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September 1-5, 2025

International Conference on

Nanoscale, Single-Molecule and Related Spectroscopies (NSM)

Book of Abstracts

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MONDAY September 1

12:00 – Registration

13:45 – 14:00 **Conference Opening**

Afternoon Session I Chair: Masuo

14:00 – 14:30
Johan Hofkens: *Trapping of (single) particles: from swarming to memory applications (Invited)*
14:30 – 14:50
Atsushi Miura: *Elucidation of Molecular Orientation Behavior toward Laser-Induced Spatiotemporal Control of Protein Crystallization*
14:50 – 15:20
Ayumi Ishii: *Spin-induced Photoelectric Functions in One-dimensional Helical Materials with Organic-inorganic Hybrid Structures (Invited)*

Coffee break

Afternoon Session II Chair: Miura

15:50 – 16:10
Sadahiro Masuo: *Energy Transfer from a Perovskite Nanocrystal to Organic Dyes*
16:10 – 16:40
Arindam Chowdury: *Role of Metastable Quenchers in Determining Spatially (Un)Correlated PL Blinking of Perovskite Crystals (Invited)*
16:40 – 17:00
Shun Omagari: *Variance of the Photophysical Properties of CsPbBr3 Nanocrystals at Ensemble and Single-particle Level*

17:30 – **Welcome Reception (open to all participants)**
at Wine Cluster Hokkaido

Trapping of (single) particles: from swarming to memory applications

Johan Hofkens [1,2]

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Gaining control on particle–particle interactions and in this way on their (self)-assembled structures is essential for colloidal and material sciences. Currently, different strategies are described to achieve such control, however, all of them lack the high spatiotemporal resolution required at the micro/nanoscale, which is provided by light. In this presentation, I will discuss the possibility of creating optical matter with different colloidal materials and different irradiation regimes. On one hand, we have reported the formation of a large assembly composed by 200 nm Au NPs with dynamically fluctuating swarms by tightly focusing a laser beam at the glass/solution interface1-4. During the initial stages, an antenna-like structure with wavelength periodicity was formed inside the focus due to the optical binding. Upon increasing the NP number, this structure was further expanded outside the focus, forming a dumbbell-shaped swarming assembly, which extends up to several tens of micrometers. We proposed that the photons scattered from the trapping laser by the trapped Au NPs within the irradiated area propagate toward the NPs located outside of the focus. The expanded scattering field enables the trapping and assembling of more than 1000 NPs outside the focal spot through optical binding, yielding sub-micrometer size assemblies [1-5]

The methodology developed to image colloidal particles in an optical trap can be used to follow trajectories of particles under other force fields as well. We started investigating how sub 100 nm particles move under the influence of dielectrophoretic forces (DEP). I will show how the insights gained can in the long run result in novel types of memory devices.

References

[1] C.-H. Huang, et. al. The Primeval Optical Evolving Matter by Optical Binding Inside and Outside the Photon Beam. Nat. Commun. (2022), 13, 5325.

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Elucidation of Molecular Orientation Behavior toward Laser-Induced Spatiotemporal Control of Protein Crystallization

Atsushi Miura
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Nearly a century has passed since the successful observation of the world's first X-ray diffraction image of a protein crystal. Today, single-particle analysis and cryo-electron microscopy have enabled protein structure determination without the need for crystals, prompting some to question whether crystallization remains essential. Nevertheless, due to its relative simplicity and ability to achieve high resolution, X-ray crystallography using protein crystals continues to be widely employed, and the demand for high-quality single crystals remains strong. Although crystallization techniques have advanced significantly—shortening screening times and improving crystal formation efficiency—a universally applicable, expedited, and simple method for growing high-quality protein crystals has yet to be established. To address this need, we aim to realize spatiotemporally controlled crystallization of proteins, and are working to develop a novel crystallization methodology using laser trapping (also known as optical tweezers). In this study, we investigated laser-induced changes in local concentration and molecular orientation using *in situ* Raman and polarized Raman microspectroscopy, as a step toward achieving laser-induced orientation-regulated crystallization.

Commercially available, highly purified proteins (hen egg white lysozyme (HEWL), bovine pancreatic ribonuclease A (RNase A), α-amylase), and amino acids (glycine, phenylalanine, and so on) were dissolved in heavy water to prepare the sample solutions. For protein solutions, deuterated buffer, salts, and precipitants were added as necessary to adjust the solution conditions. A drop of each sample solution on a cover slip with ~100 μm thick was sealed in a closed cell, and a continuous-wave (CW) near-infrared laser (1064 nm) and a CW visible laser for optical tweezing and fluorescence/Raman excitation, respectively, were coaxially introduced and focused at the air–liquid interface of the droplet. Time-dependent changes in local concentration and molecular orientation under laser irradiation were monitored by polarized Raman spectroscopy, and crystallization behavior was concurrently observed. The parallel and perpendicular components of polarized Raman signals were used to calculate the depolarization ratio (ρ) that was defined based on the angle between the polarization directions of the near-infrared and visible lasers.

In all tested samples, laser irradiation resulted in increases in both Raman peak intensity and the depolarization ratio (ρ), indicating an enhancement in both local concentration near the laser focus and anisotropic molecular alignment. These results suggest that the laser-induced increase in concentration and anisotropic molecular orientation occurs regardless of molecular species or size, supporting the potential of laser trapping as a method for spatiotemporally controlled protein crystallization. In this presentation, we will also discuss the detailed molecular orientation behavior under the influence of laser electric fields, based on polarized Raman data obtained for each system.

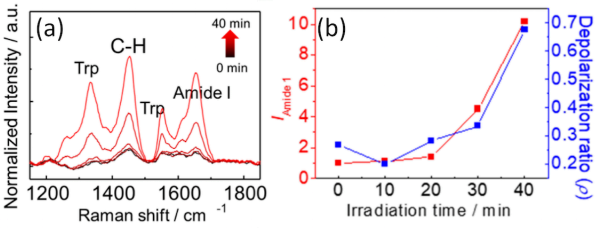

Figure. Temporal changes of (a) the Raman spectrum and (b) HEWL concentration and the depolarization ratio.

Spin-induced Photoelectric Functions in One-dimensional Helical Materials with Organic-inorganic Hybrid Structures

Ayumi Ishii[1]

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One-dimensional (1D) anisotropic crystal structures with broken spatial inversion symmetry exhibit unique spin-induced physical phenomena, attracting significant attention. This study investigates 1D helical lead halide perovskites incorporating organic chiral molecules, demonstrating controlled formation of helical and polar structures for circularly polarized light (CPL) detection [1] and bulk photovoltaic effects (BPVE) [2] (Fig.1).

The 1D structure consists of face-sharing $[PbI_6]^{4-}$ octahedral chains, surrounded by aromatic chiral cations such as *R*-(+)- and *S*-(-)-1-(1-naphtyl)ethylamine (*R*(*S*)-NEA⁺). The large π-conjugated naphthalene skeleton in NEA⁺ interacts with neighboring molecules within the crystal, significantly affecting the helicity of the $[PbI_6]^{4-}$ chains. The resulting helical 1D perovskites exhibit giant circular dichroism (CD) signals compared to reported chiral perovskites with 1D or 2D structures, which arise from their indirect electronic transition. Photodetectors of the helical 1D perovskite demonstrate highly sensitive CPL detection, achieving a high polarization extinction ratio (R_L/R_R = 25.4). The photoconductivity under CPL irradiation results from polarized electronic spin in 1D helical structures with the strong spin-orbit coupling of Pb and I.

The crystal structure as a CPL detector adopts a chiral but non-polar $P2_12_12_1$ space group, which can be transformed into a chiral and polar *C*2 space group through thermally controlled crystallization. Notably, 1D helical perovskite crystals with a chiral and polar structure exhibit an anomalous BPVE, characterized by open-circuit voltages up to 15 V—five times larger than the bandgap. This study highlights how organic chiral molecules induce chiral and polar properties of inorganic materials, offering promising applications in photovoltaics, photodetection, and spintronics.

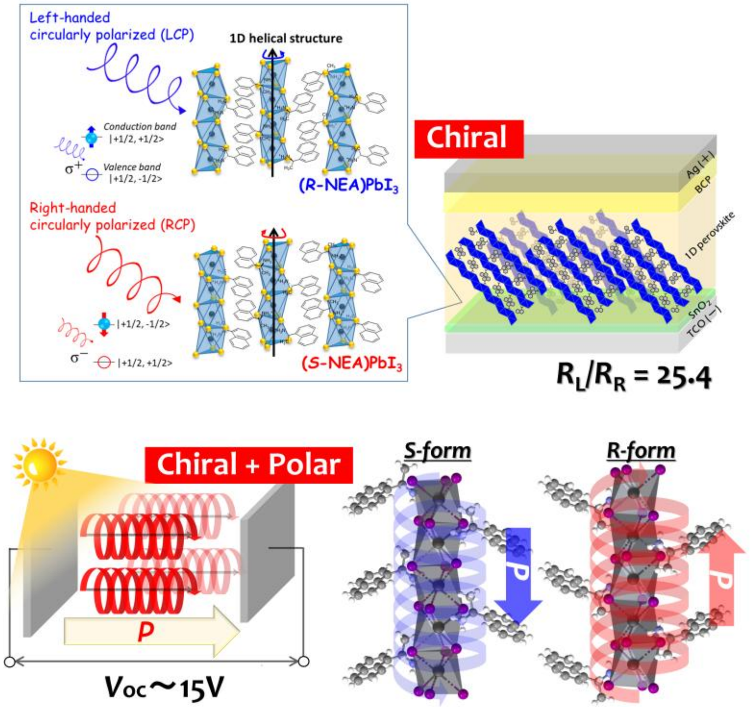

Fig. 1 Circularly polarized light detection and bulk photovoltaic properties of (*R*-NEA)$[PbI_3]$ and (*S*-NEA)$[PbI_3]$ with chiral and chiral/polar structure, respectively.

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[2] A. Ishii, et al., *Angew. Chem. Int. Ed.*, **2025**, 64, e202424391.

Energy Transfer from a Perovskite Nanocrystal to Organic Dyes

Sadahiro Masuo[1]

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Colloidal semiconductor quantum dots (QDs) are attractive materials owing to their remarkable optical properties, such as size-tunable photoluminescence (PL) wavelength, narrow PL line width, and high photodurability. With the recent development of lead halide perovskite nanocrystals (PNCs), QDs are being increasingly studied for optoelectronic applications. In addition to their PL properties, QDs—including PNCs—possess a large absorption cross-section, enabling them to readily absorb multiple photons and generate multiple excitons (MXs) within a single QD. However, MXs typically decay non-radiatively to single exciton (SX) via Auger recombination (AR), which occurs at a faster rate than the radiative rate of MXs. As a result, excitons are inefficiently consumed through AR.

As a strategy to reduce the influence of AR, extracting the excitation energy from MXs to organic molecules adsorbed on the QD surface via energy transfer (ET)—effectively converting MX energy into multiple excited states in the molecules—is a promising approach. If such ET is feasible, QDs can serve as light-harvesting moieties, paving the way for the development of various photosensitized systems. Recently, we investigated the possibility of ET from MXs in CdSe/ZnS core/shell QDs to a perylene bisimide derivative adsorbed on the QD, using photon correlation measurements.[1] This research indicated that ET from MXs was difficult to observe due to the faster AR in QDs compared to ET, suggesting that a slower AR rate could enable ET from MXs to multiple dyes. The AR rate can be reduced by increasing QD size. In this regard, PNCs are promising candidates because even those larger than their Bohr diameters retain "quantum dot-like" sharp PL spectra with high PL quantum yield. Therefore, efficient utilization of MXs is expected via ET from larger-sized PNCs to adsorbed organic dyes.

In this work, we employed lead bromide PNC-cyanine dye (Cy) linkage systems (referred to as PNC-Cy systems) to investigate the dependence of ET from MXs on PNC size. PNCs with three distinct size distributions were synthesized, and dyes were adsorbed onto the PNCs to evaluate the size dependence of ET. First, ET from the PNCs to Cy was confirmed in solution at the ensemble level. Next, ET was examined at the single-PNC level. Initially, the adsorption of multiple Cy dyes onto individual PNCs was confirmed by analyzing the emission photon statistics of Cy upon direct excitation. Then, the possibility of ET from MXs in PNCs to multiple dyes was investigated by measuring the emission photon statistics of Cy under selective excitation of the PNCs. In the case of small PNC-Cy systems, single-photon emission from Cy was observed via ET from the PNC to the dye. As the PNC size increased, the emission photon statistics of Cy transitioned from single-photon to multiphoton emission, suggesting successful ET from MXs in a single PNC to multiple Cy dyes adsorbed on its surface.

References

[1] M. Yoshioka, M. Yamauchi, N. Tamai, and S. Masuo, *Nano Lett*. 23 (2023) 11548-11554.

Role of Metastable Quenchers in Determining Spatially (Un)Correlated PL Blinking of Perovskite Crystals

Tejmani Behera, Nithin Pathoor and Arindam Chowdhury

Department of Chemistry, Indian Institute of Technology Bombay, Mumbai, India

While ubiquitous for quantum-confined luminescent nanocrystals, stochastic optical instability in terms of photoluminescence (PL) intermittency (*aka* blinking or flickering) is seldom reported for objects which extend beyond nanoscale dimensions. Interestingly, upon investigation of organo-metal halide perovskite (OHP) crystals where charge carriers are not confined, we discovered a phenomenon where entire microns-sized $(CH_3NH_3)PbBr_3$ disks/rods undergo multi-level flickering on top of a base emission [1]. Intriguingly, such photoinduced optical instability was found to be spatially-synchronous across each micro-crystal, however, emissivity fluctuations become uncorrelated beyond certain length scales (few microns) [2-3]. Moreover, we often observe such spatial correlations in flickering to vary over both space and time – being either synchronous within certain spatial and temporal windows and uncorrelated in other zones/time-scales – which makes it challenging to interpret their origins. Supported by measurements, we propose a phenomenological model which involves the activation-deactivation of very few, highly efficient metastable traps, coupled with long-range correlated carrier migration. Considering excited carriers recombine non-radiatively within a certain zone of influence of a transient quencher, we show that spatiotemporal heterogeneity, in terms of blinking events at different locations on each crystal, stems from the relative positions and quenching capability of the active traps, as well as their activation-deactivation processes. In this presentation, I will show how analyses of spatially resolved trajectories can be used to interpret the locations of few metastable quenchers each of which affect blinking events in their vicinity. This presentation will provide plausible mechanisms for the onset of spatiotemporally (in)homogeneous optical instabilities in OHP disks and rods, which may be relevant in terms of their usage as active layers in solar photovoltaic and/or light emissive devices.

References

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Synthetic-level Variability in the Photophysical Properties of CsPbBr$_3$ Nanocrystals

Shun Omagari[1], Eisuke Hirao[1], and Martin Vacha[1]

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Introduction

Cesium lead bromide (CsPbBr$_3$) nanocrystals (Fig.1) are promising luminescent materials due to their high quantum yield, tunable emission, and straightforward synthesis. However, substantial variability in their photophysical properties has been reported. The need for detailed synthesis protocols reflects the challenge of achieving reproducible CsPbBr$_3$ nanocrystal production. For instance, D. H. Son and co-workers showed that the addition of ZnBr$_2$ narrows the size distribution, yet emission quantum yields still varied from 80–95% across batches [1]. The origin of this variability remains unclear, although some studies attribute it to changes in ligand composition during post-synthetic purification [2].

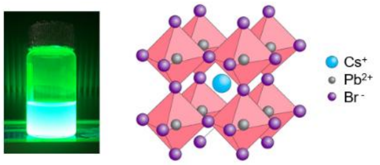

Fig.1 Luminescence of CsPbBr$_3$ nanocrystals and their crystal structure.

In this study, we examined photophysical and structural variation across nine identically synthesized CsPbBr$_3$ batches, with and without post-synthetic oleylamine treatment. Our dataset includes ensemble and single-particle emission spectra, quantum yield measurements, and TEM-based size distributions.

Results and Discussion

Post-synthetic addition of oleylamine consistently increased quantum yield, sometimes by 2-fold (Fig. 2a), in agreement with previous reports attributing such enhancement to improved surface passivation[2]. If the passivation was the sole mechanism, we would expect a corresponding increase in ON-time fraction in single-particle blinking. However, we observed little to no change in ON-time behavior (Fig. 2b), indicating that quantum yield enhancement cannot be fully explained by blinking suppression alone.

TEM analysis showed no significant difference in particle size or distribution between treated and untreated samples, which indicates that morphological changes are not responsible. Similarly, absorption edges remained consistent across all batches. Interestingly, a subset of samples exhibited spectral blueshifts following oleylamine treatment. Single-particle fluorescence imaging revealed a notable increase in emissive particle density, suggesting that the spectral shifts result not from size changes but from the activation of initially non-emissive nanocrystals.

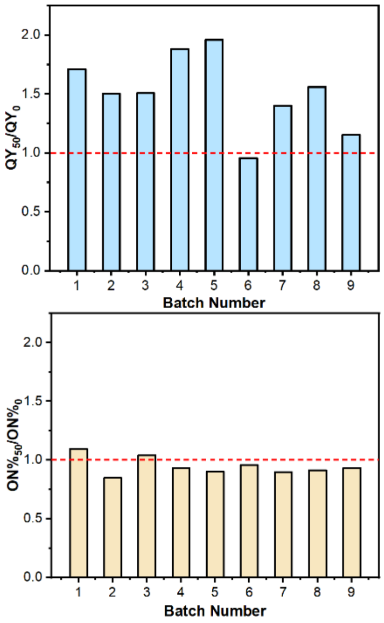

Fig.2 a) Quantum yield ratio, and b) ON-time fraction ratio. The red dotted lines represent 1.0 (no difference).

References

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TUESDAY September 2

Morning Session I Chair: Basché

9:00 – 9:45

W. E. Moerner: *Single-Molecule Detection, Spectroscopy, and Imaging Over the Years: From 1.4K to SARS-CoV-2 Nanostructures in Cells (Plenary online)*

9:45 – 10:15

Richard Hildner: *Structure-Property Relationships in Single Conjugated Co-Polymers (Invited)*

Coffee break

Morning Session II Chair: Hildner

10:45 – 11:05

Nicola Fairbairn: *Highly efficient exciton-exciton annihilation in single conjugated polymer chains*

11:05 – 11:35

Jürgen Köhler: *Natural and Biomimetic Light-Harvesting Nanotubes in the Light of Single-Molecule Spectroscopy (Invited)*

11:35 – 11:55

Syoji Ito: *Local photoexcitation of conjugated polymer solids through excitation energy transfer from single upconversion nanoparticles towards characterization of energy migration length*

Lunch

Afternoon Session I Chair: Sandoghdar

13:30 – 14:00

Kohei Imura: *Near-field imaging of plasmon modes and chiral optical fields near single gold nanoplates (Invited)*

14:00 – 14:20

Kosei Ueno: *Controlling Chemical Processes via Modulation of Electronic and Vibrational Relaxation Dynamics under Plasmonic and Polaritonic Coupling*

14:20 – 14:50

Vladimíra Petráková: *High-Yield Assembly of Plasmon-Coupled Nanodiamonds using DNA Origami for Tuned Emission (Invited)*

Coffee break

Afternoon Session II Chair: Petráková

15:10 – 15:30

Christoph Kertzscher: *Polariton-Mediated Energy Transfer Between Organic Molecules at Distances of Several µm*

15:30 – 16:00

Sabrina Jürgensen: *Collective optical states in one- and two-dimensional molecular lattices (Invited)*

Conference photo taking

16:30 – 18:30 **Poster session**

Single-Molecule Detection, Spectroscopy, and Imaging Over the Years: From 1.4K to SARS-CoV-2 Nanostructures in Cells

W. E. Moerner

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First observed optically 36 years ago in my laboratory at IBM Research, single molecules have enabled a new field of optical microscopy of the nanoscale. Since ensemble averaging is removed, each single molecule can act as a reporter of not only its position, but also of local information about the nearby environment. Combined with blinking and photoswitching (first observed at low temperatures in 1992 and then for single GFP proteins at room temperature in 1997) to ensure sparsity, in the mid-2000's, super-resolution fluorescence microscopy based on single molecules has opened up a frontier in which structures and behavior can be observed in materials and in fixed and live cells with resolutions down to 10-20 nm and below. These methods have been enhanced by PSF engineering to extract 3D position and orientation information, blinking by reversible binding (PAINT), deep learning to estimate molecular variables and structured backgrounds, light sheet illumination, and much more. A recent study shows fascinating intracellular structures formed by SARS-CoV-2 viral RNA and proteins in infected cells. As a complement to detailed nanoscale structure measurements, three-dimensional single-molecule tracking in live cells provides time-dependent information about biological regulation, condensed complexes, as well as anomalous diffusion of DNA loci in nuclei and more. Isolated single molecules in solution also have much to tell about multi-chromophore complexes, when multiple parameters are measured such as brightness, lifetime, anisotropy, emission/excitation spectra, redox potential, and much more. And beginning with optically detected ODMR in 1993, single molecules continue to have potential in the new area of molecular qubits for quantum information processing.

All of these developments in my laboratory over several decades have been enabled by an extremely talented cadre of students and postdoctoral researchers, to whom I am very grateful.

Structure-Property Relationships in Single Conjugated Co-Polymers

Erik F. Woering[1], Michael Sommer[2], Richard Hildner[1]

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[2] TU Chemnitz, Institute for Chemistry, Str. der Nationen 62, 09111 Chemnitz, Germany

Conjugated copolymers with alternating electron-accepting and -donating moieties along the backbone are an important class of functional materials for applications in e.g. organic solar cells, organic transistors, and organic thermoelectric generators [1]. In particular, blends of the conjugated copolymer PM6 and the non-fullerene acceptor Y6 are exploited in organic solar cells that feature some of the highest power conversion efficiencies to date [2]. However, the complex chemical structures and the intrinsic conformational flexibility of such conjugated macromolecules gives rise to a large degree of disorder in solutions as well as in films and blends. Clear correlations between optical and electronic properties as well as chain conformation and chemical structure (of backbone and solubilising side groups) are therefore largely elusive. Here, I will discuss results from single-molecule spectroscopy on a series of copolymers each with systematically varied chemical structure in e.g. the side groups or backbone fluorination. As a benchmark system we studied PCDTBT and showed that this prototypical copolymer is well-behaved in the sense that bulky side groups induce backbone torsion, which blue-shifts single-molecule spectra due to excited-state wavefunction localisation [3]. Comparing PM6 with fluorinated and non-fluorinated backbone on the single-chain level, we find that fluorination does not impact on the spectral position of the emission, but it does strongly reduce electron-phonon coupling to an effective carbon-bond stretch vibration, which we attribute to backbone planarisation by non-covalent interactions involving fluorine atoms, and which provides a molecular scale explanation for its efficiency in solar cells. Finally, for Y6 we are able to show that the intra-molecular charge-transfer state is highly emissive at room temperature. In contrast, in films emission is quenched at room temperature via efficient, thermally-driven formation of inter-molecular charge-transfer states. These data shed new light on the molecular scale properties of PM6 and Y6 and why blends of these materials are so efficient in solar cells.

