SCOM5: Strong Coupling with Organic Molecules Conference 2025



16–19 June 2025, Odense, Denmark









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Welcome

It is with great pleasure that we welcome you to SCOM5, the 5th edition of the conference on Strong Coupling with Organic Molecules. The conference will take place from the 16th to the 19th of June, on the main campus of the University of Southern Denmark in Odense. To celebrate the 5th anniversary, we extended the usual three-day duration to four days, with the first day devoted, for the first time, to an advanced PhD school, anticipating to offer young researchers and newcomers a forum for detailed presentation and discussion of the fundamentals of polariton science.

As far as the conference programme is concerned, we tried to maintain a good balance between the experienced and the emerging leaders of the field. Topic-wise, and following the discussion at the end of the previous edition in San Diego, we attempted to expand towards more traditional quantum optics, while alternative to organic molecules strongcoupling templates such as excitons in transition-metal dichalcogenides and perovskites were also included. Finally, to mark the 5th anniversary, we decided to give the floor, for the first time as invited speakers, to the four founding fathers of SCOM: Javier Aizpurua, Bill Barnes, Francisco García-Vidal and Jaime Gómez Rivas.

Combining this exciting programme with social activities such as a boat trip along Odense Å and dinner at a famous traditional restaurant, we hope that all participants will enjoy their time in Denmark, get the opportunity to reconnect with good friends and establish new collaborations, that will drive the further growth of the field of polaritonics!

Christos Tserkezis Conference chair N. Asger Mortensen Conference co-chair

Niels Asar Mortensen



SDU JNIVERSITY OF



Sponsors



Otto Mønsteds Fond

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Organizers

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Practical Information

How to get here

Both the PhD School (16 June) and the Conference (17–19 June) take place at the University of Southern Denmark, in Odense, Denmark. Odense is situated on the island Funen (called *Fyn* in Danish and *Fionia* in Latin) in the middle of Denmark. The nearest airport is Copenhagen Airport. Direct trains leave from Copenhagen Airport to Odense hourly and the duration of the trip is approximately 1 hour 45 min. More information about public transport in Denmark can be found on Rejseplanen. Tickets for the train can be bought online on DSB or at all stations.









Moving around in Odense

To move around in Odense, especially between city centre and SDU campus (where both the PhD School and the Conference take place), the preferred option is the tram (Letbane). A map of the tram network in Odense is shown below, with the most relevant stops highlighted. Tickets can be bought via QR codes at the tram stops with credit card, or online. Rejseplanen is very useful to find your way around in Odense.









SDU campus

The main SDU Odense campus is a continuous long building built along the tram line, with several vertical branches. Each of the corridors has a street name, with the main corridor (starting at tram stop Campus Odense and extending all the way to tram stop SDU/University) called Gydehutten. The names are written on the floor and above the doors at crossroads. You can also use SDU Maps to find directions.

PhD school venue

The venue for the PhD School (16th of June) is room U47 at the main SDU Odense campus, level 1 at the main corridor (corner between Gydehutten and Knoldene).

- <u>How to get to U47 at SDU by public transport</u>: Take the tram/Odense Letbane from Odense train station with direction towards Hjallese Station. Get off the tram at the stop 'Campus Odense' (see map below). Enter the building by the main entrance, and walk up the stairs. At the end of the stairs walk straight ahead along Gydehutten for a few hundreds of meters until you meet U47 at your left hand.
- <u>How to get to U47 by car</u>: Follow Google Maps for the address: Moseskovvej 67, 5230
 Odense M. Parking outside the building is possible free of charge and without registration, except for specific marked spots reserved for SDU vehicles. Enter the building at the entrance marked on the map below (Knoldene Ø), go to the first floor, and walk along Knoldene until you reach Gydehutten.

Conference venue

The venue of the conference (17-19 June) is the main auditorium (room O100) at the main entrance of the SDU Odense campus.

