a high level of carbon dioxide. A cost-effective and green alternative to current methods is photocatalytic H_2 evolution using nanoparticles of organic semiconductors. Typically, this process uses light to generate singlet excitons, which then undergo charge separation at interfaces between electron donor and acceptor materials. The generated charges can migrate to the photocatalyst's surface to facilitate redox reactions (left figure). For example, the generated electron can migrate to the surface bound platinum co-catalyst to reduce protons to form H_2. In this presentation, however, we discuss a novel approach of using triplet energy transfer (right diagram) to generate triplet excitons of an organic semiconductor to increase the efficiency of H_2 evolution. Triplet excitons are longer-lived species than singlet excitons due to their spin-forbidden relaxation to the ground state. As a result, the longer triplet exciton lifetime may allow a higher probability for charge separation than that using singlet excitons. The use of triplets could pave the way for singlet fission, a process that converts a singlet into two triplets, to be explored for maximising the efficiency of H_2 evolution in organic semiconductors.



Evolving Hydrogen Gas Using Triplet Excitons



Triplet Energy Transfer

Probing Singlet Fission in Phenyl Diketopyrrolopyrrole Dimers with Transient Absorption Spectroscopy

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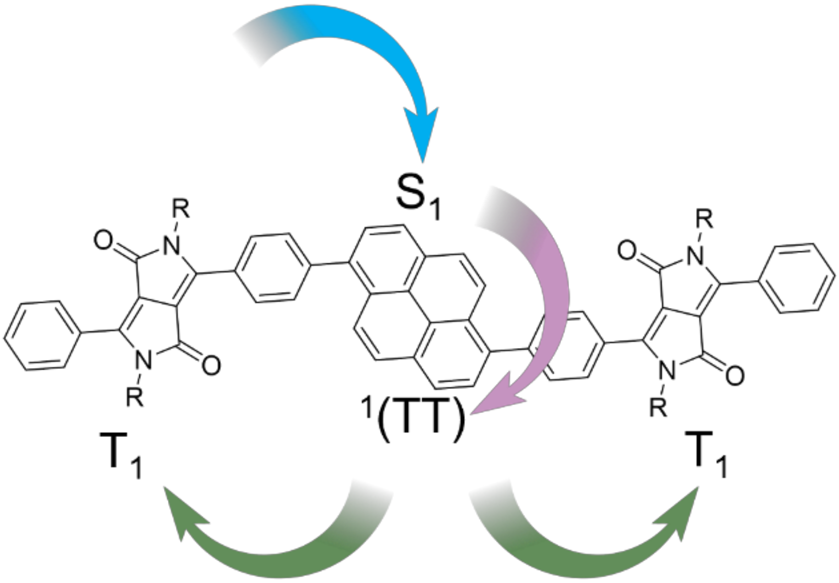
Singlet fission is a potential pathway to overcome the Shockley-Queisser limit of homojunction silicon solar cells. This down-conversion process has been well-established in thiophene diketopyrrolopyrrole (TDPP) derivatives with examples of quantitative triplet exciton yields of 200%, however, as is the case for most singlet fission systems, the T_{1}- energy of TDPP falls below the silicon band gap.[1] Conversely, phenyl diketopyrrolopyrrole (PDPP) has been shown to have a relatively higher T_1 energy but has been previously reported to fail to undergo singlet fission.[2] Herein, we report singlet fission in a pyrene bridged PDPP dimer. The combination of ultrafast transient absorption experiments in both solution and solid state have given insight into the mechanism leading to the singlet fission process.



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Charge carrier dynamics of photo-functional semiconductor films in correlation with their device performance

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The global energy transition from fossil fuels to renewable sources is well underway. Solar energy conversion is one of the most attractive processes to generate electricity (PV) or fuel (solar fuel) using sun light. Their devices function by absorbing sun light at a photo-functional semiconductor, followed by charge carrier generation and dissociation at or inside the semiconductor. PVs generate electricity by extracting the separated charge carriers at their respective electrode, while solar fuel such as hydrogen gas or alcohol is generated by reducing water or carbon dioxide with the generated and separated electrons.