References
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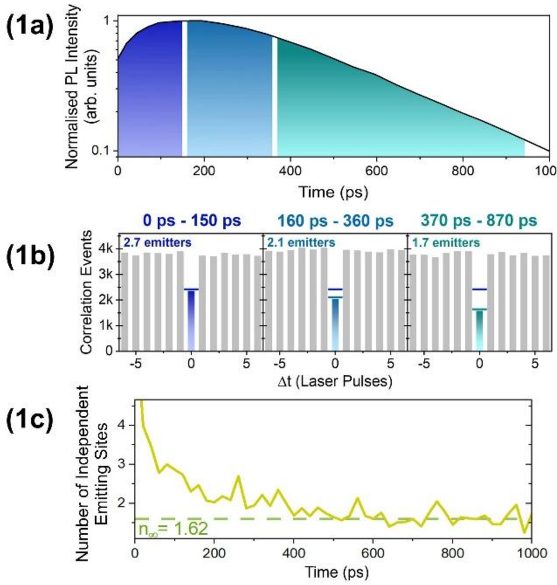
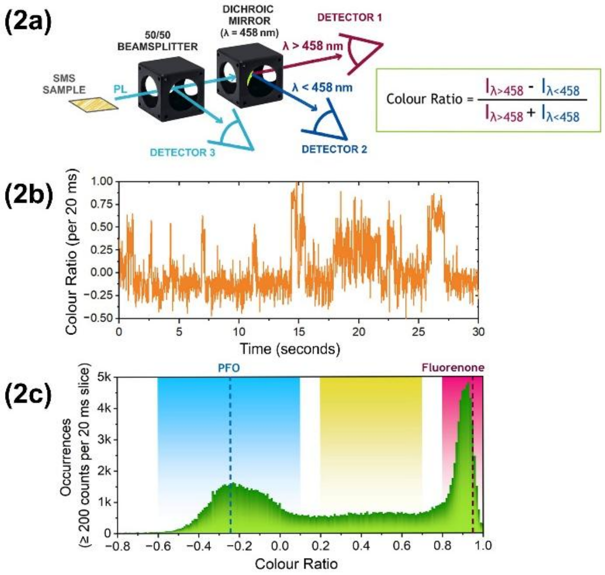
Highly efficient exciton-exciton annihilation in single conjugated polymer chains

Nicola J. Fairbairn[1], Olga Vodianova[2], Gordon J. Hedley[2]

[1]*Department of Engineering Science, The University of Electro-Communications, Tokyo, Japan*
[2]*School of Chemistry, The University of Glasgow, United Kingdom*

The unique benefit offered by single molecule spectroscopy (SMS) is that individual molecules are measured, free from ensemble averaging effects. This approach becomes particularly valuable for heterogeneous light-emitting materials such as conjugated polymers, where every individual chain can have different electronic properties. The small number of light-emitting chromophores on an individual chain allows its photon statistics to be analysed. This is particularly important as we can understand how the emitted photons are temporally distributed. Polyfluorene, (PFO), is an archetypal blue-emitting conjugated polymer incorporated into organic light-emitting diodes and organic lasers. The occurrence of exciton-exciton annihilation in PFO is a loss-mechanism for light emission that is desirable to minimise but difficult to observe free from other background signals. By diluting the PFO chains to a low concentration in a PMMA matrix, SMS measurements are made by addressing each chain consecutively with a piezo-scanning confocal fluorescence microscope. Photoluminescence (PL) is collected and focused onto two detectors in a Hanbury Brown & Twiss (HBT) geometry. Time-correlated single-photon counting is used to measure the PL decay (Fig. 1a) and generate photon antibunching graphs for determining how many independent emitting sites exist in the chain. Here, we have used novel picosecond time-resolved antibunching (psTRAB)[1] to monitor the varying rate of exciton-exciton annihilation in single chains of PFO. By altering the time window that the antibunching graph is constructed from using psTRAB, different degrees of photon antibunching are observed in PFO, indicating that the number of independent emitters is reducing with time – a signature of annihilation (Fig. 1b and 1c). We further tailor the psTRAB experiment to measure the colour ratio of incoming photons (Fig. 2a). This allows us to monitor spectral fluctuations in single PFO chains (Fig. 2b), construct a colour ratio population histogram by calculating psTRAB from spectrally filtered regions (Fig. 2c) and explore how spectral position alters annihilation.





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Natural and Biomimetic Light-Harvesting Nanotubes in the Light of Single-Molecule Spectroscopy

Jürgen Köhler[1]

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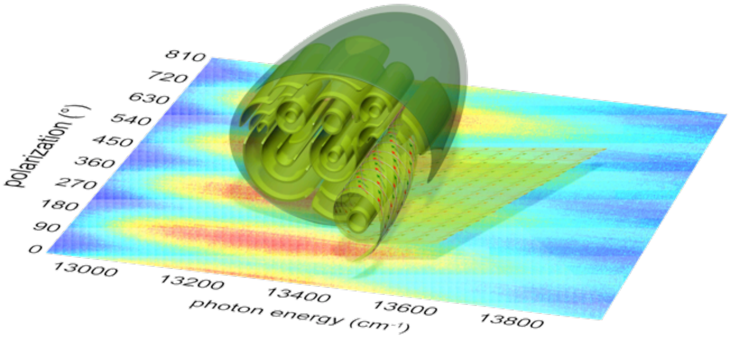
The light-harvesting apparatus of green sulfur bacteria - so called chlorosomes - is highly efficient and allows these species to grow photosynthetically under extremely low light conditions. The chlorosomes contain supramolecular structures comprising hundreds of thousands of bacteriochlorophyll molecules, and details of the structural organization of the molecular aggregates are the subject of ongoing debates. Since the electronic energies and the photophysical properties of a molecular aggregate are imposed by its geometrical structure, spectral information obtained can be compared with predictions derived from structural models. Thereby, the single-particle approach is particularly suited for the elucidation of specific, distinctive spectral features that are key for a particular model structure, and that would not be observable in ensemble-averaged spectra due to the heterogeneity of the biological objects. In other words, spectroscopy can search for the presence (or lack) of spectral signatures that are characteristic of specific structural features and thereby test whether a proposed structural model is compatible with the experimentally observed spectra. The talk will discuss these topics on the example of natural chlorosomes as well as biomimetic light-harvesting nanotubes.



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Local photoexcitation of conjugated polymer solids through excitation energy transfer from single upconversion nanoparticles towards characterization of energy migration length

Syoji Ito[1,2], Hirotaka Kageyama[1], Bhagya Lakshmi S. B.[1], Hikaru Sotome[1], Ali Eftekhari,[3] Aude Bouchet,[3] and Michel Sliwa[3,4]

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Excitation energy transfer (EET) plays a fundamental role in light-energy conversion across many photo-responsive molecular systems. To date, researchers have extensively studied EET dynamics using time-resolved spectroscopic techniques, yielding ensemble-averaged timescales across various systems. However, in real molecular solids, the rates and distances of EET often vary with position due to inherent inhomogeneities in intermolecular spacing and molecular orientation. While directly measuring spatial diffusion of excitation energy in molecular solids is generally difficult because of the diffraction limit in optical detection. In this study, we visualized excitation energy diffusion within a conjugated polymer solid by employing upconversion nanoparticles as nanometer-scale excitation sources in combination with a localization technique.

Films of MEH-PPV were excited at the nanoscale via excitation energy transfer (EET) from individual upconverting nanoparticles (UCNPs) containing thulium (Tm) ions as emitters and ytterbium (Yb) ions as sensitizers. These Tm-doped UCNPs embedded in the MEH-PPV films were photoexcited using a continuous-wave (CW) laser at 976 nm. The Yb sensitizers absorbed the 976-nm light, facilitating a stepwise multiphoton energy transfer to the Tm ions, which resulted in the formation of excited states with sub-millisecond lifetimes. The long-lived visible emission from the excited Tm ions enabled localized excitation of MEH-PPV through EET from individual TmUCNPs.

We verified the occurrence of EET from the UCNPs by employing spectrally resolved single-particle imaging and single-particle emission dynamics measurements using a hyperspectral confocal microscope. By analyzing the spectrally separated luminescence images of individual TmUCNPs and the surrounding MEH-PPV using localization techniques, we were able to determine both the precise positions of the UCNPs and the spatial distribution of emissive trap sites within the MEH-PPV matrix. Based on the two-dimensional spatial mapping of these emissive sites, the EET length in the MEH-PPV film was quantitatively estimated to range from several nanometers up to approximately 50 nm.

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Near-field imaging of plasmon modes and chiral optical fields near single gold nanoplates

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Plasmons excited in noble metal nanoparticles have been extensively studied because of their potential applications such as chemical sensing, nanoscale imaging, and photochemical reactions. For these applications, amplified optical field induced by plasmons are utilized. Plasmons also amplify chiral optical field near the nanoparticle and are promising for enantio-selective reactions and sensing. Plasmonic optical field is closely related to plasmon mode excited. Hence, to utilize the plasmons for the advanced applications, understanding of plasmon mode and chiral optical field is indispensable. We have developed various near-field imaging methods to reveal spectral and spatial features of plasmons.[1-3] In this study, we applied the near-field methods to visualize plasmon modes and chiral optical fields near a gold nanoplate.[4]

Chemically synthesized gold nanoplates were dispersed on a glass substrate. Aperture-type near-field optical microscope was used to examine optical properties of samples. Gold nanoplate was locally illuminated through an aperture of the near-field probe and transmitted light was detected after dispersing by the monochromator. For chiral optical field imaging, a quarter waveplate and a polarizer were installed in prior to the monochromator for selective detection of circularly polarized components.

Figure 1(a) shows near-field transmission image of a gold nanoplate. In near-field optical microscopy, photonic local density-of-states is visualized. Based on theoretical calculations, the observed image is assigned to plasmon mode excited. Figure 1(b,c) shows chiral optical fields visualized for the identical gold nanoplate. Spatial feature is not identical with that observed in Fig. 1(a). We also found that the spatial distribution of chiral optical field is dependent on the incident field. To reveal origin of the spatial feature visualized, we performed electromagnetic simulations and found that the chiral optical field is originated from in-plane polarized plasmon mode nearly resonantly excited. We also performed three-dimensional near-field imaging and found that the spatial extension of the chiral optical field is larger than that of the optical near-field. We unraveled from theoretical calculations that the difference originates from spatial properties of electric and magnetic fields near the nanoplate. Details will be discussed at the conference.

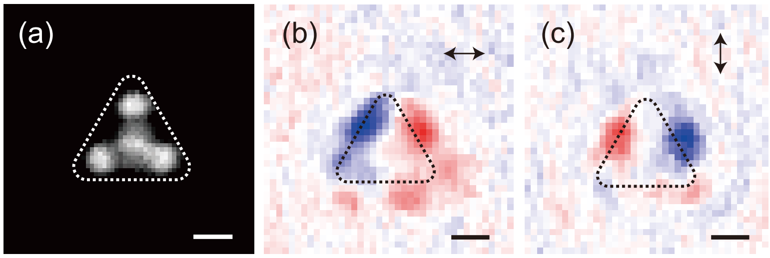

Figure 1. Near-field transmission (a) and chiral optical field (b,c) images near gold nanoplate. Scale bars: 200 nm.

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Controlling Chemical Processes via Modulation of Electronic and Vibrational Relaxation Dynamics under Plasmonic and Polaritonic Coupling

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In recent years, the exploration of strong light–matter interactions—particularly between optical resonators or localized surface plasmons and molecular electronic or vibrational states—has opened new frontiers in controlling chemical reactivity. This interdisciplinary field, known as polariton chemistry, is attracting growing attention for its potential to tailor reaction pathways, selectivity, and charge transport properties at the quantum level. Our group has been engaged in this area since its early stages, proposing that modulation of photochemical and electronic dynamics arises not only from the Purcell effect, which modifies radiative and non-radiative decay rates, but also from coherent quantum phenomena intrinsic to hybrid light–matter systems. While most studies focus on strong coupling, the high concentration required often hinders quantitative interpretation of the underlying physicochemical mechanisms. To address this, we have also investigated weak coupling regimes at lower concentrations, aiming to construct a more comprehensive understanding of how light–matter interactions govern molecular dynamics.

We have conducted systematic experimental studies under both strong and weak coupling conditions using tailored nanostructured platforms. For strong coupling, hybrid systems of merocyanine-doped PMMA films or J-aggregated porphyrins deposited on silver or gold nanoblock arrays were fabricated via electron-beam lithography and lift-off processes. Photoreactivity and dephasing dynamics were investigated using high-resolution micro-spectroscopy and interferometric femtosecond pump–probe spectroscopy. In the weak coupling regime, merocyanine in 1 mM acetone solution was studied in the presence of dispersed gold nanoparticles, with steady-state absorption and sub-picosecond transient absorption spectroscopy used to trace the photoisomerization dynamics. To probe vibrational weak coupling in the mid-infrared, gold nanochain structures with IR plasmon resonances were fabricated, and vibrational relaxation of eosin Y and out-of-plane breathing modes of bilayer MoS_2 on a microcavity substrate were analyzed using PL spectroscopy and coherent vibrational measurements, respectively.

Our results reveal a complex interplay between photonic/plasmonic coupling and excited-state dynamics. Under strong coupling, merocyanine photoisomerization was significantly suppressed, though high chromophore concentrations limited mechanistic insight. For porphyrin J-aggregates, we observed prolonged dephasing times, with ongoing studies addressing corresponding population dynamics. In the weak coupling regime, similar suppression of photoisomerization was observed, alongside a two-fold acceleration of the fast decay component in ground-state bleach recovery when gold nanoparticles were present. Concurrently, fluorescence was enhanced twofold, suggesting accelerated excited-state relaxation via the Purcell effect, leading to increased radiative decay and reduced non-radiative pathways. In the mid-infrared vibrational coupling regime, vibrational dephasing times were shortened, indicating enhanced energy dissipation consistent with the infrared Purcell effect. These findings demonstrate that both strong and weak plasmonic or polaritonic coupling regimes provide versatile means to modulate electronic and vibrational relaxation dynamics, enabling controlled manipulation of chemical reactions and processes.

High-Yield Assembly of Plasmon-Coupled Nanodiamonds using DNA Origami for Tuned Emission

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Controlling the spatial arrangement of optically active elements is crucial for the advancement of engineered photonic systems. Color centers in nanodiamonds offer unique advantages for quantum sensing and information processing; however, their integration into complex optical architectures is limited by challenges in precise and reproducible positioning, as well as efficient coupling. DNA origami provides an elegant solution, as demonstrated by recent studies that showcase the nanoscale positioning of fluorescent nanodiamonds and plasmonic gold nanoparticles. Here, we present a scalable and robust method for covalently functionalizing nanodiamonds with DNA, enabling a high-yield and spatially controlled assembly of diamond and gold nanoparticles onto DNA origami. By precisely controlling the interparticle spacing, we reveal the distance-dependent modulation of a nitrogen-vacancy center photoluminescence. We find a 10-fold increase in the fastest decay pathway at short interparticle distances. Our findings indicate selective plasmon-driven effects and an interplay between radiative and non-radiative processes. This work overcomes key limitations in current nanodiamond assembly strategies and provides insights into engineering NV photoluminescence by plasmonic coupling. These advancements bring us closer to quantum photonic and sensing applications.

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Polariton-Mediated Energy Transfer Between Organic Molecules at Distances of Several µm

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ABSTRACT

Strong light-matter coupling between an exciton and the mode of an optical resonator leads to the formation of hybrid light-matter states called polaritons which possess new characteristics found neither in the original exciton or optical mode. Consequently, certain material properties might undergo significant changes. One such example is polariton-mediated energy transfer [1-4]: If two different molecules (a donor and an acceptor) are coupled to the same microcavity, it has been shown recently that a new polariton based energy transfer mechanism can emerge bridging distances much larger than conceivable *via* Förster- (dipole-dipole interaction) or Dexter-type (electronic exchange) transfer mechanisms, which are inherently short-ranged. For the measurements we will present at this conference, we used two different J-aggregates (organic dye molecules with favorable optical properties due to supramolecular self-organization [5]) as energy donor and acceptor, respectively, embedded in a tunable Fabry-Pérot resonator, with controllable mirror distances up to several micrometers [6]. Transmission spectra show the expected splitting of the absorption peaks, indicating polariton formation. Fitting to a phenomenological Hamiltonian reveals the achieved coupling energies and cavity distances. Fluorescence measurements suggest efficient energy transfer from the donor to the acceptor molecules at distances of several µm.

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Collective optical states in one- and two-dimensional molecular lattices

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Nanomaterials such as two-dimensional layers and one-dimensional nanotubes can act as templates and containers for the formation of highly ordered molecular lattices in one or two dimensions. These ordered but not covalently bound molecular systems show fascinating optical properties, arising from the Coulomb coupling of the molecules' transition dipole moments which result in a delocalized collective optical state. We investigate the formation of well-ordered one-dimensional molecular chains inside nanotubes [1] and two-dimensional lattices on atomically flat two-dimensional substrates. [2] Using organic α-sexithiophene, MePTCDI (perylene derivate) and phthalocyanine molecules, we show the formation of large ordered domains of collective coupled molecules, as well as the change in the optical response of the molecules upon the coupling of their transition dipole moments. It is characteristic for collective states to have a strong and narrow emission, shifted emission/absorption, and a vanishing Stokes shift.

Using different spectroscopic methods we study how the interaction of the different organic molecules with the one- and two-dimensional hosts and substrates induce hybridized states with focus on changes in the molecule-related response. Further, we study a bundle of boron nitride nanotubes (BNNT) that contain molecules that build up chains. The difference in the optical response of single- and multi-file filled tubes gives us the opportunity to investigate the optical response of the bundle yielding unexpected strong red shifts for tubes with small inner diameter that are only filled with one chain of molecules.

The high transition energy tunability of the molecular systems makes them promising candidates for components in future optoelectronic devices and for analytic spectroscopy.

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WEDNESDAY September 3

Morning Session I Chair: Scheblykin

9:00 – 9:30
Masaru Kuno: *Widefield mid-infrared photothermal heterodyne imaging (Invited)*
9:30 – 9:50
Satoru Fujiyoshi: *Cryogenic Fluorescence Nanoscopy of Individual Molecules using Three-Dimension Camera System*
9:50 – 10:20
Mikako Ogawa: *Cancer therapy based on light-induced chemical structural transformation (Invited)*

Coffee break

Morning Session II Chair: Lauret

10:50 – 11:10
Linda Biondelli: *Local environment influence on mid-infrared photothermal contrast*
11:10 – 11:40
Hugo Levy-Falk: *Hybrid Interfaces for Sensing and Control in Fluorescent Organic Molecules (Invited)*
11:40 – 12:00
Tsuyoshi Fukaminato: *Efficient photoswitchable fluorescent nanoparticles: Design and development*

Lunch (International Committee meeting)

Afternoon Session I Chair: Kuno

13:30 – 14:00
Yoshihiko Kanemitsu: *Single quantum dot spectroscopy of halide perovskite nanocrystals (Invited)*
14:00 – 14:20
Hanses Niklas: *Enhancing Molecule Detection in Localization Microscopy through False Positive Probability Selection*
14:20 – 14:50
Xiaoyong Wang: *Up-Converted Photoluminescence from a Single Perovskite Nanocrystal at Cryogenic Temperatures (Invited)*

Coffee break

Afternoon Session II Chair: Wang

15:10 – 15:30
Nithin Pathoor: *Enhanced exciton diffusion in anthracene-based supramolecular fibers and perovskite micro-disks*
15:30 – 16:00
Ivan Scheblykin: *“Digitization” Effects in Luminescence of Micro- and Nanostructured Semiconductors (Invited)*
16:00 – 16:20
Marc Bröckel: *Probing the Mesomeric Effect and its Influence on the Molecular Electron Density via Surface-Enhanced Raman Spectroscopy*

19:00 – **Conference Dinner**

Widefield mid-infrared photothermal heterodyne imaging

Masaru Kuno[1,2]

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This talk will describe recent work we have conducted to develop a superresolution, mid-infrared absorption microscopy. Called infrared photothermal heterodyne imaging (IR-PHI), the technique is a new superresolution microscopy approach that offers a mid-infrared spatial resolution of order 300 nm which is significantly below the mid-infrared diffraction limit. IR-PHI has been demonstrated on a number of model systems including individual polymer nanoparticles and perovskite thin films. More recent work we have conducted has introduced a widefield modality to the technique. The new IR-PHI variant is called widefield IR-PHI (wIR-PHI) and is capable of large area hyperspectral imaging and spectroscopy. The technique also has a temporal resolution of approximately 100 ns, enabling it to monitor transient species. In whole, these photothermal-based superresolution techniques open the door to highly sensitive and chemically specific measurements of local material properties.

Cryogenic Fluorescence Nanoscopy of Individual Molecules using Three-Dimension Camara System

Satoru Fujiyoshi and Kanta Naruse

Department of Physics, Institute of Science Tokyo

In living cells, numerous biomolecules regulate biological functions. For example, a single mammalian cell contains approximately 10^9 proteins, which interact through covalent and noncovalent bonds to form large, complex three-dimensional (3D) networks that enable diverse and sophisticated cellular functions. To elucidate the functional roles of these networks, it is critical to visualize 3D spatial orgnizations of individual biomolecules throughout the entire cellular volume. Far-field fluorescence microscopy under cryogenic conditions has emerged as promising approaches for single-molecule whole-cell imaging. Although the resolution of fluorescence microscopy was long constrained by the diffraction limit, far from molecular dimensions, the localization precision (a measure of reproducibility) of individual fluorophores in cryogenic fluorescence microscopy can reach the angstrom scale [1]. However, the localization accuracy (a measure of trueness) in the axial dimension was limited to several tens of nanometers [2]. In this presentation, we introduce a 3D localization microscope with nanometer accuracy, minimizing three major source of localization errors: (1) photo-blinking, (2) dipole orientation effect, and (3) background emission. To evaluate the accuracy, two fluorophores were conjugated to the end of a 17 nm double-stranded DNA (dsDNA) molecule. The measured 3D distance between the two fluorophores at 1.9 K was 20 ± 8 nm, consistent with the designed length of 17 nm.