- <u>How to get to O100 by public transport</u>: Take the tram/Odense Letbane from Odense train station with direction towards Hjallese Station. Get off the tram at the first stop in the university campus, Campus Odense (see map above). At the tram stop, you will immediately see the main entrance on the left. Walk up the stairs and you will meet the registration desk right in front of you.
- How to get to the main auditorium (O100) by car: Follow Google Maps for the address: Moseskovvej 47, 5230 Odense M. Parking outside the building (P1) is possible free of charge





and without registration, except for specific marked spots reserved for SDU vehicles. Enter the building at the entrance marked on the map below (Agrene \emptyset), move one floor up, and walk until you meet Gydehutten; then turn right.

SDU campus map

A map of the SDU Odense campus can be found below, with all relevant points for the PhD School and the Conference marked. The venues for the PhD School (U47) and the Conference (O100) are marked in red, and the relevant tram station is marked in blue. When buses are used for tranferring participations to social activities, the meeting point will be Parking P1 (in green). Additionally, you can also use SDU Maps to find directions. The photo to the right shows the square in front of the main entrance.









What to do in Odense

We are looking forward to welcoming you to the city of Odense. First, you should be aware that the Danish weather is always a bit unpredictable. So bring clothes to be prepared for rain showers, a bit of cold wind and, if you are lucky, nice sunshine. Below you can find some suggestions for activities in Odense. For more options, visit the official tourist website of Odense. Welcome to Odense!

The Funen Village

The Funen Village is an open-air historic museum with buildings from Funen and the islands showing how a local village could look in the 1800s. Peasant wives, farm workers of both sexes, blacksmiths and many others are here—in contemporary costumes—to show you how people lived and worked on the land in the 1800s. The conference dinner on 18 June will take place at Sortebro Kro in the Funen Village.









A walk in the footprints of Hans Christian Andersen

Go for a walk in the old part of Odense and follow the footprints of Hans Christian Andersen. By following the suggested route, you will pass by Skt. Hans Kirke (where Hans Christian Andersen was baptized), H.C. Andersens Hus (the birth place of Hans Christian Andersen), Fattigskolen (the school for poor kids that Hans Christian Andersen attended), Hans Christian Andersens Barndomshjem (where Hans Christian Andersen lived during his childhood) and ending the walk at the Odense Cathedral and the town hall of Odense. The walk is approximately 1.6 km long, and will take you through some of the nicest parts of Odense.



Link to map of the walk







Odense Harbor Bath

What about going for a swim in Odense Harbor Bath in the morning before the conference starts? Odense Harbor Bath resembles the top deck of a ship, with a pool in the center plus sauna and changing facilities are available. The Harbor Bath is open in the morning 6.00-7.30 and later from 11.00-18.30. Access is free of charge. The walking distance from Odense Station to the Harbor Bath is 1 km.

Address: Gamle Havnekaj 3, 5000 Odense C

In 2023, Vogue wrote an article about Danish Harbor Baths, which is a year-round activity in Denmark.









Food and beers at Storms Pakhus!

Grab some food and a beer at Storms Pakhus, which is a very popular street food market in Odense with both indoor and outdoor seating. It is open every day 11.00-23.00. This is a great place to bring your old and new friends from the conference! The walking distance from Odense Station to Storms Pakhus is 500 m. This is also going to be the venue for a relaxed and informal dinner in the evening of the PhD-school day.

Address: Lerchesgade 4, 5000 Odense C









Art Museum Brandts



If you are interested in a cultural experience, we recommend visiting the Brandts Art Museum, where 250 years of art history are gathered under one roof. Brandt shows classic, modern, and contemporary art. In June 2025, Brandts presents, among others, an exhibition with Kinga Bartis' unique works. Brandts is located in the heart of Brandts Klædefabrik, which was once a clothing factory, and the distinctive old-fashioned industrial architecture with its yellow bricks and green mullioned windows makes it stand out from its surroundings and feels like a small town within the city.









Social activities

PhD School dinner at Storms Pakhus

16 June, 18.00—Storms Pakhus

Discover how the hundred-year-old warehouse has been turned into the largest street food mecca in Odense, with an abundance of food stalls and exciting bars. Every PhD summer school participant will receive a voucher worth 200 DKK for purchasing their own food and drinks at Storms Pakhus. We will eat together in a designated SCOM5 area.

The costs for bus transportation and the food & drink voucher are included in the registration for the PhD summer school.