Highly efficient charge transfer reactions in addition to high charge separation efficiency and swift charge transport with minimum charge recombination are required to improve their device performance. Although numerous studies were conducted to understand charge carrier dynamics at photo-functional semiconductors, their dynamics has hardly been compared with their device performance. To improve their device performance, it is of immense importance to identify underlying key parameters controlling charge carrier dynamics in correlation with the device performance. In this presentation, we will demonstrate how charge carrier dynamics at typical photo-functional semiconductors such as halide perovskite films or TiO_2 photocatalysts influences their device performance.

This work was partly supported by JSPS KAKENHI Grant (19H02813) and (22H02182), and the Collaborative Research Program of Institute for Chemical Research, Kyoto University (grant number 2023-45), Japan. We would like to acknowledge supports from the Australia-Japan Foundation for the international collaborative project. We also acknowledge supports from ARC LIEF fund (LE200100051 and LE170100235), Australia and School of Engineering in RMIT University, and Forefront Research Center, Faculty of Science at Osaka University.

Ultrafast Spectroscopy and Microscopy of Thin Film Materials

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Transient absorption (TA) spectroscopy is used to understand complex, ultrafast excitonic dynamics in materials of many types. It can be used to probe the formation and decay of intermediate species that absorb in the visible, near and mid- infrared, and teraHertz spectral regions. TA measurements can now also be recorded at micron spatial scales. Uncoupling the entangled information contributed by excited-state processes including electron-phonon coupling, different chemical components in the film, and thermal energy transport in the system requires the combination of multiple techniques. In particular, transient infrared (TrIR) spectroscopy can be used to monitor temporal changes in the intensities and frequencies of vibrational transitions that are expected to occur due to excited-state processes such as electron transfer and singlet fission. When coupled with visible/NIR TA spectroscopy, TrIR can provide the additional information required to identify the species and kinetics of complex systems.

In this talk we will illustrate the use of transient infrared (TrIR) spectroscopy to study thick film active layers based on a multi-component electron donor-acceptor system, AITC/DRTB-T-C4:L8-BO. We will also report on the use of (visible) transient absorption microscopy (TAM) measurements that allow the ultrafast spectroscopic behaviour to be mapped over thin transparent films. This has been applied to study materials including WS2 and conjugated polymer films.

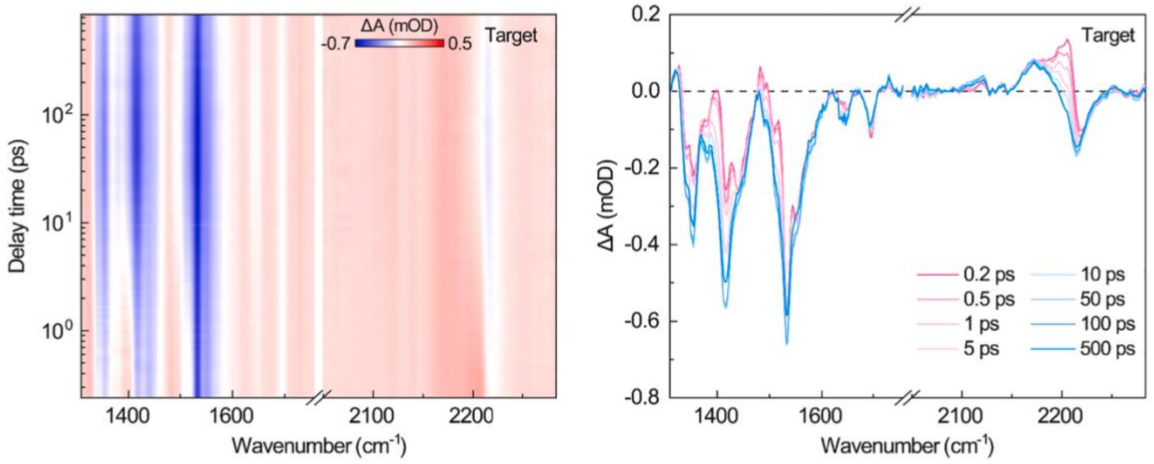

Figure 1. TRIR data for 300-nm-thick AITC/DRTB-T-C4:L8-BO blended film (pump @ 800 nm).