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Cancer therapy based on light-induced chemical structural transformation

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Photoimmunotherapy (PIT) is a targeted cancer therapy using antibody and photoreactive molecule (IR700) conjugates. IR700 is a hydrophilic silicon phthalocyanine, which has large Q-band in its absorption spectrum and absorbs near infrared (NIR) light around 690 nm. After the intravenous administration of the conjugate, the tumor is irradiated by NIR light. The irradiated cancer cells rapidly undergo necrotic/immunogenic cell death in a highly selective manner.

In photodynamic therapy, singlet oxygen production is a key for cancer cell injury. Upon absorption of near-infrared light, IR700 transitions from its excited singlet state to a triplet state via intersystem crossing, and like porphyrins for PDT, it can transfer energy to oxygen to generate singlet oxygen. However, in PIT, cell damage still occurs even when singlet oxygen is scavenged. When the antibody–IR700 conjugate is internalized into cells, cell injury can occur through the generation of singlet oxygen upon photoexcitation of IR700, as in PDT, but the extent of damage is relatively limited.

We have previously investigated the mechanism underlying cell membrane damage in PIT and demonstrated that the formation of aggregates composed of the drug and antigen proteins on the cell membrane via photochemical reactions leads to cellular injury (Fig. 1) [1]. Specifically, the cleavage of IR700's water-soluble axial ligands expose the hydrophobic phthalocyanine ring, resulting in the formation of insoluble aggregates. These aggregates incorporate antibody and antigen molecules, exerting irreversible stress on the cell membrane and ultimately leading to membrane disruption. As for the ligand cleavage mechanism of IR700, we have reported that upon photoexcitation, the phthalocyanine ring accepts an electron from an electron donor, forming a phthalocyanine anion radical. Subsequent coordination of a water molecule and protonation results in cleavage of the axial ligands [2].

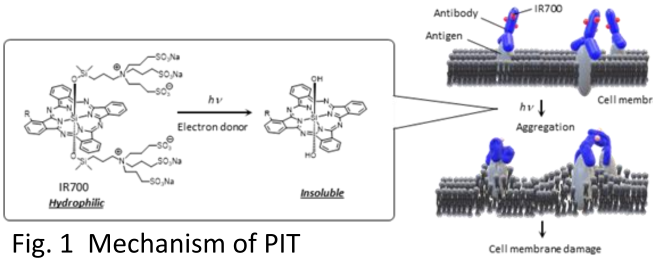

Fig. 1 Mechanism of PIT

We also investigated the activation of compounds using ionizing effects induced by hard X-rays. The energy of hard X-ray photons does not correspond to the electronic excitation energies of most molecules, making them poorly absorbed by molecules and thus highly penetrative in biological tissues. We focused on hydrated electrons generated when X-rays irradiate water. As mentioned above, IR700 produces a radical anion during the cleavage of its axial ligands. We hypothesized that if this radical anion could be generated through reaction with hydrated electrons, the ligand cleavage reaction could also be triggered by X-ray irradiation. Upon X-ray exposure, as anticipated, the axial ligands were cleaved through reaction with hydrated electrons [3]. We are currently working on the development of other compounds that can be activated by X-rays based on different chemical structures.

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Local environment influence on mid-infrared photothermal contrast

Linda Biondelli[1], Eduard Podshivaylov[2], Yang Ding[1], Umberto Filippi[3,4], Pavel Frantsuzov[2], and Masaru Kuno*[1,4]

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Mid-infrared (MIR) photothermal microscopy represents a new, ultrasensitive, superresolution infrared absorption spectroscopy[1,2]. The origin of its signal contrast is complex[3,4,5], containing competing contributions from photothermal-induced changes to specimen refractive indices and volumes. In this study, we investigate the interplay between the specimen thermo-optic and thermal expansion contributions with those of the local surrounding medium to model observed MIR contrast in photothermal microscopy images and spectra. This entails size-dependent measurements on individual polystyrene and polymethylmethacrylate nanoparticles with diameters below 200 nm and as low as 50 nm. From this we now establish that O-PTIR/IR-PHI[6] MIR contrast contains competing contributions from the thermo-optic and thermal expansion coefficients of both specimen and local environment. Comparison of acquired data to predictions of a modified Mie scattering model of O-PTIR/IR-PHI probe backscattering cross sections shows complex size- and environmental-dependencies for major MIR contrast contributors. Between $d = 188$ nm and $d < 140$ nm, a significant crossover occurs, following which the medium thermo-optic coefficient dominates that of the specimen. This aligns with existing single particle photothermal studies, conducted in the visible region of the spectrum. An important conclusion from the study then is that photothermal effects on local environment refractive indices enhance O-PTIR/IR-PHI signal contrast, paving the way for future superresolution IR absorption measurements within complex environmental matrices.

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Hybrid Interfaces for Sensing and Control in Fluorescent Organic Molecules

Hugo Levy-Falk[1], Elena Fanella[1,3], Daniele De Bernardis[1], Rocco Duquennoy[1],
Valerio Digiorgio[4], Giacomo Scalari[4], Maja Colautti[1,2], and Costanza Toninelli[1,2]

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The physics of single molecules involves complex interactions between electronic and vibrational modes. In particular, at liquid-helium temperatures, polycyclic aromatic hydrocarbons present a lifetime-limited optical transition, making them a platform of choice for the generation of undistinguishable photons and for quantum sensing [1, 2]. Our recent theoretical work on a framework for a vibronic model of molecules [3] highlights the possibility of inducing a coherent interaction using laser excitation. We particularly highlight the similarity between this system and a cavity QED system, where the electronic two-level transition plays the role of the two-level atom, and the vibrational mode corresponds to the cavity bosonic mode. The coherent population of the vibrational mode alters the fluorescence of the molecule and thus provides indirect evidence that the system enters this regime.

I will present our latest experimental results on the characterization and manipulation of vibronic coupling on a single molecule of dibenzoterrylene embedded in an anthracene nanocrystal. In particular, I will discuss the use of continuous-wave and pulsed excitation schemes in the high-saturation regime, providing insights on the cooperativity of the system and our work toward coherent excitation of vibrational modes. Additionally, I will present our efforts towards porting these techniques to engineered long-lived vibrational states, as was previously proposed in other theoretical works [4]. Finally, I will discuss the potential of this system for leveraging the vibrational modes of the molecule in order to realize THz to optical transducers.

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Efficient photoswitchable fluorescent nanoparticles: Design and development

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The field of advanced photoactive nanomaterials with photoswitching capabilities is extremely active and has attracted increasing interest in recent years due to their wide range of potential applications, including biological sensors, targeted medicine, high-resolution fluorescence imaging, and innovative materials science [1].

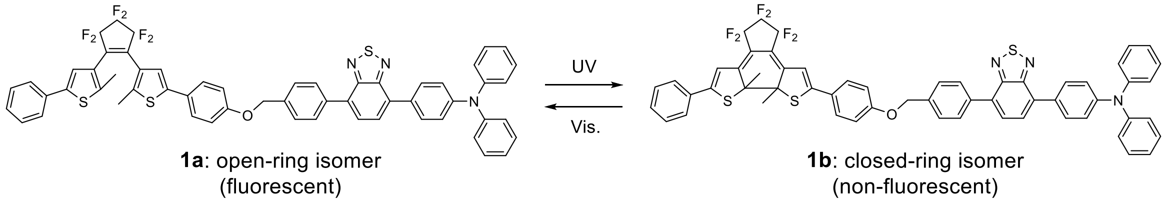

Fig. 1. Molecular structure and photochromism of a BTD-DAE dyad **1**

Recently, we successfully prepared a highly efficient fluorescent photoswitchable nanoparticle (NP), in which a photochromic diarylethene (DAE) is combined with a highly fluorescent benzothiadiazole (BTD) dye (Fig. 1) [2]. These NPs represent a state-of-the-art system, exhibiting bright red emission, reversible fluorescence photoswitching upon UV–visible irradiation, and complete ON–OFF contrast (10,000:1). Most interestingly, upon UV irradiation, the NPs exhibit complete fluorescence quenching even at a very low conversion (<5%) of the photochromic unit. This "giant amplification of fluorescence photoswitching (GAFP)" observed in both NP suspensions and individual NPs, originates from long-range intermolecular Förster resonance energy transfer (FRET) within each NP.

Although the BTD-based NPs were attractive for several applications, their photostability was insufficient. To overcome this limitation, we introduced the perylenediimide (PDI) dye as an excellent fluorophore candidate due to its bright and stable emission. The resulting DAE–PDI dyad also exhibited GAFP effect with high photostability [3]. Furthermore, to further enhance the performance of the NPs, we recently designed and synthesized new DAE–PBI dyads that exhibit much brighter emission in the NP state or incorporate multiple DAE units [4]. The detailed fluorescence and photoswitching properties will be presented in this talk.

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Single quantum dot spectroscopy of halide perovskite nanocrystals

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Halide perovskites are receiving much attention as a new class of semiconductor materials from viewpoints of the fundamental physics and optoelectronic device applications [1,2]. Their high-quality nanocrystal samples can be fabricated by low-temperature chemical solution processes. Halide perovskite nanocrystals (NCs) exhibit nearly 100% photoluminescence (PL) quantum yields without any special surface treatments, because of their inherent properties of ionic crystals. This talk discusses the optical and electronic properties of halide perovskite NCs, summarizing our group's research on the photophysical properties of halide perovskites [3-15].

Single-dot PL spectroscopy reveals the intrinsic optical properties of halide perovskite NCs. Excitons, trions, and biexcitons determine the PL spectra and dynamics of perovskite NCs [3-5]. At low temperatures, weak trion and biexciton PL peaks appear at the low energy region below the strong exciton PL peak [9-12]. The binding energies of trions and biexcitons increase with decreasing NC size [11]. Moreover, the long-range exciton-phonon interaction evaluated from the Huang-Rhys factor increases with a decrease in NC size [9,10]. The short-range exciton-phonon interaction determined from the Urbach energy also increases with a decrease in NC size [6]. Both the long-range and short-range exciton-phonon interactions increase in small NCs [13-15]. We discuss the size dependence of the exciton-exciton and exciton-phonon interactions in halide perovskite NCs.

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Enhancing Molecule Detection in Localization Microscopy through False Positive Probability Selection

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Single-molecule localization microscopy (SMLM) allows imaging beyond the diffraction limit. Detection of molecules is a crucial initial step in SMLM. False positive detections, which are not quantitatively controlled in current methods, are a source of artifacts that affect the entire SMLM analysis pipeline. Furthermore, current methods lack standardization, which hinders reproducibility. Here, we present an optimized molecule detection method which combines probabilistic thresholding with theoretically optimal filtering. The probabilistic thresholding enables control over false positive detections while optimal filtering minimizes false negatives. A theoretically optimal Poisson matched filter is used as a performance benchmark to evaluate existing filtering methods. Overall, our approach allows the detection of molecules in a robust, single-parameter and user-unbiased manner. This will minimize artifacts and enable data reproducibility in SMLM.

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Up-Converted Photoluminescence from a Single Perovskite Nanocrystal at Cryogenic Temperatures

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As an intriguing optical phenomenon in fluorescent materials with the emitted photons being higher in energy than the absorbed photons [1], up-converted photoluminescence (UC-PL) has been utilized in a variety of practical applications such as biological imaging [2], anti-counterfeit technology [3], solar energy harvesting [4], and laser cooling [5]. Here we employ resonant and near-resonant laser excitations on single lead-halide perovskite $CsPbI_3$ nanocrystals (NCs) to study their UC-PL properties mainly at the previously unexplored temperature of ~4 K. Under resonant excitation of the exciton state of a single $CsPbI_3$ NC, there appear several longitudinal optical (LO) phonon modes at the red-shifted energies from -2 to -16 meV, whose activation and emission can also be resolved from the down-converted PL excitation measurement. Surprisingly, the UC-PL of a single $CsPbI_3$ NC can still take place even at ~4 K, with the intensity decreasing monotonously as the red-shifted laser energy is detuned away from that of the exciton state. Only when a single $CsPbI_3$ NC is heated above ~10 K will the LO phonon modes be clearly resolved from the above UC-PL excitation measurement, whose thermal populations can be well described by the Bose-Einstein statistics. We attribute the intriguingly broad UC-PL excitation spectrum observed at ~4 K to the dynamic electric field created by polar LO vibrations, which is present even at zero temperature. Just as in the Franz-Keldysh electro-absorption process, this dynamic electric field gives rise to an absorption tail below the exciton state to facilitate photon absorption and the subsequent UC-PL process.

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Enhanced exciton diffusion in anthracene-based supramolecular fibers and perovskite micro-disks

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Here we present the observation of enhanced exciton diffusivity in supramolecular fibers and lead-halide perovskites. The green emitting supramolecular fibers are formed by self-assembly of the dumbbell shaped 9,10-bis(phenylethynyl)anthracene molecules (Fig. 1ai).[1] These fibers when aligned with gold nanoholes on plasmonic substrates (Fig. 1aii) show a significant increase of more than two-fold in the exciton diffusivity. Unlike surface plasmon polariton (SPP) induced leakage of waveguided emission from supramolecular nanofibers,[2] the increased carrier diffusivity and diffusion lengths observed here require the laser wavelength to fall within the SPP spectral range. This indicates that the exciton diffusion enhancement is caused by the laser-induced SPP. At the same time, the position-dependent lifetime data show that the phenomenon is caused by exciton diffusion itself as opposed to SPP propagation.

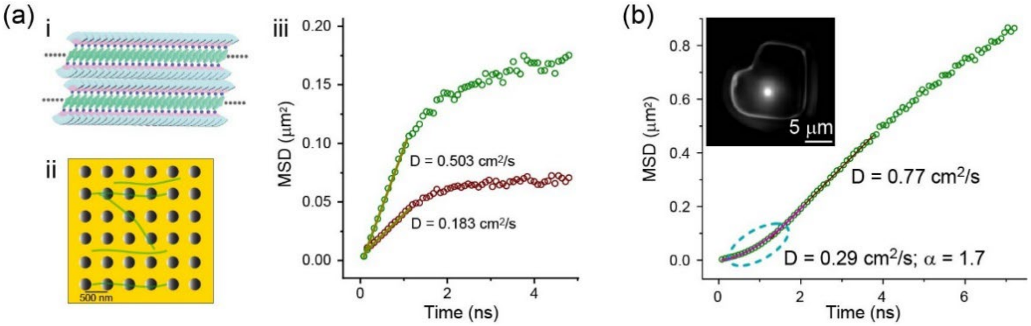

Figure 1. (a) Plasmon enhanced carrier diffusivity of anthracene-based supramolecular fibers (i) aligned with gold nano-holes (ii) of the SPP substrate. (iii) The time evolution of mean-square displacement for the fibers on gold nano-holes (green) and on glass (red). (b) The super-diffusive excited carrier propagation in $MAPbBr_3$ micro-disks.

Another observation we made is the anomalous super-diffusion of excited carriers in mesa-shaped methylammonium lead bromide ($MAPbBr_3$) micro-disks. As exemplified in Fig. 1b, the carriers get accelerated from initial diffusivity of 0.29 cm^2/s to 0.77 cm^2/s with a high truncation coefficient ($\alpha = 1.77$) under 375 nm excitation. Unlike the previously observed ballistic propagation of hot-carriers in femto-second time scale,[3] the super-diffusive non-linear behavior is sustained over few nanoseconds. The suppression of super-diffusive carrier migration under laser wavelength close to the band gap supports the involvement of hot electrons. The stabilization of hot electrons is likely related to the coupling of electronic excitation with the phonons and formation of polarons.[3] However, the exact mechanism of hot-carrier stabilization in the timescale of nanoseconds is yet to be comprehended.

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“Digitization” Effects in Luminescence of Micro- and Nanostructured Semiconductors

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Single-molecule spectroscopy, traditionally applied to nano-objects like individual molecules and quantum dots, also proves powerful for studying larger systems such as, for example, sub-micrometer semiconductor crystals.[1] Despite their size, charge dynamics in these crystals are still governed by the behaviour of only a few discrete "actors"—namely, a small number of defect states and excitations (e^-, h^+, excitons). This low-count regime renders traditional bulk semiconductor models—based on continuous variables and differential equations—inadequate. Instead, these systems exhibit what can be termed *digitization*: a regime where charge carriers, defects, and excitations are countable integers rather than averaged continuously varying quantities.[2] For instance, a small crystal either contains one excitation or none; describing it as having e.g. 0.1 excitations is physically meaningless. This digitized nature manifests in measurable phenomena such as discrete intensity variations in photoluminescence. I will discuss the consequences of this digitization on the luminescence properties of nano- and microstructured semiconductor materials. Single-molecule spectroscopy uniquely enables access to this regime, allowing us to probe charge dynamics and emission properties at the level of individual crystals—where integer statistics, not continuous distributions, dominate the physics.

Switching of metastable non-radiative centers on and off proves an effect of super-resolution in terms of the ability to interpret time-resolved luminescence decays. I will show how studying the luminescence of individual blinking sub-micrometer metal-halide perovskite crystals reveals the nature of non-radiative recombination centers in these materials.[3]

Finally, I will discuss potential applications of luminescence from micro- and nanostructures in neuromorphic optical computing, introducing the concept of *memlumors*—materials that combine memory and luminophore functions.[4]

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Probing the Mesomeric Effect and its Influence on the Molecular Electron Density via Surface-Enhanced Raman Spectroscopy

Marc Bröckel, Alfred J. Meixner and Kai Braun

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Understanding the molecular electron density, a fundamental property governing molecular interactions, plays a vital role in determining molecular structures, bonding patterns, and overall chemical reactivity. This knowledge is crucial for e.g. designing molecules with tailored properties in fields such as pharmacology, materials science, or organic optoelectronics. The most common way to tailor the molecular electron density is to introduce substituents with a mesomeric (M-) or inductive (I-) effect. Recently, we have shown via tip-enhanced Raman spectroscopy that a positive bias voltage applied to a gold tip in proximity to a molecule on a gold surface leads to a reversible electron density shift from the Au-substrate to the molecule observed as intensity increase in the corresponding TERS-spectra. [1] This positive bias voltage can be viewed as an external I-effect.

In this study, we employ surface-enhanced Raman spectroscopy (SERS) to probe the electron density of molecules from the 2-mercaptobenzothiazole (MBT) family. Introducing an ethoxy group (EMBT) or a chlorine atom (CMBT) drastically changes the molecular electron density which can be directly observed in the SERS intensity. We present a method with which it is possible to partition the total Raman intensity into atomic contributions. Combining such "atomic Raman spectra" with visualizing the electron density shifts that occur during a normal mode cycle and Weinberg's NBO analysis yields a deep understanding of the Raman intensity and allows for experimental tracking electron density shifts with atomic resolution.

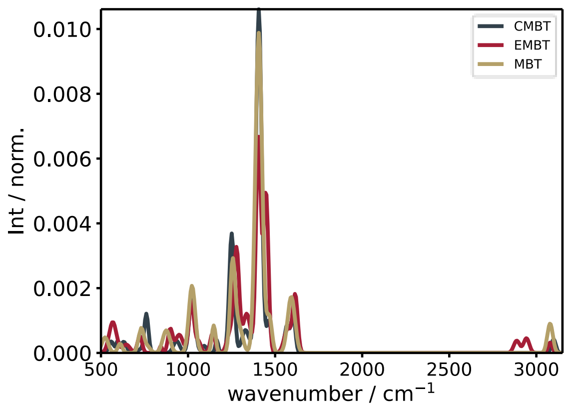

Figure 2: SERS Spectra of CMBT, EMBT and MBT. The peak intensities differ due to the different substituents

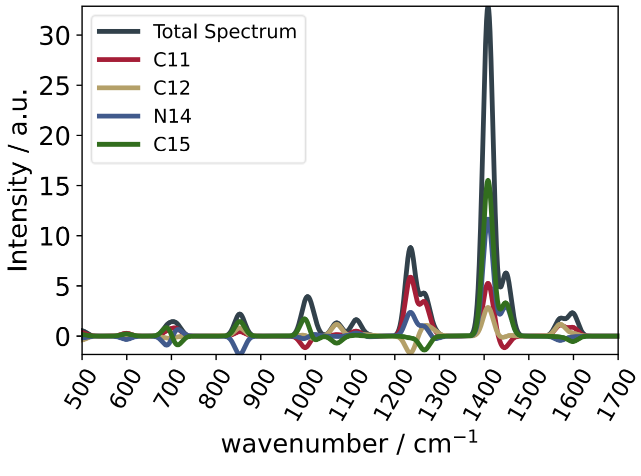

Figure 1: Calculated total SERS spectrum of MBT and the atomic contributions of atoms C11, C12, N14 and C15

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THURSDAY September 4

Morning Session I Chair: Köhler

9:30 – 10:00
Thomas Renger: *Energy Transfer in Far-Red-Light and White-Light Photosystem I (Invited)*
10:00 – 10:20
Yutaka Shibata: *Variability of energy-transfer pathway in photosynthetic pigment-protein complex revealed by single molecule excitation-emission spectroscopy*
10:20 – 10:50
Toru Kondo: *Photosynthetic light harvesting observed by single-molecule spectroscopy (Invited)*

Coffee break

Morning Session II Chair: Kondo

11:10 - 11:40
H. Peter Lu: *Single-Molecule Force Manipulation and Nanoscopic Imaging of Protein Structure-Dynamics-Function Relationship (Invited)*
11:40 – 12:10
Tjaart Krüger: *Protein aggregation studied by real-time feedback-driven single-particle tracking spectroscopy (Invited)*

Lunch

Afternoon Session I Chair: Jelezko

13:30 – 14:00
Vahid Sandoghdar: *Fourier-limited transition of single surface-adsorbed molecules (Invited)*
14:00 – 14:20
Kenji Hirai: *Directed Molecular Assembly within Optical Cavities*
14:20 – 14:50
Jean-Sébastien Lauret: *Nanographenes as single quantum emitters (Invited)*

Coffee break

Afternoon Session II Chair: Lu

15:10 – 15:30
Heyou Zhang: *Photoswitchable Nanoparticle Arrays*
15:30 – 16:00
Fedor Jelezko: *Coherent control of single color centers in diamond (Invited)*
16:00 – 16:20
Norio Murase: *Next-Generation Phosphors: Encapsulation of Quantum Dots in Silica Nanoparticles and Single Particle Analysis Insights*

Energy Transfer in Far-Red-Light and White-Light Photosystem I

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Certain cyanobacteria adopt to far-red light by incorporating a few chlorophyll *f* (Chl *f*) pigments in their photosystems. It has been controversially discussed in the literature, whether there is a Chl *f* in the reaction center (RC) of photosystem I (PSI). Although from a functional point of view, a low-energy Chl in the RC would be beneficial, earlier cryo-EM studies did not find any evidence for a Chl *f* in the RC. I will present such evidence from calculations of optical difference spectra (light-minus-dark) and note that the Rutherford group (Imperial College London, UK) has found independent evidence from cryo-EM, using an improved analysis. In the second part of my talk I will present simulations of single molecule excitation-emission spectra of white-light PS1, measured by the Shibata group (Tohoku University, Sendai) revealing a correlation between excitation and emission wavelengths under certain conditions. The simulations offer a structure-based explanation of this correlation.