How to get to Storms Pakhus: At 18:00, we will leave together by bus (P1) for Storms Pakhus. The address is: Lerchesgade 4, 5000 Odense C



Opening reception

17 June, 18.00—Gydehutten

We celebrate the opening of SCOM5 with a reception. Enjoy a glass of wine, tapas, catch up with old and new friends, plus take the opportunity to visit the SCOM5 poster session. More than 50 researchers present their research at the poster session. The reception takes place at the conference area at the main entrance of SDU, in front of room O100.







Boat trip and Conference dinner

18 June, 17.00—Munke Mose park

At 17.00, the conference bus will depart from SDU (P1) and take everybody to the starting point of the boat trip on Odense Å (the small river), at Munke Mose park. The old boat service has been sailing since 1882. The boat will take us on a scenic sail along the gardens of Odense's fine old neighborhoods, past Odense Zoo, and out to the beech forest Fruens Bøge.



18 June, 19.00—Sortebro Kro

The conference dinner takes place at Restaurant Sortebro Kro in the Funen Village. Sortebro Kro is a Royal Licence country inn dating from 1805, which was moved from its original location and rebuilt in the working museum town of the Funen Village, to represent all the country inns that were established in Denmark's past. Please note that a visit to the Funen Village is not possible during the conference dinner.









If you have signed up for the conference dinner, this will be indicated on your conference name badge.

- <u>How to get to Sortebro Kro:</u> From the boat stop it is 1.2 km walk through the beech forest to Sortebro Kro. Bring clothes and shoes fit for the walk.
- How to get to Sortebro Kro by car:

If you choose not to go on the boat trip, it is also possible to go to the restaurant by taxi or your own car. The address is: Sortebro Kro, Sejerskovvej 20, 5260 Odense. A large car park is available free of charge in front of the building.







Programme

Programme at a glance

	Monday, June 16	Tuesday, June 17	Wednesday, June 18	Thursday, June 19
08.00	Registration, breakfast	Registration, breakfast	Breakfast	Breakfast
08.30		Welcome		
08.40	Stéphane	Thomas Ebbesen	Päivi Törmä	Vinod Menon
08.50	Kéna-Cohen			
09.00				
09.10				Darius Urbonas
09.20				
09.30		Mini break	Mini break	Akshaya G
09.40		Bill Barnes	Javier Aizpurua	
09.50				Daniele Sanvitto
10.00	Mini break			
10.10	Antonio I.	May Godsi	Rakesh Arul	
10.20	Fernandez-			$Coffee \ break$
10.30	Dominguez	Dominik Sidler	Carla Estévez Varela	
10.40				
10.50		Coffee break	$Coffee \ break$	Marzena Szymanska
11.00				
11.10				
11.20		Christoph Lienau	Klaus Mølmer	Pietro Tassan
11.30	Coffee break			
11.40				David Hagenmüller
11.50		Xabier Arrieta	Ben Yuen	
12.00	Nicolas Ubrig			Daniel J. Tibben
12.10		Thomas Schnappinger	Yassir El Moutaoukal	
12.20				Girish Lakhwani
12.30		Lunch break	Lunch break	
12.40				Lunch break
12.50				
13.00				

continued on next page







	Monday, June 16	Tuesday, June 17	Wednesday, June 18	Thursday, June 19
13.00				
13.10				
13.20	Lunch break			
13.30				
13.40				
13.50				
14.00		Joel Yuen-Zhou	Libai Huang	Jaime Gómez Rivas
14.10				
14.20				
14.30		Jussi Toppari	Luca Sortino	Marco Marangi
14.40				
14.50	Michael	Anael Ben-Asher	Ishita Jena	Philip Thomas
15.00	Ruggenthaler			
15.10		Frank Huo	Coffee break	Mikhail Vasilevskiy
15.20		Cleffer harri		Ceffee here h
15.30		Сођее отеак	NI:1: M£:	Coffee oreak
15.40 15.50			INICOlo Maccalerri	
16.00		Timur Shogai		
16.00	Coffee break		Aleesha George	Francisco J. Carcía Vidal
16.10	Coffee orean		Micesna George	Garcia viuai
16.20		Frederik Schröder	Fritz Joshua Schnur	Kayya S. Mony
16.40	Karol Kołataj		11112 Jobinati Sommar	itavya 5. Mony
16.50		Jonas M. Peterson	Departure to	Dmitry Dovzhenko
17.00			boat trip	
17.10		Michael Papachatazakis	*	Mini break
17.20				Awards & discussion
17.30		Sara Núñez Sánchez		
17.40				
17.50				
18.00	Departure to dinner	Poster session,		
19.00		reception	Conference dinner	
20.00				
21.00				
22.00				
23.00				