References

[1] A. Name, *Journal title* volume (year) page

Variability of energy-transfer pathway in photosynthetic pigment-protein complex revealed by single molecule excitation-emission spectroscopy

Xianjun Zhang[1, 2], Joachim Seibt[3], Ryo Nagao[4], Tatsuya Tomo[5], Takumi Noguchi[6], Shen Ye[1], Thomas Renger[3], and Yutaka Shibata[1]

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Photosynthetic pigment-protein complexes realize superefficient light harvesting with the dense packing of pigment molecules. Such a pigment crowded system gives rise to an intriguing coherent delocalization of excited states over multiple pigments. The pigment crowding also brings high sensitivity of the system to slight perturbations to the pigment environment induced by conformation alterations in the protein matrix. The high sensitivity to slight conformation changes has been suggested to be related to the protective mechanism of photosynthetic antenna complexes. Though its detailed mechanism is still controversial, slight conformation changes in the antenna proteins probably switch on and off the quenching channel of exciton under excessive light conditions [1].

Here, we investigate the light-harvesting excitation energy transfer (EET) in a photosynthetic pigment-protein complex, photosystem I (PSI) based on the single-molecule excitation-emission spectroscopy. PSI binds ca. 100 chlorophyll (Chl) molecules, majority of which serve as antenna. It emits relatively strong fluorescence from the characteristic red-shifted Chls (Red Chls) at low temperatures, where the EET from the Red Chls to RC (quencher) is inhibited. The high susceptibility to conformation fluctuation has emerged in PSI as the blinking of fluorescence intensity upon single molecule fluorescence detection [2]. The blinking has been hypothesized to originate from the dynamic alteration of EET pathway: in the emitting state EET toward Red Chls is favored, whereas in the dark state that toward RC is favored [2]. To confirm the above hypothesis, one needs to reveal the EET pathway in the antenna Chls. However, single molecule detections in the fluorescence emission mode do not provide access to the antenna Chls on the upstream of the EET flow. By contrast, the excitation spectral detection gives information on the antenna Chls. Using cryogenic single-molecule excitation-emission spectroscopy, we study EET in PSI at low temperature where the protein conformation motions are frozen. The different conformations of individual PSI may result in different EET pathways, which will be reflected in the excitation-emission spectra [3]. We found a large variability in both the excitation and emission spectra of single PSIs, showing the variability in the EET pathway. We also conducted theoretical study to simulate the EET dynamics in PSI based on its known structure. It revealed variable EET pathways depending on the random fluctuations in the site energies of Chls, supporting the above hypothesis.

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Photosynthetic light harvesting observed by single-molecule spectroscopy

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Photosynthetic reactions are driven by sunlight. Light capture occurs in light-harvesting antenna complexes, followed by photoelectric conversion in reaction center (RC) proteins. The near-unity quantum yield of the photoreaction process is highly attractive, prompting extensive studies on its regulatory mechanisms. Structural information provided by X-ray crystallography and cryogenic electron microscopy enables the interpretation of spectroscopic data and the construction of theoretical models based on precise architecture. This has led to a general consensus that the conformation of biological systems responsible for the photoreaction is highly optimized at the angstrom level. However, nuclear coordinates are not static but randomly perturbed at physiological temperatures, as indicated by molecular dynamics simulations. Additionally, the conformation exhibits significant heterogeneity. These factors raise new questions about how dynamic and heterogeneous properties contribute to and optimize the photoreaction. To address this question, we have applied single-molecule spectroscopy available for the identification of small but significant conformational variations associated with inhomogeneous photophysical properties. Furthermore, combining with transient absorption spectroscopy allows us to directly investigate how conformational heterogeneity and dynamics contribute to the ultrafast photoreaction process, which is inaccessible with conventional fluorescence-based single-molecule spectroscopy. Our approaches offer insights into the importance of conformational heterogeneity in the photosynthetic light-harvesting function. In particular, it is made possible to discuss multistep energy transfer processes in chlorosomes, light-harvesting system organizing pigment aggregates without a protein scaffold.

Single-Molecule Force Manipulation and Nanoscopic Imaging of Protein Structure-Dynamics-Function Relationship

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Proteins in living cells experience thermal fluctuation and local force fluctuations. We focused our studies on understanding the structural changes in protein to compressive forces at the piconewton level. We have revealed a hidden protein property: under the piconewton compressive force, a protein tertiary structure can abruptly and spontaneously rupture like a balloon.[1-7] The threshold of the compressive force amplitude at piconewtons can be biologically available in living cells due to the thermal fluctuations, molecular crowding, and other cellular physiological conditions. We have found that this hidden property can be dependent on the ionic strength, protein surface changes, protein conformational rigidity, and solvation local environments. Single-molecule conformational manipulation provides unique methods for studying the relationship between function and structure of biomolecules, and for exploring novel properties of biomolecules under complex local environments. We have developed and applied AFM/Magnetic Tweezers force manipulation combined with single-molecule spectroscopy to study the mechanisms and dynamics of protein enzymatic reactions, crowding, signaling, and nucleosome dynamics. We have also conducted MD simulations and theoretical model analyses on our experimental discoveries. Our new approach applications have provided new knowledge of protein-crowding initiated aggregation, Tau protein-associated neurodegenerative diseases, nucleosome for epigenetics, EGFR for cell signaling, and mechanical force-activated enzymatic reaction pathways.[1-7]

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Protein aggregation studied by real-time feedback-driven single-particle tracking spectroscopy

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Single-molecule spectroscopy (SMS) has significantly advanced our understanding of the properties and dynamics of biomolecules. However, the environment used in SMS experiments is a poor representation of the natural cellular environment, and the results of these studies may consequently be of limited physiological relevance. One limitation of conventional SMS experiments is the need to immobilise the particles via surface attachment. This limitation is overcome by real-time feedback-driven single-particle tracking (RT-FD-SPT), a technique that allows spectroscopic measurements on individual, freely diffusing particles, with the added benefit of diffusion information [1, 2]. We employed RT-FD-SPT to study the aggregation of the main plant light-harvesting complex, LHCII. LHCII aggregates are generally considered a model system for the fast, reversible, energy-dependent component of non-photochemical quenching, an important photoprotective process in plants. We combined spectroscopic and diffusion information to disentangle the interplay between aggregate size, detergent concentration, fluorescence intensity, and fluorescence lifetime in these aggregates [3]. The data was complemented by fluorescence correlation spectroscopy to investigate the time-dependent aggregation of LHCII during detergent removal to quantify size-dependent singlet-triplet annihilation, a dominating process in MHz pulsed experiments that is often downplayed or overlooked [4].

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Fourier-limited transition of single surface-adsorbed molecules

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For more than 35 years, single molecules have been investigated in a variety of environments and applications. A particularly elegant line of studies was performed at liquid helium temperatures, overcoming spectral inhomogeneities and dephasing. By reaching the Fourier limit of the homogenous linewidth, single polycyclic aromatic hydrocarbons (PAHs) were shown to behave as well-defined quantum optical systems. However, so far, this exquisite performance has only been reached for PAHs *embedded* in organic crystalline matrices. In this presentation, we will quickly review the most recent advances in the context of quantum optics with PAHs and then report on the first demonstration of Fourier-limited transitions in single adsorbed molecules on the surface of an organic crystal.

Directed Molecular Assembly within Optical Cavities

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The self-assembly of molecules enables the formation of intricate architectures, as exemplified by supramolecular systems and higher-order protein structures. The key to controlling these architectures lies in fine-tuning the underlying intermolecular interactions. Recently, vibrational strong coupling (VSC) has emerged as a powerful tool for modulating such interactions.[1, 2] Employing the ability of VSC to alter intermolecular forces, we aim to integrate this approach into molecular chemistry, with a particular emphasis on molecular assemblies.

We investigate the effect of VSC of solvent molecules on the crystallization of metal–organic frameworks (MOFs). Specifically, VSC of the OH stretch in water selectively promotes the formation of ZIF-8, whereas in its absence, a mixture of ZIF-8 and ZIF-L is obtained.[3] Furthermore, we show that combining supramolecular fibers with VSC grants access to metastable states, enabling the formation of unique toroidal structures that would otherwise be inaccessible.[4] These findings underscore the potential of VSC as a versatile tool for directing molecular assembly, opening new interdisciplinary frontiers at the interface of quantum photonics and molecular chemistry.

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Nanographenes as single quantum emitters

Huynh Thanh Trung[1], Suman Sarkar[1], Océane Capelle[1], Sébastien Quistrebert[1], Hugo Levy-Falk[1], Daniel Medina-Lopez[2], Cynthia Banga-Kpakao[2], Nikos Fayard[1], Elsa Cassette[1], Stéphane Campidelli[2], and Jean-Sébastien Lauret[1]

[1] *Université Paris-Saclay, ENS Paris-Saclay, CentraleSupélec, CNRS, LuMIn, Orsay, FR*
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Nanographenes synthesized by bottom-up chemistry are tunable emitters with promises in optoelectronic, biosensing, and quantum technologies [1-2]. Recent investigations on the intrinsic properties of these nanographenes classify them as stable and bright single photon sources at room temperature [3-6]. The next step towards using nanographenes as quantum emitters is to reach lifetime-limited linewidth. When embedded in a polystyrene matrix, the spectral linewidth is reduced to ~ two meV at low temperature [6]. Despite this reduction, the linewidth is still a few orders of magnitude higher than the radiative limit, estimated to be a few µeV [7]. Inspired by pioneer works on small organic molecules [8], we designed a new guest/host system to decouple as much as possible the nanographene from its local environment [9]. This presentation will show our recent results on the low-temperature spectroscopy of new nanographenes [7] embedded in a new molecular crystal host [9]. Finally, some perspectives on using nanographenes to build arrays of entangled emitters will be presented.

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Photoswitchable Nanoparticle Arrays

Heyou Zhang[1], Pankaj Dharpure[2], Michael Philipp[1], Paul Mulvaney[3], Mukundan Thelakkat[2], and Jürgen Köhler[1]*

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[2]*Applied Functional Materials, University of Bayreuth, 95440 Bayreuth, Germany.*
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Abstract:
Recent advancements in nanophotonics have significantly transformed optical data storage and display technologies, with an increasing focus on developing materials and systems capable of dynamic, reversible control of light emission. A key challenge lies in achieving precise manipulation of the emission properties of materials at micro to nano scales across solid-state substrates.

This study introduces a approach leveraging surface-templated electrophoretic deposition (STEPD) to fabricate arrays of polymer beads incorporating photo-switchable diarylethene (DAE) molecules. These DAE molecules exhibit reversible switching between high and low emission states under direct photoexcitation, enabling precise optical modulation. The integration of these photoswitchable DAE molecules within polystyrene beads allows for individual addressing, while the STEPD assembly method ensures the formation of well-aligned photonic unit arrays with high precision and scalability.

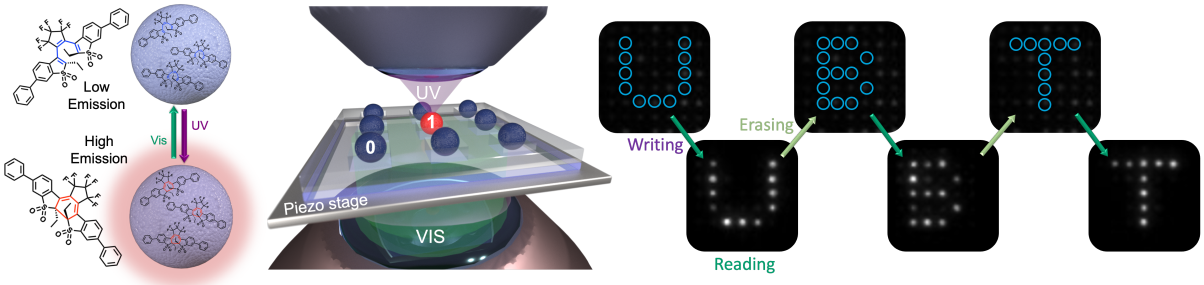

Figure 1 The molecular structure of the DAE molecules embedded in polystyrene beads with open (low-emissive) and closed (high-emissive) states. The assembled bead arrays are switched individually using focused UV excitation and read using defocused visible light illumination. A 5x5 array successfully displays the pixelated "UBT" characters, demonstrating a reversible writing-reading-erasing cycle.

Spectroscopic characterization of micro-patterned arrays was conducted to optimize critical parameters affecting on/off signal contrast and crosstalk suppression. Experimental results demonstrate that the optimized system achieves stable reversible on/off cycles in a micro-patterned array, with no significant attenuation of contrast even after 500 cycles, highlighting exceptional durability.

The research also explores binary data processing capabilities through selective writing, reading, and erasing operations within specific photonic units. This capability validates the feasibility of continuous recording and opens new possibilities for advanced optical memory devices, marking a significant step toward practical applications in optoelectronic information storage systems.

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Coherent control of single color centers in diamond

Fedor Jelezko[1],

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Single spins in diamond are attracting increasing attention due to their potential applications in quantum technologies. In this talk, we will show how dipolarly coupled qubit pairs can be generated and entanglement can be created in small quantum registers. We will also discuss optical detection of single germanium-vacancy (GeV) color centers at millikelvin temperatures, and present elements of a quantum light-matter interface based on GeV qubits.

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Next-Generation Phosphors: Encapsulation of Quantum Dots in Silica Nanoparticles and Single Particle Analysis Insights

N. Murase[1,2], T. Sawai[2], R. Mori[2], K. Inada[3], D. Eguchi[3], N. Tamai[3]

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When this conference series first began, spectroscopic insights helped establish the applicability of photochemical hole-burning for optical memory technologies. [1] The single molecule detection method emerging from this field [2] played a pivotal role in advancing spectroscopy. Although the chromophore transitioned from dyes to colloidal quantum dots (QDs), achieving robustness remains a significant challenge for widespread applications.

We have developed sol-gel derived silica glass as an optimal matrix for the encapsulation of emitting QDs in various applications.[3] Among the different approaches, silica nanoparticles with encapsulated dozens of emitting QDs have proven to be the most promising in terms of both brightness and robustness. Single-particle detection methods have clarified that the shielding effect of silica is highly dependent on the sol-gel preparation method. We have applied this sophisticated encapsulation method, initially developed for CdSe-based QDs, to InP-based QDs (InP/ZnSe/ZnS), as illustrated in the figure. In this case, the encapsulation resulted in a significant reduction in photoluminescence quantum yield. The quantum mechanical calculations together with experiments clarified that the structure of InP-QDs (core size and shell thickness) must be optimized differently from those of CdSe-based QDs. This is because of the lighter effective electron mass and lower barrier height between the InP core and the ZnSe/ZnS shells.

The silica nanoparticles we developed will be utilized in display technologies, offering bright, robust, and non-toxic features as user-friendly phosphors. These advancements will be brought to the market through the start-up we have recently established.

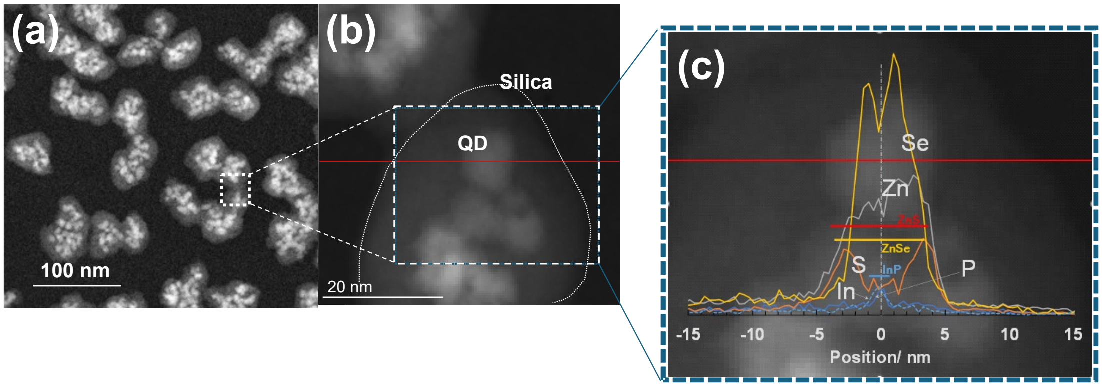

Figure (a) HAADF-STEM image of silica nanoparticles encapsulated with InP-based QDs. (b) Enlarged version of Figure (a), providing a closer examination of the encapsulated QDs. (c) EDX analysis data along the red line on the HAADF-STEM image, highlighting elemental composition variations.

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FRIDAY September 5

Morning Session I Chair: Krüger

9:00 – 9:30
Miya Ishihara: *In vivo spectroscopic photoacoustic imaging and system development (Invited)*
9:30 – 9:50
Hiroshi Ujii: *Novel Microscopic Tools for cellular Reponses to anti-cancer stimuli ~From Single-cell to Cell Populations~*
9:50 – 10:10
James Hutchinson: *Lanthanide nanomaterials for anti-cancer applications (Invited)*

Coffee break

Morning Session II Chair: Hofkens

10:40 – 11:00
Martin Vacha: *Nanoscale photophysics and long-range exciton transport in supramolecular and perovskite meso-hierarchical structures*
11:00 – 11:30
Thomas Basché: *Singlet fission among two single molecules (Invited)*

11:30 - Conference Closing

Free sightseeing

In vivo spectroscopic photoacoustic imaging and system development

Miya Ishihara[1,2]

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[2]*Information Center, National Defense Medical College, 3-2 Namiki, Tokorozawa, Saitama 359-8513 Japan*

A definitive cancer diagnosis is determined by histopathology. A histopathological image is obtained by staining living tissue and observing it under a microscope. There are situations in which dye is administered during a surgical or diagnostic procedure. In other words, color is used in medicine in many ways. With precision medicine, diagnostic treatments are now performed up to molecular levels.

Photoacoustic imaging is based on spectroscopy methods, and its biological applications are being actively investigated [1, 2].

We are developing a microscopic system and an in vivo system for basic research (Figure 1). Both systems have wavelength tuning capability [3, 4, 5, 6].

In my talk, I will present photoacoustic spectroscopic data to date.

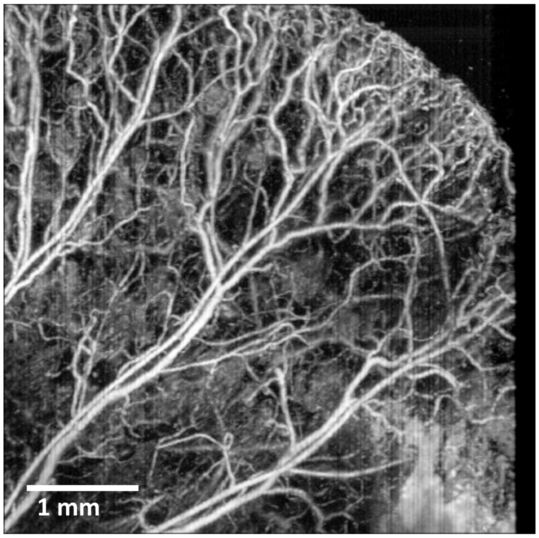

Figure 1. Vascular network of an anesthetized mouse ear imaged with our photoacoustic microscopic system. Excitation wavelength; 550-575 nm

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Novel Microscopic Tools for cellular Reponses to anti-cancer stimuli

~ From Single-cell to Cell Populations ~

Hiroshi Uji-i[1,2,3]

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With recent advances in life science, understanding the behavior of molecules and nanoparticles within cellular tissues, along with the corresponding response mechanisms of these tissues, has become increasingly important for elucidating fundamental biological phenomena. This is particularly critical In the field of drug discovery and drug delivery systems, where uncovering tissue-level mechanics, such as how effectively drug molecules reach a tumor and how its effects propagates throughout the tumor, is now recognized as an essential factor. In this context, there is a growing demand for new analytical approaches capable of evaluating cellular responses to stimuli, such as anticancer drugs or (photo)thermal treatments, across a broad scale from individual cells to multicellular tissue structures.

At the cellular level, plasmonic waveguiding endoscopic Raman spectroscopy could reveal the detailed analysis of intracellular behavior of anticancer drug molecules [1, 2, 3]. However, there remains a significant gap between conventional two-dimensional cultured cells and actual three-dimensional tissue architectures. For instance, many drug candidates that demonstrate promising results in 2D cell cultures are later rejected during animal model testing (pre-clinical evaluation). This discrepancy is largely due to our limited understanding of not only drug distribution within tumor, but also the complex collective behavior of cells, including the roles played by non-cancerous cells such as fibroblasts and immune cells.

Thus, empirical drug development strategies based on sorely on 2D cell cultures have reached their limitations. There is a strong demand for rational, mechanism-based approached grounded in tissue-level pharmacodynamics. In this presentation, we introduce a novel approach for analyzing cellular reponses to anticancer stimuli in three-dimensional cellular architectures [4].

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Lanthanide Nanomaterials for Anti-Cancer Applications

Samantha Zaman[1,2], Maria Bravo[1,2], Beatrice Fortuni[2], Hiroshi Uji-i[2,3], Susana Rocha[2], James Hutchison[1]

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Despite significant advances in cancer therapy over the past decades, cancer remains a major cause of death worldwide. In the last years there has been increasing interest in the development of new, more targeted treatment modalities with reduced side effects for difficult-to-cure cancers. Prominent examples include Photothermal Therapy (PTT), designed to selectively kill cancerous tissue in the body through light-induced thermal stress, and Photodynamic Therapy (PDT), designed to selectively kill cancerous tissue through localized, light-induced oxidative stress. The development of nanomaterials marks a significant step forward in the treatment of both PDT and PTT [1]. Nanomaterials smaller than 200 nm can accumulate in tumour tissue and exhibit strong absorption in the near-infrared range, making them effective photocytotoxic agents with cancer targeting capabilities. Nanomaterial-mediated PDT and PTT are at the stage of clinical trials but knowledge of their efficiency, mechanism and toxicity remains limited.

This presentation will outline recent works in our labs aiming to unravel the mechanisms involved in nanomaterial-mediated PDT and PTT. In particular we have employed lanthanide-doped nanomaterials, as luminescent nanothermometers when combined with gold nanoheaters for PTT, and as up-converters to drive black phosphorus quantum dots to generate singlet oxygen in PDT. To mimic the physiological features present in solid tumors most accurately, multicellular tumor spheroids were employed as well as 2D cell monolayers. More accurate models for solid tumors are crucial for the rational design of next-generation nanomaterials for targeted light-mediated cancer therapies.

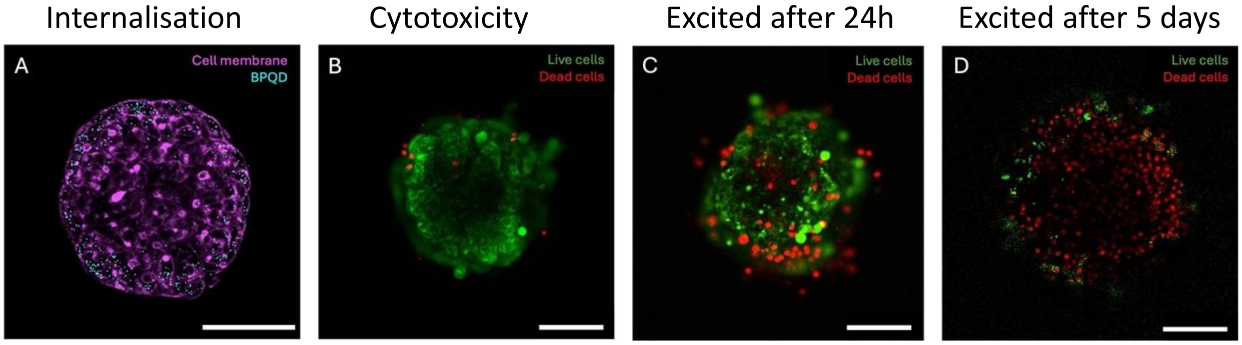
Internalisation
Cytotoxicity
Excited after 24h
Excited after 5 days

Figure 1: A spheroidal tumor mimic exposed to black phosphorus quantum dots which can generate singlet oxygen after energy transfer from photoexcited, up-converting lanthanide particles ($NaYF_4$:Yb,Er).

Reference: [1] M. Bravo *et al.*, *Journal of Controlled Release* 372 (2024) 751

Nanoscale photophysics and long-range exciton transport in supramolecular and perovskite meso-hierarchical structures

Martin Vacha

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Institute of Science Tokyo, Ookayama 2-12-1, Meguro-ku, 152-8552 Tokyo, Japan.*

Low-dimensional supramolecular structures or quantum dot superlattices represent meso-hierarchical systems that exhibit specific properties, including long-range transport of charges or excitons, potentially leading to novel optoelectronic properties and functionality. We use single molecule spectroscopy in combination with atomic force microscopy to examine new properties and functions at various scales from individual supramolecular structures to complex meso-hierarchical systems.

Position-dependent photoluminescence (PL) lifetime technique was used to study long-range exciton transport in supramolecular nanofibers. While nanofibers self-assembled from bis(phenylethynyl) anthracene linked with two dendritic hydrophilic structures show exciton diffusion coefficient on the order of 0.1 cm^2/s, the exciton diffusion in anthracene dimer nanotubes is much slower, on the order of 0.01 cm^2/s. This difference reflects differences in structural order and stability between the two mesostructures, and points to the need of well-defined and fixed chromophore arrangement for the realization of long-distance exciton transport in molecular systems.

For $CsPbBr_3$ perovskite quantum dot (QD) superlattices with sub-wavelength sizes we observe collective blinking and photon bunching phenomena at room temperature. Individual superlattices composed of thousands of QDs exhibit two-level blinking similar to that of a single QD, demonstrate photon bunching with a degree of up to 2.75, and have PL lifetimes significantly longer than individual QDs. These observations are consistent with long-range migration of singlet excitons which are trapped at a local defect in the superlattice and form biexcitons, the cascade-like emission of which gives rise to the observed photon bunching. These findings establish perovskite QD superlattices as a promising platform for room-temperature collective optical phenomena and quantum light generation.

Singlet fission among two single molecules

Thomas Basché

Department of Chemistry, Johannes Gutenberg-Universität, Mainz, Germany

Singlet fission is a photophysical process in which two triplet excitations are generated from one singlet exciton.[1] Despite the ultrafast emergence of the dark correlated triplet pair $^1(T_1T_1)$ in a terrylenediimide (TDI) dimer, a mixed optically bright $[(S_1S_0) \leftrightarrow {}^1(T_1T_1)]$ state persists lending the TDI dimer for single molecule fluorescence spectroscopy. By measuring the fluorescence intensity autocorrelation function we determine the population and depopulation rates of SF born triplet states of individual TDI dimers. We find an average overall SF rate of $1.2\cdot10^8$ s^{-1} which is almost three orders of magnitude larger than the ISC rate of the TDI monomer. The SF rate as well as the triplet decay rate are widely distributed, demonstrating the large heterogeneity of the SF process.

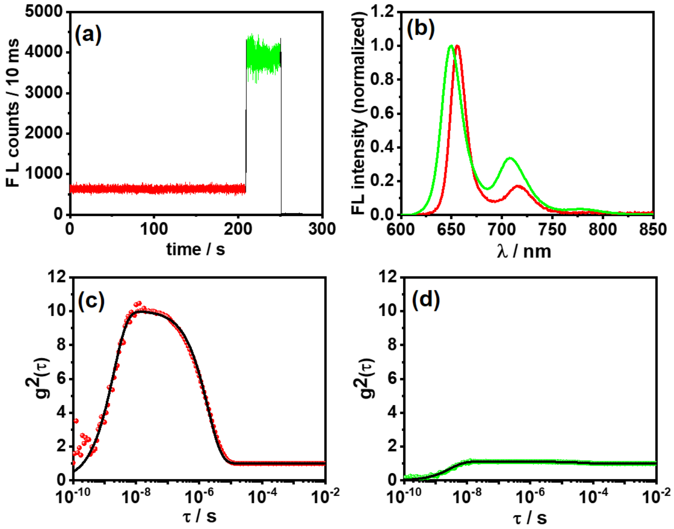

Figure 1. Fluorescence intensity time trace (a) of a single TDI dimer. After ~ 210 s a strong increase in the count rate is observed due to photobleaching of one of the monomers. The fluorescence spectra during the red and green phases are presented in (b) with a blue shift and modified intensity ratio in the green phase. The fluorescence intensity correlation functions $g^2(\tau)$ with strongly different contrasts in the red and green phases are shown in (c) and (d).

In addition to static heterogeneities, dynamic heterogeneities are observed. In particular, the SF rate and the rate of delayed fluorescence of single dimers are found to vary in a correlated manner indicating their origin from the same correlated triplet pair state. These variations are attributed to slow spin evolution which can be followed optically at the single molecule level. Our results establish single molecule spectroscopy as a new avenue into mechanistic details of the SF process which are typically drowned by ensemble averaging.

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Poster Session

P1 Wenhao Zhang: *Twisted or Planar? Packing-Dependent Photophysical Behavior in Self-Assembled Nanofibers of Dendronized Anthracene Derivatives*

P2 Jingtao Li: *Remote excitation of surface enhanced Raman in a single membrane-less organelle*

P3 Keita Abe: *Introduction of π-Conjugated Molecules into Optical Cavities for Strong Coupling*

P4 Ya Tian: *Spatial Heterogenous Diffusion Dynamics in Arginine-rich Dipeptide Coacervates*

P5 Aito Takeuchi: *In situ exploration of charge carrier and structural dynamics of perovskite photocatalysts under dynamic equilibrium condition*

P6 Yudai Yamashita: *Silver Nanowire Remote-Thermal-Sensing using Lanthanide-based Nanoparticles*

P7 Xin Xu: *Electric Field-Induced Cation Migrations in 3D/2D Perovskite* $CsPbBr_3/PEA_2PbBr_4$ *Nano-Heterostructures*

P8 Toranosuke Takagi: *Towards electrically driven single-photon sources using* $CsPbBr_3$ *perovskite quantum dots*

P9 Koki Asano: *Study of nanoscale photophysical properties of organic-inorganic hybrid perovskite films and interfaces*

P10 Reina Machida: *Plasmonic photo-thermia with Liposome-Coated Gold Nanostars: Exploring Apoptosis in 3D Tumor Spheroids*

P11 Shruti Singh: *Elucidation of Origin of Electroluminescence Intermittency in Hybrid Lead Halide Perovskite Crystals*

P12 Sasank Pattnaik: *Stochastic PL Fluctuations in All-inorganic* $CsPbBr_3$ *Microcrystals*

P13 Haruka Tsuchiya: *The investigation of substituent effect in NO releasing moiety of PeT-driven caged NO*

P14 Tsuyoshi Fukminato: *Luminescence photoswitching in perovskite quantum dots hybridized with photochromic diarylethenes*

P15 Qiwen Tan: *Probing Long-Range Energy Transfer in* $CsPbBr_3$ *Quantum Dot Superlattices with Photoluminescence and Cathodoluminescence*

P16 Jinwoong Jo: *Impact of Trivalent* Sb^{3+} *-Ion Doping on Charge Carrier Recombination Dynamics of Cesium Lead Bromide Perovskite Quantum Dots*

P17 Jaesang Yu: *Effects of Polymer Matrix and Atmospheric Conditions on Photophysical Properties of a Cesium Lead Bromide* ($CsPbBr_3$) *Perovskite Quantum Dot*

P18 Yuto Masuda: *Extension of plasmon dephasing time in asymmetric Au trimer nanogap structures*

P19 Rina Su: *Photophysical Control in Aggregation-Induced Emission–Plasmon Hybrid Systems Using Nano-Engineered Gold Arrays*

P20 Nicolas Huang: *Relationship between size and emission behavior of* $CsPbI_3$ *perovskite nanocrystals*

P21 Ami Goto: *Energy Transfer in a Quantum Dot–Perylene Bisimide Systems – Investigation of Energy Transfer from Multiple Excitons*

P22 Yoshiyuki Watanabe: *Synthesis of diarylethene derivatives having benzo[b]selenophene and turn-on type fluorescence switching in aqueous environment*

P23 Arisa Hamada: *Development of highly fluorescent and stable photochromic nanoparticles*

P24 Shin-Chwen Yeh: *Broadband Near-Field Plasmonic Enhancement in Metal–Insulator–Metal Nanostructures for Efficient Hydrogen Evolution*

P25 Shun Arai: *Single-particle transient absorption microscopy unveils hidden diversity in photosynthetic light-harvesting antenna, chlorosome*

P26 Kosuke Yoshihara: *Evaluation of nanostructures of phospholipid membranes using single-molecule tracking based on Monochromatic Excitation Fluorescence Switching (MEFS)*

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Twisted or Planar? Packing-Dependent Photophysical Behavior in Self-Assembled Nanofibers of Dendronized Anthracene Derivatives

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Supramolecular stacking governs the optical and mechanical properties of self-assembled luminescent materials. Here, we report two distinct nanofiber assemblies of dendron-functionalized 9,10-bis(phenylethynyl)anthracene, controlled solely by environmental humidity and solvent quality.[1] Under ambient conditions, a green-emitting nanofiber λ_{em} ~ 500 nm) forms through planar π-π stacking and J-aggregation. Atomic force microscopy (AFM)-assisted mechanical perturbations reveal that fiber deformation (especially transverse pressure) significantly alters the emission spectra and fluorescence lifetimes, indicative of structural flexibility and mechano-responsive luminescence.

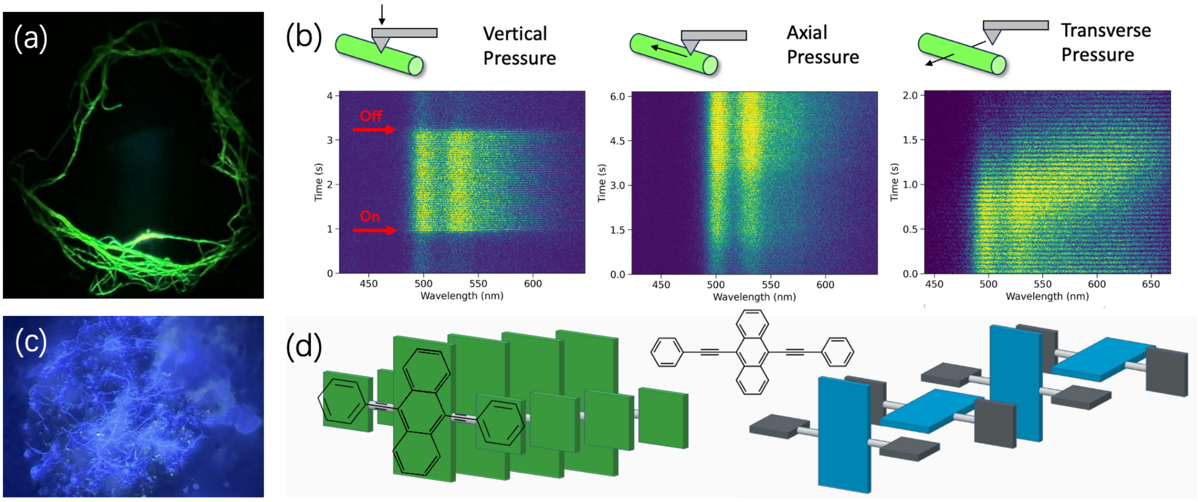

Figure 1.(a) Green-emitting nanofibers; (b) Emission changes of green nanofibers under different AFM-applied pressures; (c) Blue-emitting nanofibers; (d) Schematic models of emissive core packing in green and blue nanofibers.

In contrast, under low humidity (< 50%) with $MgSO_4$-dried solvents, we obtained a remarkably stable blue-emitting nanofiber (λ_{em} ~ 405 nm). Its emission resembles the anthracene core with slight red-shift, suggesting twisted, non-coplanar conformations. Molecular dynamics simulations (GROMACS, vacuum conditions) revealed small clusters adopting metastable orthogonal interlocks, indicating that such local motifs, although rare, can form spontaneously and exhibit notable stability throughout the simulation window (~0.9 ns). Experimentally, the blue nanofiber maintains both its morphology and emission spectra even after one-year ambient exposure. Polarization-dependent fluorescence[2] and exciton diffusion measurements further confirm this rigid, uniform packing, showing exciton diffusion reduced by a factor of 3 compared to the green nanofibers.

Our results highlight how subtle environmental control can drive distinct supramolecular architectures, yielding nanofibers with either responsive luminescence (green, flexible) or extraordinary stability (blue, interlocked). These findings offer new design strategies for supramolecular materials with tunable optical properties and enhanced structural robustness.

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Remote excitation of surface enhanced Raman in a single membrane-less organelle

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Membrane-less organelles (MLOs), such as nucleoli, stress granules, and P-bodies, are biomolecular condensates formed through liquid–liquid phase separation. Although these organelles lack membranes, they play indispensable roles in RNA processing, ribosome biogenesis, and stress responses. Emerging evidence has revealed that anticancer drugs exhibit physicochemical properties that cause them to preferentially partition into specific MLOs in cancer cells[1]. Such selective accumulation directly influences their pharmacological activity and cellular toxicity. Moreover, mutations in key scaffold proteins that drive condensate formation can disrupt drug partitioning and contribute to drug resistance. Despite these findings, the molecular basis for drug partitioning into MLOs and how this influences therapeutic outcomes remains poorly understood.

In this study, we aim to elucidate the interaction landscape between anticancer drugs and MLO components at the single-cell level using a combination of endoscopic enhanced Raman spectroscopy (SERS) and proteomic profiling. Specifically, we focus on doxorubicin (DOX), a DNA-intercalating agent widely used in chemotherapy, and its subcellular localization in A549 lung cancer cells. To probe this behavior, we designed and employed a novel gold-etched silver nanowire (AgNW) endoscopy probe that can be inserted directly into subcellular compartments of living cells[2]. Using 638 nm laser excitation, we recorded thousands of Raman spectra from the nucleoli and nuclei of DOX-treated A549 cells. Statistical analysis revealed that characteristic Raman peaks corresponding to DOX-DNA intercalation—particularly near 1245 and 1504 cm^{-1}—appeared with significantly higher frequency in nucleolar spectra. These findings underscore the importance of sub-organelle localization in understanding drug distribution patterns and hint at specific interactions between DOX and nucleolar components.

Moving forward, this research aims to provide a framework for the rational design of new drug delivery systems (DDS) that exploit the selective partitioning behavior of MLOs. By engineering drugs or delivery vehicles that specifically accumulate in disease-relevant MLOs, we hope to enhance therapeutic efficacy while minimizing off-target effects and resistance. This MLO-targeting strategy represents a novel paradigm in drug discovery, with potential applications in oncology, neurodegeneration, and beyond.

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Introduction of π-Conjugated Molecules into Optical Cavities for Strong Coupling

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When fluorescent molecules are embedded within an optical cavity, they can enter the strong coupling regime, wherein the cavity photons interact coherently with the molecular excitons. This results in the formation of hybrid light–matter quasiparticles known as polaritons. By photoexcitation, these polaritons can undergo condensation, leading to coherent light emission at a single wavelength—referred to as polariton lasing (**Fig.1**).[1] Unlike conventional semiconductor lasers, polariton lasing occurs at an ultra-low threshold due to the absence of population inversion, making it a promising alternative light source for next-generation optoelectronic devices.

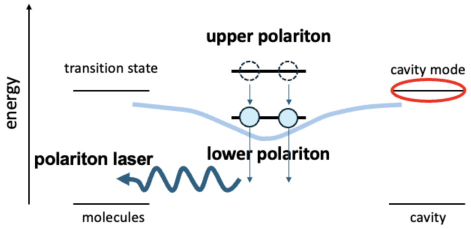

Fig 1. Energy diagram under strong coupling

In this study, we introduce π-conjugated molecules into Fabry-Perot cavity. The optical cavity was constructed using distributed Bragg reflectors (DBR) that selectively reflect light, which corresponds to the excitation and emission spectrum of the target fluorophore. A solution containing fluorophore was spin-coated onto the surface of a DBR. After drying, a second DBR was carefully coated on top to form the complete cavity, ensuring the polymer layer was sandwiched between two reflective mirrors.

To investigate the photonic behavior within the cavity, angle-resolved photoluminescence (ARPL) spectroscopy was performed. The emission spectra exhibited clear angular dispersion, showing an anti-crossing behavior consistent with the lower polariton branch, confirming that the system was operating in the strong coupling regime. In addition, a nonlinear increase in intensity was observed above a critical threshold, which is a key indicator of lasing. These findings suggest that the DBR cavity facilitated strong coupling and polariton emission.

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Spatial Heterogenous Diffusion Dynamics in Arginine-rich Dipeptide Coacervates

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Arginine-rich dipeptides, poly-PR (proline-arginine) and poly-GR (glycine-arginine), translated from aberrant hexanucleotide GGGGCC repeat expansion in the intron of mutant *C9ORF72*, are implicated in neurodegenerative diseases, such as amyotrophic lateral sclerosis (ALS) and frontotemporal dementia (FTD), by forming toxic protein aggregates in neurons.[1] These dipeptides disrupt the dynamics of various membrane-less organelles (MLO) by concentrating within and interacting with their components through liquid-liquid phase separation (LLPS). The dipeptides, with different charge properties, show different molecular diffusion dynamics within MLOs.[2] However, the mechanism regulating the diffusion dynamics of charged molecules in MLO remains unclear.

Here, we employed Raman spectroscopy and fluorescence recovery after photobleaching (FRAP) to elucidate the spatial distribution and molecular diffusion behavior of the dipeptides and poly-A RNA within LLPS-derived liquid droplets *in vitro*. Raman spectra revealed that $(GR)_{20}$ / poly-A RNA complexes have a higher density of RNAs than $(PR)_{20}$ / poly-A RNA droplets. Furthermore, we observed a concentration gradient of RNA within individual liquid droplets in Raman spectra maps, while the intensity ratios between dipeptide and RNA remained uniform, suggesting a hierarchical structure and the formation of stable RNA-dipeptide complexes.

FRAP analysis of RNA and various small dye molecules with different net-charges further indicated heterogeneous diffusion dynamics, with slower diffusion at the droplet center compared to the inner boundary. This trend aligns with the observed concentration gradient of RNA and dipeptides in the Raman spectroscopic measurements. Notably, only the negatively charged fluorescein showed a significant difference in the ratio of center-to-boundary recovery time between $(GR)_{20}$ and $(PR)_{20}$ droplets, suggesting stronger interaction with $(GR)_{20}$.

These findings highlight spatial heterogeneity and charge-dependent interactions in LLPS droplets and provide valuable insight into the mechanisms governing molecular diffusion dynamics, contributing to a deeper understanding of protein aggregation in neurodegenerative disease.

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In situ exploration of charge carrier and structural dynamics of perovskite photocatalysts under dynamic equilibrium condition

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Organic–inorganic perovskites are promising materials for next generation light-harvesting and -emitting devices such as solar cells, lasers and LEDs owing to their favorable optical features, for example, strong absorption of visible-light [1]. However, they are instable against external stimuli, especially water. Recently, it has demonstrated that the stability of perovskites is enhanced significantly when they are saturated and reach dynamic equilibrium at the solid-liquid interfaces [2]. In this condition, the perovskites can produce hydrogen under visible-light irradiation. Fundamental understandings such as charge carrier dynamics and structural stability of perovskite photocatalysts under dynamic equilibrium condition are still unclear because of nanoscopic heterogeneities at the interfaces. In this study, we employed single-particle photoluminescence (PL) spectroscopy to investigate their charge and structural dynamics [3,4].

Figure 1 shows setup for single-particle spectroscopy. Each sample was excited by 405-nm laser (CW or pulsed).

Figure 2 shows PL intensity trajectories of perovskite ($CH_3NH_3PbI_3$) nanoparticle. Remarkable PL blinking was observed only in solution, suggesting that charge trapping by chemicals in solution possibly induce nonradiative pathways.