Monday, June 16—PhD School

d Venue: Gydehutten, U47

- **08.00:** Registration, breakfast
- 08.40: Stéphane Kéna-Cohen, Organic microcavities in the strong light-matter coupling regime
- 10.00: Mini break
- 10.10: Antonio I. Fernández-Domínguez, Electromagnetic field quantization in nanophotonic systems
- 11.30: Coffee break
- 12.00: Nicolas Ubrig, Exciton-polaritons in atomically thin materials
- 13.20: Lunch break
- 14.50: Michael Ruggenthaler, Quantum-electrodynamical density-functional theory: an introduction to a novel framework to control materials phenomena
- 16.10: Coffee break
- **16.40:** Karol Kołataj, Advances in fabrication techniques for strong coupling: harnessing DNA nanotechnology in nanophotonics
- 18.00: Dinner at Storms Pakhus







Tuesday, June 17

Venue: Gydehutten, O100

- **08.00:** Registration, breakfast
- **08.30:** Welcome
- 08.40 09.30: Chair: Christos Tserkezis
- **08.40:** Keynote talk: Thomas Ebbesen, Manipulating matter by strong coupling to the vacuum electromagnetic field

09.30: Mini break

- 09.40 10.50 : Tuesday early morning session—Chair: Christoph Lienau
- 09.40: Invited talk: Bill Barnes, Exciton de-localisation and strong coupling: work in progress
- **10.10:** May Godsi, Revealing the nature of high-order cavity polaritons under the coupling-decoupling transition
- 10.30: Dominik Sidler, On the origin of local chemical modifications under collective strong coupling

10.50: Coffee break

- 11.20 12.30 : Tuesday late morning session—Chair: Bill Barnes
- **11.20:** <u>Invited talk:</u> Christoph Lienau, Two-dimensional electronic spectroscopy probes strong coupling: what can we learn?
- **11.50:** Xabier Arrieta, Ultra-strong coupling between optical phonons and surface plasmon polaritons observed in real space
- **12.10:** Thomas Schnappinger, Cavity-Born-Oppenheimer Hartree-Fock: molecules under vibrational-strongcoupling
- 12.30: Lunch break
- 14.00 15.30 : Tuesday early afternoon session—Chair: Sara Núñez-Sánchez
- 14.00: Invited talk: Joel Yuen-Zhou, When do molecular polaritons behave like optical filters?
- 14.30: Jussi Toppari, Suppression of strong coupling effects by thermal disorder
- 14.50: Anael Ben-Asher, Strong coupling with the cavity antiresonances: forming polaritons with imaginary rather than real Rabi splitting
- 15.10: Pengfei Frank Huo, Theory and quantum dynamics of exciton polariton transport

15.30: Coffee break

- 16.00 18.00 : Tuesday late afternoon session—Chair: Joel Yuen-Zhou
- **16.00:** <u>Invited talk:</u> Timur Shegai, Strong light-matter coupling: from self-hybridized polaritons to Casimir self-assembly
- **16.30:** Frederik Schröder, Strong coupling between a dielectric nanocavity and a monolayer transition metal dichalcogenide
- 16.50: Jonas M. Peterson, Direct imaging of polariton relaxation dynamics and interactions
- 17.10: Michael Papachatzakis, Giant Rabi splitting and polariton photoluminescence in an all solutiondeposited dielectric microcavity
- 17.30: <u>Invited talk</u>: Sara Núñez Sánchez, Bioinspired free-cavity exciton polaritons with J-aggregates for light harvesting