Figure 3 shows transmission images of mixed-halide perovskites ($CH_3NH_3PbBr_{2.8}I_{0.2}$) after photoirradiation. The insets show the time after stopping irradiation. The crystal was damaged by irradiation, but under dark condition, they showed elf-healing behavior. Dynamic equilibrium-induced unique reactions are potential approach to realize new (photo)functional materials.

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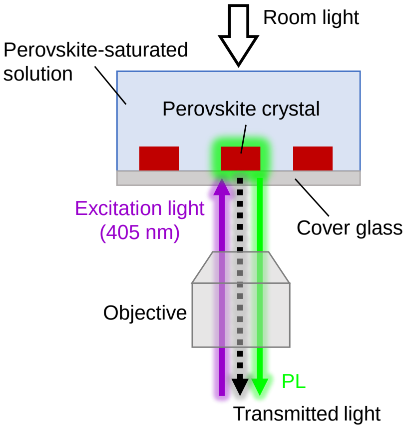

Figure 1. Experimental setup for single-particle PL spectroscopy.

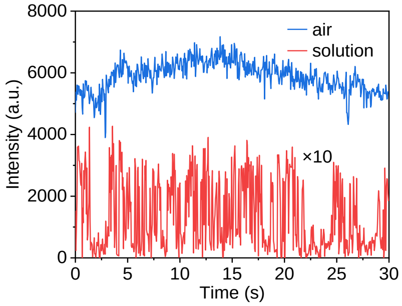

Figure 2. PL intensity trajectories of $CH_3NH_3PbI_3$ in air and in solution.

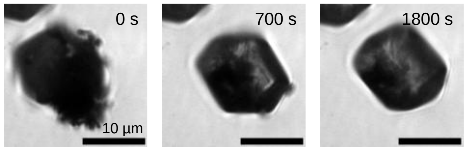

Figure 3. Self-healing reactions of mixed-halide perovskite crystal under dynamic equilibrium condition.

Silver Nanowire Remote-Thermal-Sensing using Lanthanide-based Nanoparticles

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Localized heat sensing at the nanoscale is becoming a significant issue due to the advanced semiconductor fabrications. Therefore, there is a growing need to develop a non-contact temperature sensing method at the nanoscale, which is difficult to achieve with conventional technologies.

To address this challenge, we aim to develop a novel sensing method for nanoscale temperature at nanoscale using Lanthanide-based nanoparticles (LnNPs), which exhibit temperature-dependent luminescence properties. [1]
When LnNPs are loaded on silver nanowires (AgNWs), the heat transmits along the AgNWs can be remotely measured by observing the emission spectra of LnNPs (Fig.1). In combination with AgNWs, the LnNPs can be transported to arbitrary locations.

To evaluate heat propagation in an AgNW, it is essential to heat only a localized section of the AgNW. For this, Au NPs were attached at an end of an AgNW, of which localized surface plasmon resonances (LSPRs) were excited with a laser focus. Upon the LSPRs decay, local heat was generated only at the AuNPs part. The transmitted heat was estimated by measuring emission spectra of LnNPs attached to another end of the AgNWs (Fig.2). A clear change in peak intensities ratio of the spectra (P1/P2) was observed with and without LSPRs excitation was confirmed (Fig. 3).

This result suggests that AgNWs can be used to realize the remote-thermal-sensing at the nanoscale.

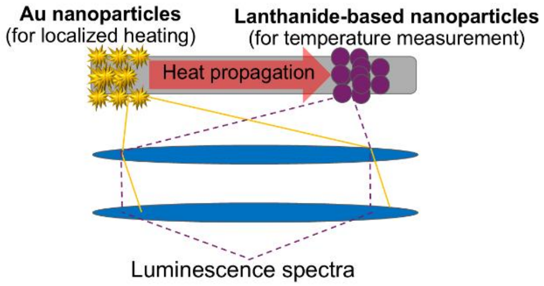

Figure 1. Heat Propagation Evaluation

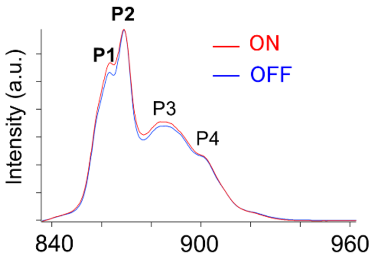

Figure 2. Spectral change by plasmon excitation

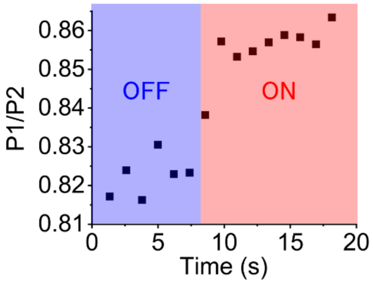

Figure 3. Intensity ratio change by plasmon excitation

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Electric Field-Induced Cation Migrations in 3D/2D Perovskite $CsPbBr_3/PEA_2PbBr_4$ Nano-Heterostructures

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Perovskite systems are of great interest due to their strong light absorption, tunable bandgaps, and enhanced environmental stability, making them promising for next-generation optoelectronic applications.[1] We have reported the structural evolution at the 3D/2D perovskite interface, where quasi-2D layers with distinct emission wavelengths can spontaneously form and dynamically rearrange at the interface shown in Fig.1(a).[2] In this study, these transformations are further shown to be directionally controllable using applied electric fields (Fig.1(b)), which induce reversible shifts in PL intensity and spectral position, clearly indicating the cation redistribution and band structure modulation.

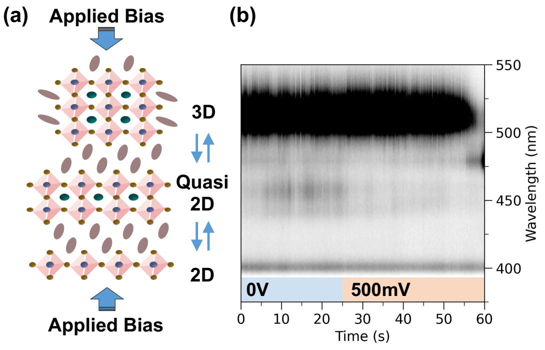

Figure 1. (a) Schematic of cation migration at 3D/2D perovskite interface and **(b)** corresponding bias-dependent real-time in-situ PL spectra.

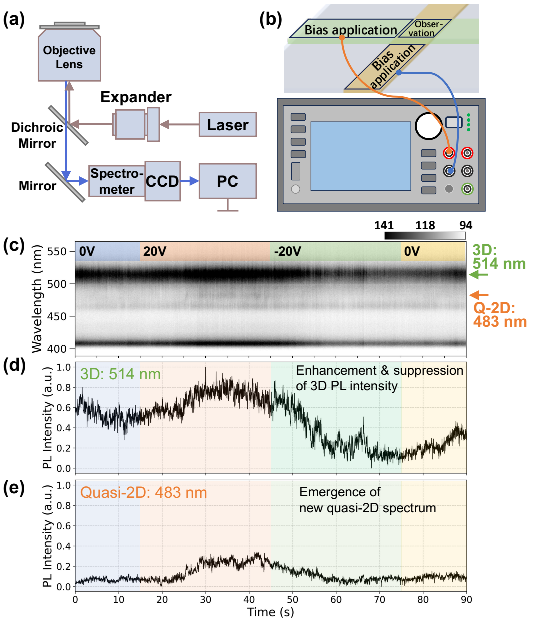

Figure 2.(a) Schematic of PL microscope and **(b)** the bias-controlled device; **(c)** in-situ PL spectra; **(d)** normalized PL intensity at 514 nm (3D phase) and **(e)** 483 nm (quasi-2D phase), highlighting field-induced enhancement, suppression, and emergence of new emission bands.

Using the integrated PL microscope for bias control shown in Fig. 2(a-b), the 3D/2D perovskite exhibits tightly time-correlated electrical and optical responses.

The concurrent in-situ PL mapping in Fig.2(c) shows a reversible structural response of the heterostructure under an electric field. Forward bias enhances the 3D PL intensity (Fig.2(d)) and introduces a new peak at 483 nm, indicating the formation of a new quasi-2D structure (Fig.2(e)). Reverse bias applied from 45 seconds suppresses the PL and eliminates the new peak, suggesting a reversible transition.

The results demonstrate that 3D/2D perovskite $CsPbBr_3/PEA_2PbBr_4$ heterostructures exhibit reversible structural changes under external stimuli. Direction-dependent PL modulation confirms reversible ion migration correlated to the field polarity. The blue shift under low forward bias indicates tunable band structure or phase composition. These findings highlight the field-tunability of heterostructure systems and the potential for reconfigurable optoelectronic applications.

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Towards electrically driven single-photon sources using $CsPbBr_3$ perovskite quantum dots

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$CsPbBr_3$ perovskite quantum dots (QDs) exhibit excellent luminescence properties with the ability to emit high-purity single photons at room temperature, making them promising candidates for single-photon sources. Most prior studies have focused on optically excited single-photon emission at the single-particle level. However, electroluminescence (EL) is essential for device applications[1]. Here, we report the first observation of single-particle EL achieved due to successful surface ligand exchange.

The $CsPbBr_3$ QDs were synthesized via a hot-injection method, followed by ligand exchange with DDAB and DLPS. Single-particle spectroscopy measurements were carried out with an epi-fluorescence microscope coupled to an EM-CCD camera.

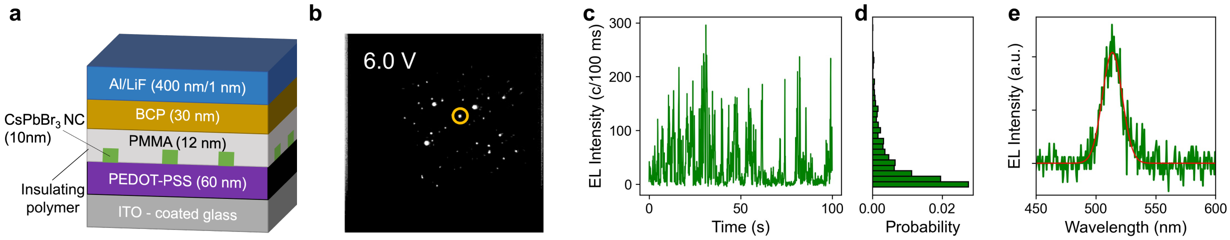

Figure 1. (a) Scheme of the multilayer EL device, showing well-dispersed $CsPbBr_3$ QDs covered with insulating polymer. (b) Electroluminescence image recorded using ligand exchanged QDs. (c) Representative single-particle EL blinking trace. (d) Emission-intensity histogram. (e) EL emission spectrum, featuring a sharp band-edge peak at ~510 nm.

To address selectively the emission of individual QDs within the EL device (Fig. 1a) and to ensure efficient charge injection, the $CsPbBr_3$ QDs were dispersed on the PEDOT:PSS layer and covered with a thin film of an insulating polymer PMMA[3]. EL images showed few spots with as-synthesized QDs. In contrast, the DDAB ligand-exchanged QDs with passivated surface defects exhibited numerous EL spots (Fig. 1b). Their blinking behavior (shown in Fig. 1c) and the emission-intensity distribution (Fig. 1d) indicate that QDs remained predominantly in low-intensity grey states, punctuated by brief and random "on" events. The EL emission spectrum (Fig. 1e) exhibits a peak at approximately 510 nm, consistent with band-edge emission of $CsPbBr_3$ QDs. Further improvement is expected from a two-step ligand-exchange strategy[4]. Partly replacing DDAB with DLPS, which binds more strongly to the QD surface, is expected to enhance surface stability and improve the single-particle EL performance.

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Study of nanoscale photophysical properties of organic-inorganic hybrid perovskite films and interfaces

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Lead halide perovskites have shown great potential for optoelectronic applications such as solar cells, light-emitting diodes and photodetectors because of their tunable bandgaps, high PLQY and capability of solution processing. Research on perovskite solar cells has progressed rapidly over the past decades, and the conversion efficiency exceeded 26% in 2024 [1]. However, the mechanisms of charge trapping that causes loss of conversion efficiency are still unclear.

Photoluminescence (PL) characterization using fluorescence microscopy has become an excellent tool to investigate local nanoscale photophysical properties of semiconductors including perovskite thin films. For example, the phenomenon of “blinking”, the fluctuations of PL intensity, was unexpectedly reported in perovskite thin films [2]. Local phenomena such as blinking provide important information on the structure heterogeneity or the effect of defects. Here, we investigate the nanoscale photophysical properties of perovskite films under various conditions.

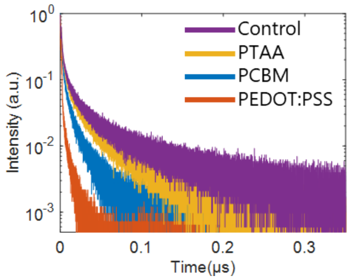

Figure 1: PL decay with transport layer

First, we investigated the effects of transport layers. For this study, we used triple cation perovskite films prepared on various transport layers by single-step spin-coating technique. We found the presence of the transport layer has resulted in a significant decrease in nanoscale PL lifetime (Fig. 1). This indicates charge loss at the interface between transport layer and perovskite layer, which was also supported by the PLQY results.

Further, we investigated the effects of electric field (EF) with the device shown in Fig. 2(a). To insulate the perovskite layer and electrode, we used polymer layers and applied voltage while irradiating the sample with light. Contrary to previous reports [3], we found PL enhancement as well as PL inhibition, depending on the direction of the bias (Fig. 2(b)). We also observed that the degree of PL change induced by the EF is spatially heterogeneous at nanoscale, and that the changes are correlated with PL lifetime. The results will be compared with other perovskite compositions and discussed in terms of ion migration.

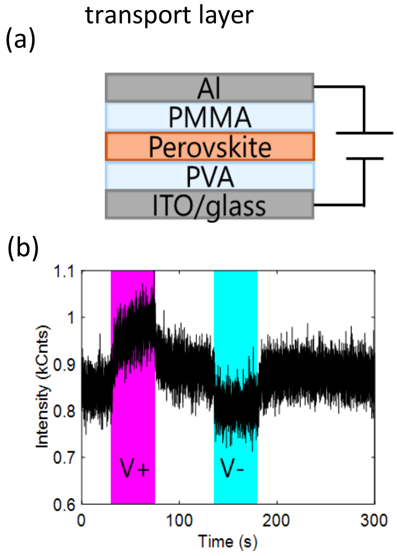

Figure 2: (a) Device structure for EF experiments. (b) Typical PL intensity time trace with EF.

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Plasmonic photo-thermia with Liposome-Coated Gold Nanostars: Exploring Apoptosis in 3D Tumor Spheroids

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Hyperthermia therapy takes advantage of the heat sensitivity of cancer cells, promoting apoptosis through localized heating. One approach for delivering heat to cancer tumor is plasmonic photothermal therapy using noble metal nanoparticles[1]. When noble metals such as gold, silver, and copper, are exposed to light at specific wavelengths, they exhibit localized surface plasmon resonances (LSPRs), exciting free electrons of conduction band to cooperative oscillations. As LSPRs decay, the released energy is converted into heat and light, with the generated heat causing localized damage to cancer cells[2]. Although this approach theoretically requires a large quantity of nanoparticles to effectively induce apoptosis in all targeted cells, many studies have reported successful treatments using significantly lower amounts, suggesting the involvement of additional, yet-to-be-understood mechanisms.

In this study, we optimized the synthesis parameters for gold nanostars, known for their high photothermal conversion efficiency and excellent material stability. Furthermore, we applied liposome coatings to these nanostars to enhance uptake-efficiency and cellular adhesion. Using three-dimensional tumor models such as spheroids, which mimic in vivo tumor microenvironment, we performed photothermal therapy and monitored heat propagation and cellular responses within individual spheroids, aiming to elucidate the mechanisms underlying the spread of apoptosis in this model system.

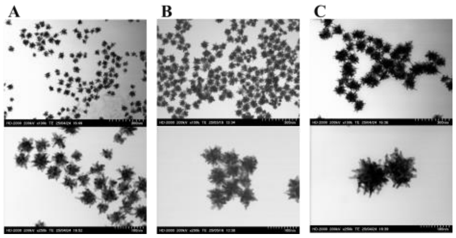

Fig 1 TEM image of gold nanostar
(The top image is 100x, the bottom one is 250x)

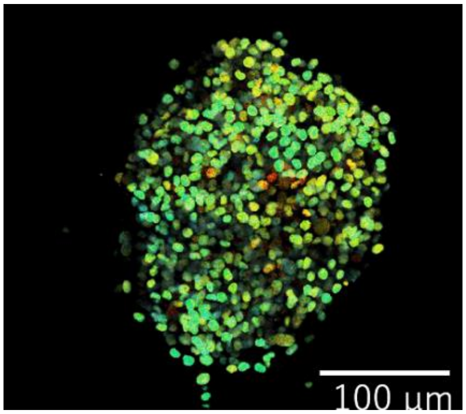

Fig 2 2-photon microscope image of 3Dspheroid upon plasmonic photo-thermia

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Elucidation of Origin of Electroluminescence Intermittency in Hybrid Lead Halide Perovskite Crystals

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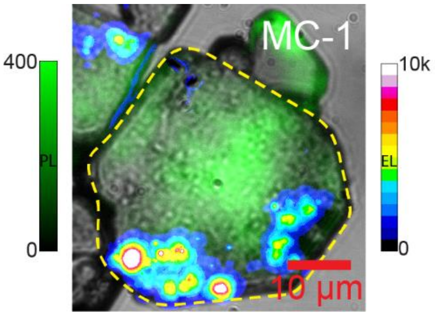
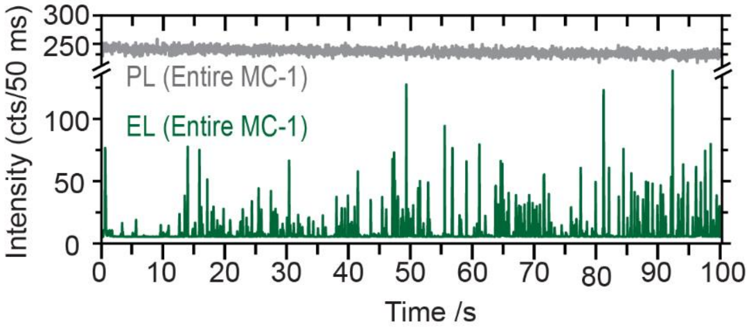
Over recent decades, metal halide perovskites (MHPs) have emerged as promising materials for advanced photovoltaic and optoelectronic devices. While the photoluminescence (PL) intermittency (blinking) of MHP nano/micro-crystals is well-documented, reports of electroluminescence (EL) intermittency remain scarce[1-3]. Here, we report intermittent EL in bulk $MAPbBr_3$ crystals, contrasting their temporally stable emission under photoexcitation. Using single-particle wide-field imaging, we delve into the dynamics of stochastic EL emission, arising from fluctuating local emission centers (ECs) within individual crystals. This behavior highlights carrier recombination dynamics during charge injection and unveils a plausible EL blinking mechanism rooted to ion migration under an applied external bias. Given its adverse impact on the efficiency of perovskite light-emitting devices, understanding EL intermittency is critical. Our findings not only illuminate this intriguing behavior but may also help to develop potential strategies to mitigate its impact, paving the way for more efficient perovskite-based technologies.





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Stochastic PL Fluctuations in All-inorganic $CsPbBr_3$ Microcrystals

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All-inorganic halide perovskites such as $CsPbBr_3$ have emerged as promising prospects for next-generation optoelectronic devices due to their good thermal stability, high photoluminescence quantum yield, and exceptional structural integrity [1,2]. Microcrystals, especially microrods, are frequently viewed as photo-physically stable systems, where emission intermittency (often observed in single emitters, NCs, QDs) is assumed to be inhibited by their lower surface defect density and high crystallinity [2,3]. Contrary to this generally held assumption, we show substantial photoluminescence (PL) flickering in quantum unconfined single CsPbBr3 microrods (length: 1-3 µm), exhibiting temporally unpredictable yet spatially correlated intensity changes across the entire crystal.

Correlation analysis demonstrates that the flickering is not spatially localized but instead displays synchronously across microrod locations, suggesting efficient charge-carrier migration and a similar quenching mechanism. These findings imply the occurrence of transitory, non-radiative defect states that intermittently suppress emission. Environmental studies support the dynamic and reversible nature of these states. Our results challenge the assumption that all-inorganic composition ensures ambient emission stability, as we unexpectedly observe flickering even in CsPbBr3 microcrystals, highlighting the presence of hidden, dynamic defect processes in seemingly ideal perovskite systems. Our study provides a deeper knowledge of carrier-trap interactions in dimensionally unconfined perovskite systems.

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The investigation of substituent effect in NO releasing moiety of PeT-driven caged NO

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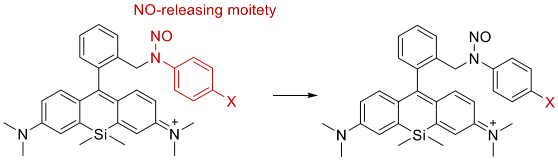
[Purpose] Nitric oxide (NO) exhibits a transient and potent vasodilatory effect in vivo and plays a crucial role in the regulation of blood flow in mammals. However, under physiological conditions, it has short half-life of only a few seconds because it rapidly reacts with biomolecules, making it difficult to handle[1]. Therefore, small molecule NO donors that can release NO are useful for applying NO in biological systems. Among them, photocontrollable NO donors (caged NOs) allow precise spatial and temporal control of NO release and are expected to serve as tools for studying NO bioactivity as well as potential therapeutic agents for cardiovascular diseases. Recently, we developed a photoinduced electron transfer (PeT)-type caged NOs that efficiently release NO in response to visible light. Through the development of PeT-type caged NOs, we found that modifying the hydroxy group at the NO-releasing site of the red-light-controllable PeT-type caged NO, NORD-1 (**1**), to a methoxy group (**2**) enhanced the NO release quantum yield (Φ_{NO}) in the presence of ascorbic acid. In **2**, NO release is proposed to be triggered by two steps of electron transfer: PeT and one-electron-reduction by ascorbic acid[2]. Based on the findings, we hypothesized that changing the methoxy group to other substituents would affect the efficiency of NO release. In this study, we synthesized compounds with various substituents at the NO-releasing site and investigated the substituent effects to identify compounds with improved NO-releasing efficiency.

[Methods] We synthesized compounds **3**–**8** with different substituents at the NO-releasing moiety and analyzed their decomposition using HPLC after 680 nm light irradiation. The decomposition products were identified by LC-MS. NO release was confirmed using an NO electrode, and Φ_{NO} were calculated. Furthermore, to identify which step was most affected by the substituents, some quantum chemical calculations were conducted and investigated the localization of radical, electron density of decomposition intermediates and energy of the π–π* and charge-transfer (CT) excited states.

[Results and Discussion] HPLC analysis revealed that **2**, bearing a methoxy group, decomposed the fastest among compounds **2**–**8**. When substituents with stronger electron-donating or electron-withdrawing properties than methoxy group were introduced, the decomposition rate decreased compared to **2**. Additionally, **2** exhibited the highest Φ_{NO}, and none of the other compounds surpassed its NO-releasing ability. LC-MS confirmed that all compounds produced the expected decomposition products upon photolysis. Moreover, quantum chemical calculations of the π–π* and CT excited states of **2**–**8** indicated that **2** showed the highest propensity for transition from the π–π* to the CT excited state. These findings suggest that PeT enhancement via electron donation from the substituent is a key factor in improving NO release efficiency.



1 (X = OH, NORD-1, $\varepsilon\Phi_{NO}$: 365)
2 (X = OMe, $\varepsilon\Phi_{NO}$: 746)

3 (X = CN)
4 (X = F)
5 (X = Cl)
6 (X = H)
7 (X = Me)
8 (X = NMe_2)

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Luminescence photoswitching in perovskite quantum dots hybridized with photochromic diarylethenes

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Perovskite quantum dots (pQDs) have emerged as a highly attractive class of materials due to their near-unity photoluminescence quantum yields (PLQY), narrow full-width at half maximum (FWHM), and ease of synthesis. The combination of these excellent optical properties with photoswitching capabilities enables their potential use in single-particle optical memories and super-resolution fluorescence imaging. Recently, we have attempted to achieve luminescence photoswitching of pQDs by hybridizing them with photochromic diarylethene (DAE) molecules [1–4]. The main challenge in this process lies in selecting an appropriate hybridization method, as pQDs are highly sensitive to environmental conditions (e.g., solvent polarity, oxygen, and light irradiation) and are prone to surface defects. In fact, our initial attempt at hybridizing DAEs and pQDs at the single-particle level resulted in poor photoswitching cycle stability [2]. A detailed investigation revealed that photoinduced electron transfer from green-emitting $CsPbBr_3$ pQDs to DAE molecules on the surface led to PL quenching. The residual holes remaining in the pQDs during this process contributed to material degradation [3]. To address this issue, we sought to suppress electron transfer from pQDs to the closed-ring isomer of DAE using two strategies. First, we increased the distance between the pQD donors and DAE acceptors to reduce the likelihood of electron transfer [4]. Second, we modified the composition of the pQDs to red-emitting $CsPbI_3$, in which the energy levels are unfavorable for electron transfer but highly favorable for Förster resonance energy transfer (FRET). Further details of our results will be presented at the conference.

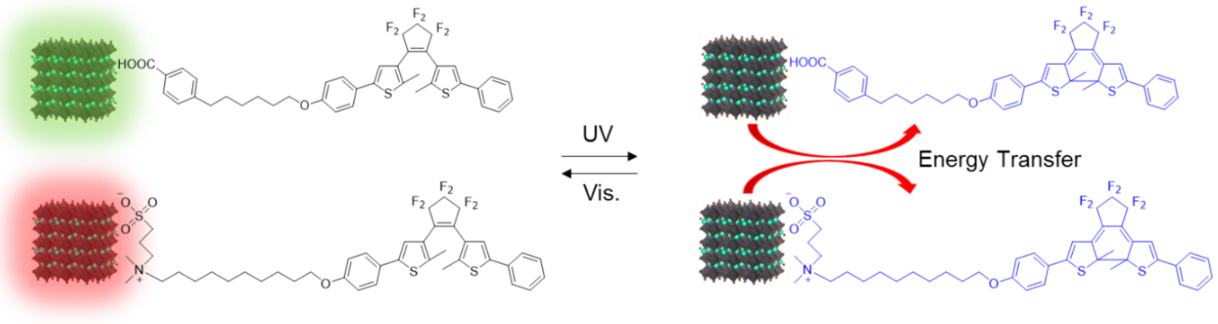

Fig. 1. Schematic illustration of the designed and synthesized DAE-pQD hybrid systems.

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Probing Long-Range Energy Transfer in $CsPbBr_3$ Quantum Dot Superlattices with Photoluminescence and Cathodoluminescence

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Perovskite quantum dot superlattices, composed of highly ordered assemblies of individual quantum dots, have emerged as promising materials for quantum photonics and optoelectronic applications due to their enhanced exciton coupling and long-range energy transfer properties[1-3]. However, direct visualization of energy transfer pathways and local defect states within these superlattices remains challenging, particularly due to the spatial resolution limits of conventional photoluminescence (PL) measurements. In this work, we employ both photoluminescence (PL) and cathodoluminescence (CL) measurements to investigate energy transfer dynamics in $CsPbBr_3$ PQD superlattices. PL measurements reveal photon bunching and collective blinking phenomena, which are attributed to long-range energy transfer within the superlattice. Complementary CL mapping (Fig. 1) provides spatially resolved evidence of heterogeneous CL intensity across different regions of the superlattice. High-resolution mapping of the CL spectra and lifetimes further confirm the presence of long-range energy transfer within the superlattices. These findings advance the understanding of energy transfer processes in perovskite quantum dot superlattices and demonstrate the value of employing complementary optical and electron-beam spectroscopies to probe collective and dynamic phenomena in complex nanoscale systems.

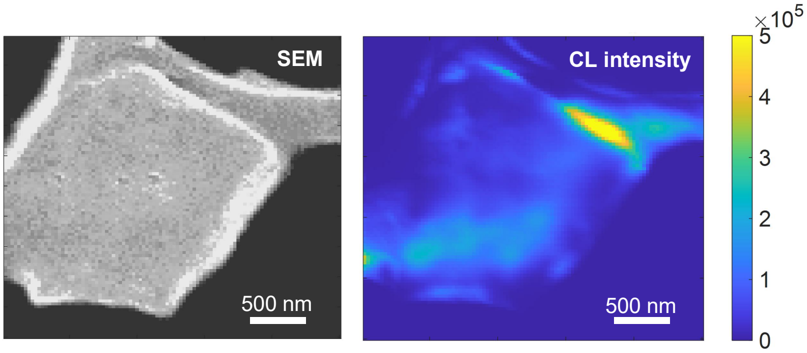

Fig 1. SEM image and CL intensity map of a $CsPbBr_3$ quantum dot superlattice

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Impact of Trivalent Sb^{3+}-Ion Doping on Charge Carrier Recombination Dynamics of Cesium Lead Bromide Perovskite Quantum Dots

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Metal-ion doping of perovskites has proven to enhance their photoluminescence (PL) properties and stability; however, the underlying charge carrier dynamics remain unclear. We synthesized a cesium lead bromide ($CsPbBr_3$) perovskite quantum dot (PQD) incorporating a heterovalent Sb^{3+} ion dopant and its pristine counterpart and performed time-resolved single-particle PL spectroscopy. The PL intensity and lifetime of the Sb-$CsPbBr_3$ PQD were remarkably enhanced compared to those of the pristine-$CsPbBr_3$ PQD because of diminished nonradiative charge carrier recombination dynamics. The charge carrier trapping (detrapping) rate was lower (higher) for the Sb-$CsPbBr_3$ PQD than for the pristine-$CsPbBr_3$ PQD, as the Sb^{3+} doping contributed to hindering the formation of the structural defects responsible for charge carrier trap states and increasing the exciton binding energy.[1] The replacement of Pb^{2+} with Sb^{3+}, which has a smaller ionic radius, in the $CsPbBr_3$ structure effectively increased the tolerance factor,[2] enabling the doped PQD to exhibit more stable local structures and, thus, suppressing its decomposition.

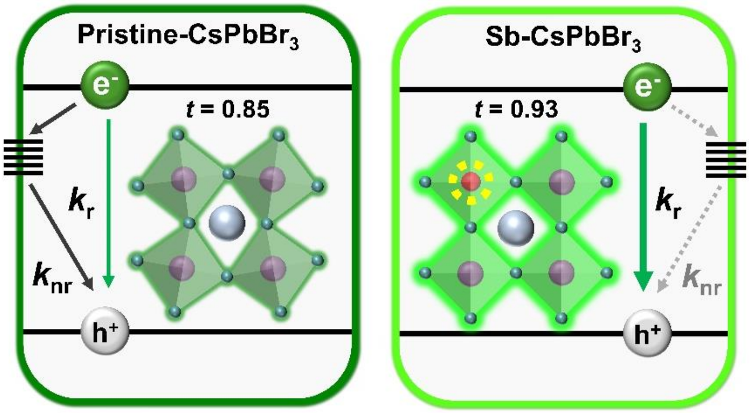

Figure 1. Schematic illustration of charge carrier recombination dynamics in single $CsPbBr_3$ PQDs, highlighting the influence of Sb^{3+}-ion doping. k_r: radiative recombination rate; k_{nr}: nonradiative recombination rate; t: tolerance factor.

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Effects of Polymer Matrix and Atmospheric Conditions on Photophysical Properties of a Cesium Lead Bromide ($CsPbBr_3$) Perovskite Quantum Dot

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Understanding the environment-dependent stability and photoluminescence (PL) properties of advanced perovskite materials remains a challenge with conflicting views. Herein, we investigated the influence of the host matrix (poly(methyl methacrylate) (PMMA) and polystyrene (PS)) and atmospheric conditions (ambient and N_2) on the PL properties of a $CsPbBr_3$ perovskite quantum dot (PQD) using single-particle spectroscopy.[1] Despite the same PL blinking mechanism, the PL properties of the PQD were considerably affected by the environmental conditions. The charge trapping and detrapping rates of the PQD were lower and higher, respectively, under ambient atmosphere than under N_2 owing to surface defect passivation by oxygen. The frequency and rate of PQD decomposition were higher in the PMMA matrix than in the PS matrix under an ambient atmosphere. PS achieved superior PQD encapsulation owing to its higher affinity toward hydrophobic surface ligands because of its aromatic rings, thereby protecting the PQD surface from moisture and thus inhibiting decomposition.

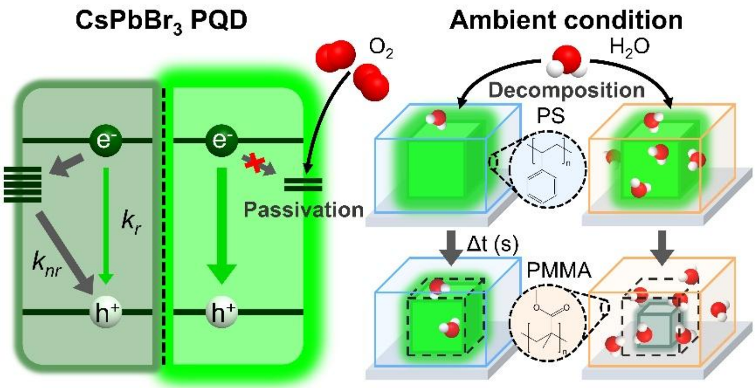

Figure 1. Schematic illustration of the environment-dependent stability and PL properties of single $CsPbBr_3$ PQDs. k_r: radiative recombination rate; k_{nr}: nonradiative recombination rate; PS: polystyrene; PMMA: poly(methyl methacrylate).

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Extension of plasmon dephasing time in asymmetric Au trimer nanogap structures

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Localized surface plasmon resonance (LSPR) enhances various optical effects, such as surface-enhanced Raman scattering. However, a significant challenge is the short dephasing time (T_2, i.e., coherence time). This is because the collective oscillations of conduction electrons in metals suffer from both elastic phase loss, arising from electron-electron scattering, and inelastic losses, due to dipole radiation. Excitation of dark plasmons reduces radiative losses from light scattering, helping to extend T_2. In this study, we aimed to extend T_2 by introducing an asymmetric structure and to elucidate the factors governing dephasing time associated with symmetry breaking.

Symmetrical and asymmetrical Au trimer structures were fabricated on glass substrates via electron beam lithography and lift-off techniques. As shown in Figure 1(a, b), each trimer consists of nanorods with lengths L_1 = 215 nm and L_2 = 105 nm, gap G = 10 nm, width W = 60 nm, height 30 nm, and pitch 400 nm. Far-field extinction spectra were measured under long-axis (L-mode) polarization, and near-field excitation spectra were obtained using a wavelength-tunable femtosecond laser. Dephasing dynamics were probed by interferometric pump-probe measurements. FDTD simulations were conducted to analyze charge distributions at the near-field peak of the E_z component.

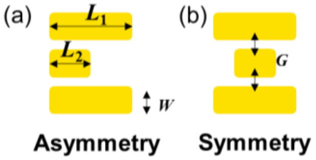

Figure 1. Schematic diagrams of asymmetric (a) and symmetric Au nanorod trimers (b). *L*, *W*, and *G* represent the length, width, height, and gap size of the nanorods, respectively.

The extinction spectrum showed two plasmon resonance peaks, and a spectral region was identified in which the near-field intensity increased slightly on the short-wavelength side between these peaks. Furthermore, the near-field peak's linewidth in the asymmetric structure was significantly narrower than that in the symmetric structure. To explain this difference in spectral linewidth, FDTD analysis was performed, revealing that in the asymmetric structure, asymmetric dipole–dipole interactions are established between the nanorods, stabilizing quadrupole modes and suppressing radiation loss. On the other hand, symmetric structures predominantly support dipolar modes, leading to enhanced radiative losses and broader spectral linewidths. Furthermore, to clarify the relationship between the linewidth of the near-field spectrum and T_2, frequency filtering was applied to the self-correlation waveform of two-photon-induced luminescence measured by the interferometric pump-probe method, and T_2 was extracted as a fitting parameter. As shown in Figure 2(a, b), T_2 was determined to be 12 fs in the asymmetric structure and 5 fs in the symmetric structure, confirming an apparent prolongation of T_2 in the asymmetric structure. This difference indicates that the suppression of dipole radiation loss in the asymmetric structure contributes significantly to the extension of the dephasing time.

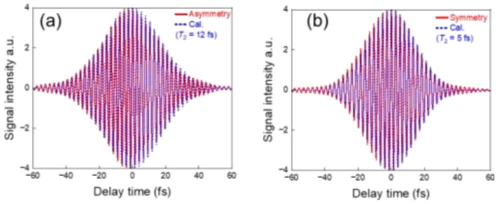

Figure 2. Frequency-filtered TPI-PL autocorrelation traces (red lines) and simulated autocorrelation traces (blue dashed lines) calculated using the dephasing time T_2 as a fitting parameter. Structural configuration: asymmetric (a) and symmetric structures (b).

Photophysical Control in Aggregation-Induced Emission–Plasmon Hybrid Systems Using Nano-Engineered Gold Arrays

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Plasmon-enhanced photoluminescence (PL) enables nanoscale control of light emission, improving quantum yields, radiative rates, and directionality. However, conventional dye-based systems face issues such as concentration quenching, poor emitter–plasmon alignment, and difficulty in quantitatively assessing photophysical parameters. Aggregation-induced emission (AIE) Illuminants overcome concentration quenching by emitting more strongly in the aggregated or solid state. Here, we introduce a methodology combining precise nanofabrication and surface engineering with the unique advantages of AIE-active molecules to build a quantitative platform for probing plasmon–emitter interactions. Using the photo-responsive AIE molecule PBYN on nano-engineered gold nanoblocks, we investigate PL enhancement and spectral behavior, establishing a basis for future studies on ultrafast dynamics under strong and weak coupling regimes.

PBYN, an AIE-active small molecule [1], was synthesized and vacuum-deposited as a 10 nm film onto gold nanoblock arrays fabricated via electron-beam lithography and lift-off. To prevent quenching from direct contact with the gold surface, a 1 nm Al_2O_3 spacer was deposited by atomic layer deposition (ALD) prior to PBYN coating. Optical absorption of a 200 nm-thick PBYN film showed a peak at 390 nm, supporting excitation near 400 nm. Accordingly, we used the 400 nm second harmonic of a femtosecond laser (800 nm fundamental, 80 MHz) to excite the system and recorded the PL spectra. By varying the nanoblock size from 60 to 100 nm in 10 nm steps, we tuned the LSPR to modulate spectral overlap with PBYN emission, enabling systematic evaluation of resonance-dependent PL enhancement.

Photoluminescence (PL) measurements demonstrated that the highest enhancement—approximately twofold—was achieved with nanoblocks having 60 nm sides. Prior extinction spectrum measurements revealed that these structures exhibit localized surface plasmon resonance (LSPR) peaks near 600 nm, which spectrally overlap with the emission peak of PBYN. This spectral alignment enabled systematic tuning of emission enhancement through the Purcell effect. As the nanoblock size increased from 60 nm to 100 nm in 10 nm increments, both the plasmon resonance and the corresponding PL emission exhibited a red-shift, reflecting modulation of emission dynamics by the shifting LSPR. These results collectively confirm that PL enhancement can be predictably modulated by engineering the plasmonic environment to match the photophysical properties of the emitter. While this red-shift is primarily attributed to Purcell-enhanced emission of PBYN, enhanced intrinsic luminescence from the gold nanostructures may also contribute. To clarify this, future studies will measure PL from the gold arrays before PBYN deposition. The use of AIE molecules such as PBYN eliminated concentration quenching, enabling accurate evaluation of photophysical changes. Moreover, spatial confinement achieved through ALD and lithography ensured high reproducibility. This platform offers a robust basis for future studies of PL dynamics, quantum yield tuning, and light–matter interactions in AIE-based plasmonic systems.

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Relationship between size and emission behavior of $CsPbI_3$ perovskite nanocrystals

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Several nm-sized perovskite nanocrystals have been known to exhibit single-photon emission due to efficient Auger recombination. Auger recombination rate is inversely proportional to PNC size, therefore, as the PNC size increases, multiphoton emission is observed. We recently reported the relationship between emission photon statistics and size of $CsPbBr_3$ PNC[1]. We found that the contribution of multiphoton emission gradually increases when the size of $CsPbBr_3$ PNC is larger than 10 nm. Recently, a relationship between size and blinking behavior was reported for $CsPbI_3$ PNCs, where the blinking behavior changes at around the size of its Bohr diameter[2]. However, further research on the size-dependent relationship of emission properties is necessary. In this work, we studied the size-dependent emission photon statistics and blinking behavior of $CsPbI_3$ PNCs by using single PNC spectroscopy technique combined with atomic force microscope. Fig. 1(a) shows the correlation between size and $g^{(2)}(0)$ obtained by photon correlation measurements. We found that PNCs under 14 nm which are in the strong quantum confinement region show high probability of single-photon emission. On the other hand, as the size increases above 14 nm, the probability of multiphoton emission increases with size. This occurs because quantum confinement effect weakens with size, thus slowing down Auger recombination rate allowing multiphoton emission to occur. In addition, Fig. 1(b) is a histogram of PNCs blinking behavior against its size. We found that PNCs under 16 nm preferred band-edge carrier blinking, whereas those larger than 16 nm preferentially show Auger blinking. This difference is thought as follows. The excitons in the smaller-sized PNC delocalizes throughout the entire PNC, thus causing the charge carriers to rapidly traps and de-traps into the surface defects. In comparison, excitons in larger-sized PNCs do not delocalize throughout the entire PNC, which slows the rate of trapping and de-trapping into the surface defects, thereby altering its blinking behavior.

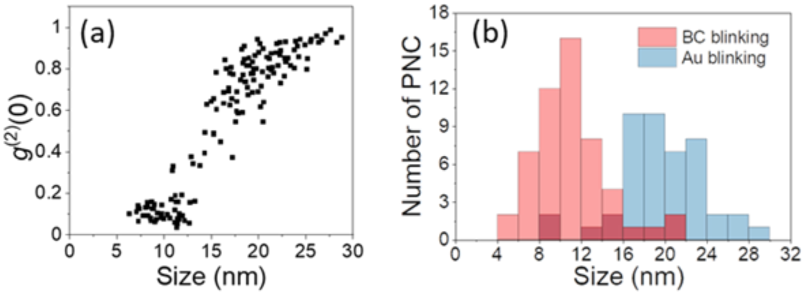

Fig. 1 (a) Correlation between size and $g^{(2)}(0)$ obtained from single PNCs. (b) Histogram of blinking mechanism and size obtained from single PNCs.

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Energy Transfer in a Quantum Dot–Perylene Bisimide Systems

–Investigation of Energy Transfer from Multiple Excitons–

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Colloidal semiconductor quantum dots (QDs) are attractive materials owing to their outstanding optical properties such as size-tunable photoluminescence (PL) wavelength, narrow PL line width, and high photo-durability. In addition to the PL properties, large absorption cross-section is also attractive property of QDs. By this property, QDs can easily absorb multiple photons and can generate multiple excitons (MX) in a single QD. Despite the generation of MXs, isolated single QDs exhibit single-photon emission because MXs rapidly decay nonradiatively to a single exciton (SX) through Auger recombination (AR). Thus, the excitons are wastefully consumed by AR. To realize the efficient utilization of generated excitons in the QD, we have investigated the energy transfer from the multiple excitons in a CdSe/ZnS to multiple perylene bisimide (PBI) molecules adsorbed on the QD. As a result, ET occurred from a single exciton to a single perylene bisimide (PBI) molecules adsorbed onto CdSe/ZnS QDs, showing that Auger recombination occurred faster than ET.[1] It is known that the Auger recombination rate is inversely proportional to the volume of a QD.

Thus, in this work, we investigated the energy transfer from a CdZnS/ZnS QD with large size to perylene bisimide derivatives adsorbed on the QD in solution and at the single QD level. By selectively exciting only the PBI, we were able to confirm that multiple PBIs were adsorbed on the QD by measuring its photon correlation. We then measured PL intensity of QD and PBI by selectively exciting only the QD. The representative time traces of PL intensities and PL decay curves detected from the single QD-PBIs are shown in Figure 1(a). The PL from PBI is predominant immediately after the start of the measurement and persists for ~140 s. The PL decay curve before 140 s (Figure 1b) exhibits a shortened decay time of 7.0 ns for the QD (τ_{QD_ET}), indicating that the PL of the QD is quenched by the ET. In Figure 1(a), the PL of the PBI suddenly disappears owing to photobleaching, and the PL of the QD appears after 140 s. The PL decay curve after 140 s (Figure 1b) exhibits a recovered decay time of 17.0 ns for the QD (τ_{QD}) is observed.

From these results, we confirmed ET at the single QD level. Furthermore, we confirmed that multiple PBIs are simultaneously emitted by measuring their photon correlation with QD excitation. Thus, we succeeded in achieving ET from multiple excitons in a QD to multiple PBIs.

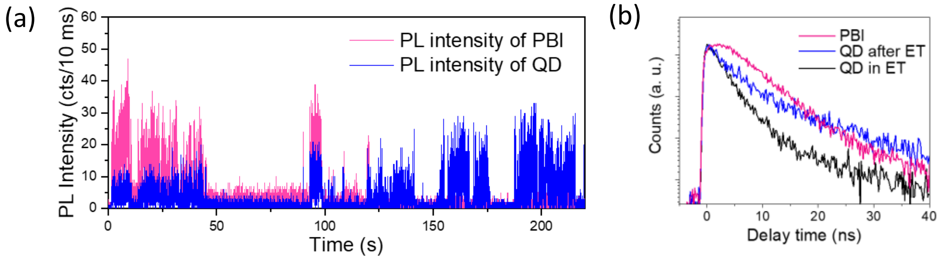

Figure 1. Time traces of PL intensity (a), PL decay curves (b) detected from a single CdZnS/ZnS QD-PBIs.

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Synthesis of diarylethene derivatives having benzo[*b*]selenophene and *turn-on* type fluorescence switching in aqueous environment

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Diarylethene derivatives (DAEs) have attracted considerable attention due to their high thermal stability and excellent fatigue resistance. Most DAEs contain sulfur (S) atoms as heteroatoms in the heteroaryl groups, but replacing sulfur with other elements like oxygen (O), nitrogen (N), phosphorus (P), silicon (Si), or sulfur dioxide (SO_2) can significantly alter their optical properties and photochromic reactivity.

In recent years, organic compounds containing selenium (Se) atoms, such as selenophene or benzoselenophene derivatives, have attracted growing interest in fields such as pharmacology, solar cells, and organic semiconductors. However, DAEs incorporating selenophene groups have been scarcely explored. Although a few studies on DAEs with selenophene units have been reported, no distinctive characteristics compared to typical thiophene-based DAEs have been highlighted.

Additionally, there is growing interest in DAEs that exhibit fluorescence photoswitching, particularly in the "*turn-on*" mode. In this mode, the open form is non-fluorescent, whereas the closed form has strong fluorescence property. Such fluorescence photoswitching behavior is crucial for applications in super-resolution fluorescence imaging. However, there are very few examples of DAEs that exhibit turn-on fluorescence photoswitching, and most of them have insufficient fluorescence quantum yields, except for those with benzo[*b*]thiophene-*S*,*S*-dioxide groups.

In this study, we designed and synthesized novel DAEs (**1H** and **2H**) featuring a benzo[*b*]selenophene moiety, and thoroughly examined their photochemical conversion behavior and fluorescence properties [1]. Notably, the closed-ring isomers both of DAEs **1H** and **2H** exhibited distinct fluorescence switching, with relatively high quantum yields ($\phi_f > 0.1$) in both solution and solid powder forms. Furthermore, leveraging their solid-state luminescence, we formulated DAE nanoparticles that successfully achieved *turn-on* fluorescence switching in aqueous environments.

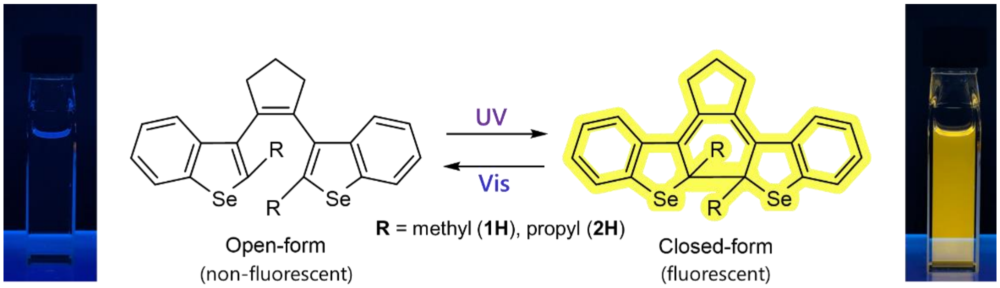

Fig. 1. Photochromism and fluorescence photoswitching of DAEs (**1H** and **2H**).

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Development of highly fluorescent and stable photochromic nanoparticles

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Fluorescent photochromic molecules, which can reversibly switch their fluorescence properties upon photoirradiation, are highly attractive for various applications, such as optical memory, displays, and molecular probes for imaging technologies. In particular, the development of fluorescence photoswitchable molecules is crucial for applications in super-resolution fluorescence imaging and anti-counterfeiting technologies.

Among various photochromic compounds, diarylethene (DAE) derivatives have attracted considerable attention due to their high fatigue resistance and excellent photoreactivity even in the solid state. Recently, we found that fluorescent photoswitchable nanoparticles (NPs) consisting of a DAE linked to a benzothiadiazole (BTD) fluorophore prepared by a reprecipitation method in aqueous solution exhibited an efficient nonlinear fluorescence photoswitching behavior, in which only a few percent of photogenerated closed-ring isomer is sufficient to completely quench the fluorescence [1].

Although the BTD fluorophore exhibits strong fluorescence both in solution and in the solid state, its photostability is insufficient for use in super-resolution fluorescence imaging applications. In contrast, perylenebisimide (PBI) derivatives are known not only for their excellent optical properties but also for their high photostability. However, under highly concentrated conditions, such as in nanoparticle (NP), their fluorescence is significantly reduced due to aggregation.

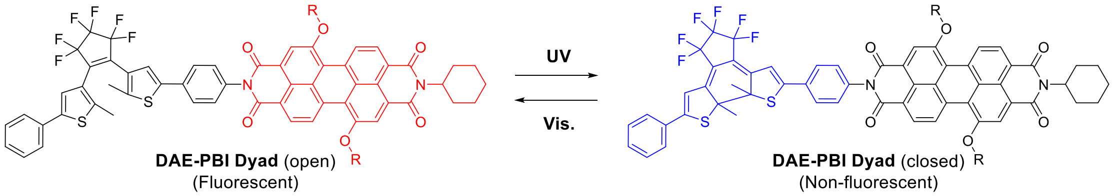

Fig. 1. Molecular structure and photochromism of DAE-PBI dyads (R is bulky substituent).

In this study, to achieve high emissive properties and enhanced stability in the nanoparticle (NP) state, we attempted to synthesize DAE derivatives linked to a perylenebisimide (PBI) fluorophore bearing bulky substituents (DAE–PBI dyads) [2,3]. NPs of the DAE–PBI dyads were prepared by a reprecipitation method, and their morphology was confirmed by scanning electron microscopy (SEM). Despite being in an aggregated state, the fabricated NPs exhibited absorption and fluorescence spectra that were nearly identical to those in THF solution, and demonstrated both high emissive properties and excellent stability in the NP state.

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Broadband Near-Field Plasmonic Enhancement in Metal–Insulator–Metal Nanostructures for Efficient Hydrogen Evolution

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The production of green hydrogen via photocatalysis has emerged as a sustainable solution to mitigate the global energy crisis. To date, extensive efforts have been devoted to semiconductor-based photocatalysis, particularly utilizing UV-active nanomaterials for hydrogen generation. However, since ultraviolet light constitutes only ~5% of the incident solar spectrum, the practical efficiency of such systems under natural sunlight is inherently limited. [1] To overcome this constraint, it is crucial to develop photocatalytic systems with broadband light absorption capabilities across the solar spectrum, enabling the generation of a greater number of hot charge carriers and thereby enhancing overall photocatalytic performance. [2]

In this work, a plasmonic Fabry–Pérot (F–P) cavity structure formed by a metal–insulator–metal (MIM) configuration is capable of achieving broadband light absorption exceeding 90% in the 400–1100 nm range. This architecture integrates top gold nanoparticles (Au) with TiO_2 nanowires (NWs) and bottom gold nanoparticle layer (Au-NP), denoted as Au@TiO_2 NW|Au-NP, was fabricated through a facile low-power visible LED photodeposition, hydrothermal growth, and sputtering process. [3] In this system, the Au NPs not only induce localized surface plasmon resonance (LSPR) to enhance light harvesting but also function as catalytic active sites, thereby improving photocatalytic performance. Moreover, two-photon-induced photoluminescence (TPI-PL) measurements demonstrate that the Au@TiO_2 NW|Au-NP hybrid structure exhibits pronounced broadband near-field enhancement, indicative of efficient hot carrier generation. Ultrafast time-resolved spectroscopy further confirms a shortened plasmon dephasing time, which is attributed to the enhanced local electromagnetic fields and hot carrier dynamics. Electrochemical impedance spectroscopy (EIS) and photocurrent response analyses indicate superior charge transport and excellent electron–hole separation properties of the Au@TiO_2 NW|Au-NP photoelectrode. Overall, this study demonstrates a promising strategy for designing broadband-absorbing plasmonic nanostructures with enhanced near-field effects, offering substantial potential for efficient and sustainable hydrogen production.

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Single-particle transient absorption microscopy unveils hidden diversity in photosynthetic light-harvesting antenna, chlorosome

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Photosynthetic organisms convert solar energy into chemical energy, a process fundamental for life on Earth. The initial step of photosynthesis is the absorption of light energy by light-harvesting antennas. Chlorosomes, the light-harvesting antennas of green sulfur bacteria, exhibit the highest efficiency in light capture among all known antennas. They absorb light energy and transfer it to the reaction center (RC) via baseplate and Fenna-Matthews-Olson (FMO) proteins (Fig.1(a)). Each chlorosome contains 50,000-250,000 bacteriochlorophylls (BChls) that self-assemble without a protein scaffold to form rod- and lamella-like structures, indicating significant structural heterogeneity (Fig.1(b)) [1]. This heterogeneity likely disrupts the excited energy transfer (EET) process, which is considered unfavorable for the growth of phototrophs. However, the relationship between the EET process and structural heterogeneity is not yet fully understood. Although single-particle fluorescence spectroscopy is useful for analyzing functional variety due to structural heterogeneity, it is limited in measuring EET in chlorosomes because of the low fluorescence emitted during EET and the insufficient time-resolution in fluorescence spectroscopy (typically, on the 10^{-12} s timescale) to monitor the EET within chlorosomes. To overcome these challenges, we developed the single-particle transient absorption microscope that allows us to probe EET dynamics at the single-chlorosome level. Our measurements revealed significant variation in the EET rate among individual chlorosomes, indicating distinct rate groups. Mutant chlorosomes with varying abundance ratios of rod- and lamella-like BChl assemblies enabled assignments of the rate group to each assembly. Moreover, we characterized the EET rate, absorbance, and energy transfer efficiency of the rod- and lamella-like assemblies, offering an opportunity to elucidate the roles of the assemblies and the light-harvesting mechanism in chlorosomes.

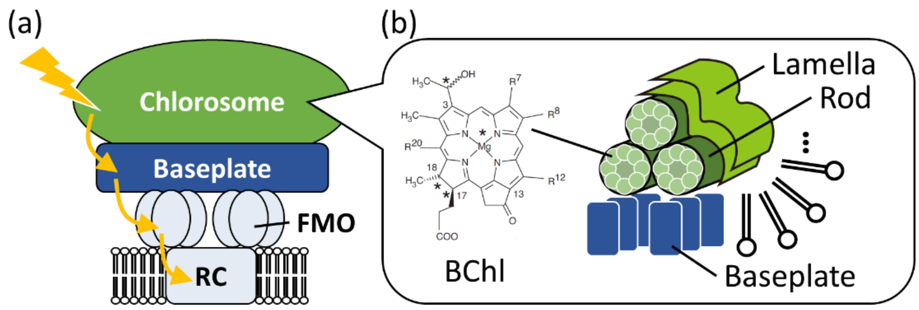

Fig.1 (a) Photosynthetic system of green sulfur bacteria. The absorbed energy by chlorosomes transfers to the RC via baseplates and FMOs. **(b)** Schematic of the chlorosome. It contains BChl assemblies forming the rod- and lamella-like structures.

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Evaluation of nanostructures of phospholipid membranes using single-molecule tracking based on Monochromatic Excitation Fluorescence Switching (MEFS)

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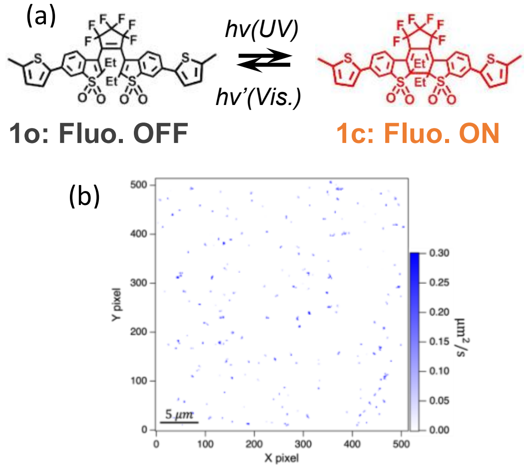
Cell membranes, primarily composed of phospholipids, play a vital role in the transport of small molecules into and out of cells. Understanding the physicochemical properties of these membranes is essential not only for elucidating cellular functions but also for designing effective drug delivery systems. In this study, we investigate the physicochemical characteristics of lipid membranes at the single-molecule level.

To achieve this, we employed single-molecule tracking (SMT) using monochromatic excitation fluorescence switching (MEFS) of fluorescent diarylethene derivatives (fDAE). The chemical structure of the fDAE used is shown in Figure (a). Our recently developed MEFS-SMT technique [1] enables the tracking of thousands of single molecules, with trajectories covering the entire imaging field of a fluorescence microscope[2]. Phospholipid membranes, such as those composed of dioleoyl phosphatidylcholine (DOPC), were prepared on glass substrates in aqueous solution. The fluorescence switching behavior and translational motion of individual fDAEs were analyzed under continuous 532 nm laser irradiation. In DOPC membranes, fDAEs exhibited spontaneous fluorescence ON-OFF switching with an average ON time of 0.3 s, allowing effective single-molecule tracking.Detailed analysis of the diffusion behavior yielded spatial maps of probability density distributions and diffusion coefficients of the fDAEs. Figure (b) shows a two-dimensional diffusion coefficient map for fDAEs in a DOPC membrane. In our presentation, we will share these findings for both single-component and multi-component phospholipid membranes and discuss how local membrane properties influence the motion of individual molecules.



(a) Chemical structures of the isomers of fDAE. (b) 2D map of the diffusion coefficient of the fDAE in a membrane of DOPC

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Hybrid 1D Silver Organochalcogenides: From Bandgap Modulation to Exciton Diffusion

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Metal organochalcogenides (MOCs) are a versatile class of organic-inorganic hybrid semiconductors known for their unique light-matter interactions.[1,2] Their hybrid nature allows for extensive structural and optoelectronic tunability through ligand engineering.[3] In this study, we systematically modulated the electronic properties of ligands by introducing Cl and Me functional groups, achieving precise control over the optoelectronic characteristics of one-dimensional (1D) Ag-based MOCs.

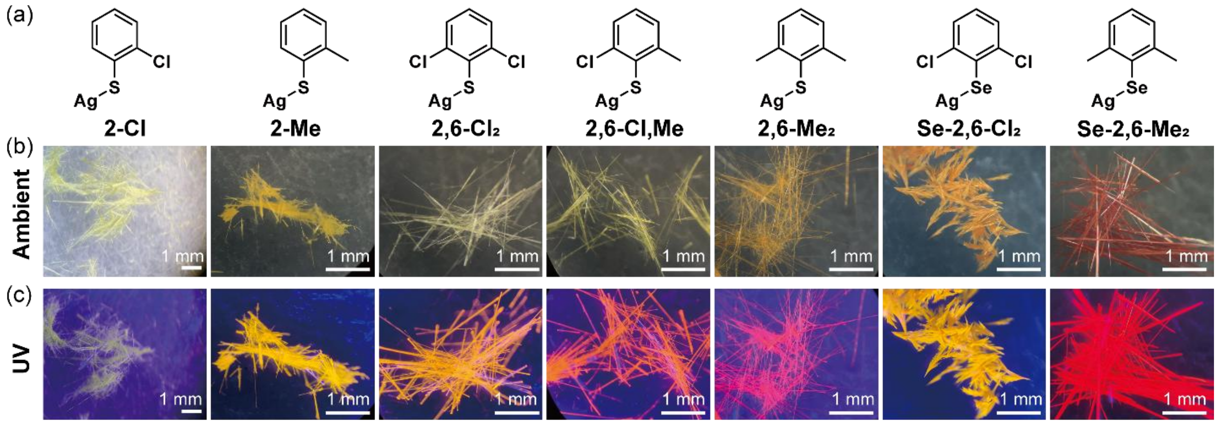

Figure. Synthesized MOCs. (a) Molecular structure of MOCs. Optical micrograph of the MOCs at (b) ambient condition and (c) under UV light.

We successfully synthesized seven Ag-based MOCs by systematically varying the number of functional groups and chalcogen elements (**Figure**). These MOCs exhibited bright luminescence peaks ranging from 560 to 690 nm, covering the yellow-to-red region. Substituting Cl with Me groups in benzene ligands induced a red-shift in both absorption and photoluminescence for S- and Se-based MOCs. Further, we performed detailed characterization of the optical properties of these MOCs on the level of single nanocrystals, including emission anisotropy, carrier diffusivity, and precise structural characteristics.

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Single-molecule spectroscopy of conjugated donor-acceptor copolymers containing non-fullerene acceptors

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In future organic solar cell or organic photodetector materials, non-fullerene acceptors (NFAs) have been attracting attention because of their versatility, tunable properties and broad absorption spectra. One such example, ITIC, has been showing promising properties in a copolymer with a donor molecule PM6. However, thin films prepared from PM6-pITIC show complex photophysical properties, including multipeak photoluminescence (PL) spectra that are still eluding interpretation. We use single-molecule spectroscopy and in-situ solvent-vapor annealing (SVA) method to prepare mesoscale aggregated structures of PM6-pITIC single chains and study the effect of intra- and interchain interactions on the photophysical properties. Single PM6-pITIC chains dispersed in PMMA matrix show emission from both the PM6 donor and the pITIC acceptor, pointing to constrained efficiency of the D-A energy transfer. In aggregates prepared by SVA from good solvent the donor emission completely disappears, presumably due to closer distances between the donor and acceptor blocks and increased energy transfer efficiency. SVA using a mixture of good and poor solvents leads, on the other hand, to compact aggregates showing PL emission shifted further to lower energies. This result is interpreted as the formation of charge-transfer states which are enabled by strong interchain interactions in the aggregate. These results were corroborated by the PL study of single chains and aggregates of the respective homopolymers of PM6 and ITIC, and by PL lifetime measurements.

Computational Modeling and Thermodynamic Simulations of Nanomaterials for Cancer Photo-Thermal Therapy

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[Introduction] Cancer remains a major global health challenge, prompting the need for more targeted and less invasive treatment strategies. Photothermal therapy (PTT) is a promising approach, utilizing gold nanoparticles (AuNPs) to convert near-infrared (NIR) light into localized heat, effectively inducing cancer cell death while minimizing damage to surrounding healthy tissues. However, several obstacles hinder the clinical translation of PTT. A key limitation is the lack of precise and predictive models for heat diffusion and dissipation within biological tissues, which can lead to under- or over-heating of the targeted area. Additionally, the non-uniform distribution of AuNPs within tumors can result in uneven heating, reducing therapeutic efficacy and increasing the risk of incomplete tumor ablation. This study addresses these challenges by focusing on computational modeling and simulation of heat transport during PTT, aiming to enhance thermal control and nanoparticle delivery uniformity for more effective and safe cancer treatment outcomes.

[Method] Current main focus of this research is computational modeling of hybrid AuNP–AgNW nanostructures used for improved hyperthermia and nanoscale thermometry. Use of silver nanowire (AgNW) probes facilitate precise heat localization, greatly improving hyperthermia control. In the model's 3D configuration, AuNP with a length of 175 nm and diameter of 50 nm is combined with AgNW with lengths of 5000 nm and diameters ranging 120~200 nm. Optical absorption by AuNP under near-infrared (NIR) irradiation was simulated using ANSYS Lumerical FDTD, and the resulting temperature fields were computed using COMSOL Multiphysics for thermal analysis.

[Results] Figure 1 shows the simulated temperature fields generated by AuNP–AgNW nanostructures under NIR irradiation, with varying AgNW diameters: (a) 120 nm, (b) 160 nm, and (c) 200 nm. Increasing the AgNW diameter slightly extends the heat distribution along the AgNW. Isothermal contours (40~24 °C) show broader heat diffusion as diameter increases, while the temperature gradient remains steep near the AuNP, confirming improved thermal spread through AgNW. This supports that optimizing nanomaterials parameters is important for controlled thermal delivery in PTT applications.

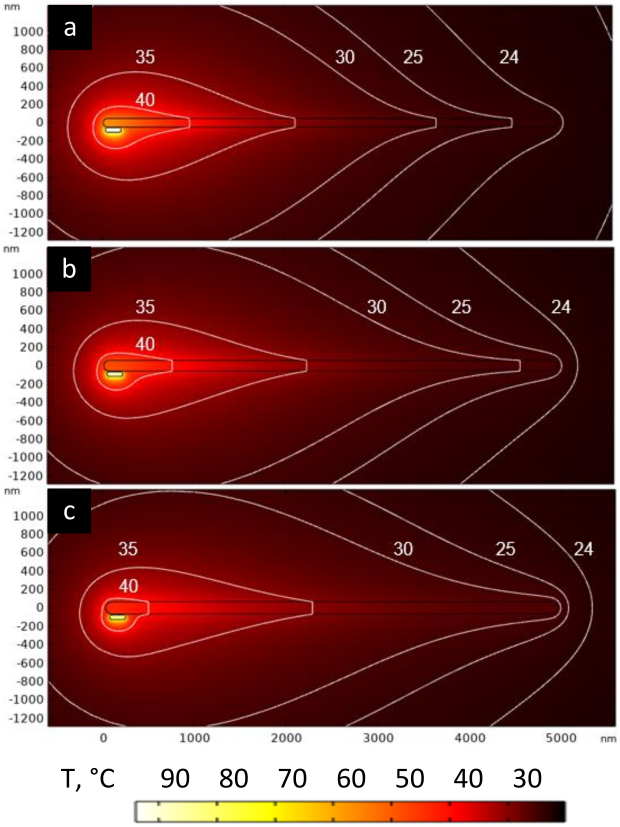

Fig.1 Simulated temperature fields and isotherms in AuNP-AgNW structures with AgNW diameters: (a) 120 nm; (b) 160 nm; (c) 200 nm;