18.00 - 21.00: Poster session, reception







Wednesday, June 18

Venue: Gydehutten, O100

08.00: Breakfast

08.40 - 09.30: Chair: N. Asger Mortensen

08.40: Keynote talk: Päivi Törmä, Strong coupling and topological phenomena in plasmonic lattices

09.30: Mini break

- 09.40 10.50 : Wednesday early morning session—Chair: Joel D. Cox
- **09.40:** <u>Invited talk:</u> Javier Aizpurua, Addressing ultrastrong light-matter interaction of organic molecular layers in optical and infrared cavities
- 10.10: Rakesh Arul, Nanophotonics to control mid-infrared light and polariton condensation
- $\textbf{10.30: Carla Est\'evez Varela, Plexcitonic nanorattles based on J-aggregates as ultra-efficient SERS-tags$
- 10.50: Coffee break
- 11.20 12.30 : Wednesday late morning session—Chair: Javier Aizpurua
- 11.20: Invited talk: Klaus Mølmer, Quantum optics with radiation on the move
- 11.50: Ben Yuen, Towards exact quantum electrodynamics in plasmonic systems
- 12.10: Yassir El Moutaoukal, Chiral strong coupling quantum electrodynamics Hartree-Fock

12.30: Lunch break

- 14.00 15.10 : Wednesday early afternoon session—Chair: Nicolò Maccaferri
- 14.00: Invited talk: Libai Huang, Ultrafast imaging of nonlinear exciton polariton interactions
- 14.30: Luca Sortino, Nonlinear polaritons in van der Waals heterostructure metasurfaces
- 14.50: Ishita Jena, Quantum dynamics in coupled plasmonic cavities

 $15.10: \ Coffee \ break$

- 15.40 16.50 : Wednesday late afternoon session—Chair: Timur Shegai
- 15.40: Invited talk: Nicolò Maccaferri, Ultrafast collapse of molecular polaritons
- 16.10: Aleesha George, Ultrafast spectroscopy reveals (dis)order-driven polariton dynamics
- 16.30: Fritz Joshua Schnur, Sub-5-fs pump-probe spectroscopy on strongly coupled exciton-cavity-polaritons

16.50: Departure to boat trip

- 17.00 19.00: Social activity: boat trip
- 19.00 23.00 : Conference dinner at Sortebro Kro







Thursday, June 19

Venue: Gydehutten, O100

08.00: Breakfast

08.40 – 10.20 : Thursday early morning session—Chair: P. André D. Gonçalves

- **08.40:** <u>Invited talk:</u> <u>Vinod Menon</u>, <u>SMILES</u>—a plug and play platform for molecular polariton condensates</u>
- **09.10:** Darius Urbonas, Condensation of cavity exciton-polaritons in perovskite nanocrystals at roomtemperature
- 09.30: Akshaya G, Room-temperature polaritons in a tunable open microcavity using 2D hybrid perovskites
- 09.50: Invited talk: Daniele Sanvitto, Polariton waveguides for nonlinear optical phenomena
- $10.20: \ Coffee \ break$
- 10.50 12.40 : Thursday early morning session—Chair: Daniele Sanvitto
- **10.50:** <u>Invited talk:</u> Marzena Szymańska, Driven-dissipative superfluids: a compact Kardar-Parisi-Zhang dynamics of the phase
- 11.20: Pietro Tassan, On-chip ultrafast polariton transistors
- 11.40: David Hagenmüller, Polariton-assisted hopping in amorphous non-conducting polymers
- **12.00:** Daniel J. Tibben, Extending the self-discharge time of Dicke quantum batteries using molecular triplets
- **12.20:** Girish Lakhwani, Device physics of organic solar cells under strong light-matter coupling
- 12.40: Lunch break
- 14.00 15.30 : Thursday early afternoon session—Chair: Marzena Szymańska
- 14.00: <u>Invited talk</u>: Jaime Gómez Rivas, Double-probe near-field THz microscopy for probing dark modes and strong light-matter coupling
- 14.30: Marco Marangi, Metasurface-enhanced superradiance in strongly-coupled J-aggregates
- **14.50:** Philip Thomas, Generating phase singularities using surface exciton polaritons in an organic natural hyperbolic material
- **15.10:** Mikhail Vasilevskiy, Fano resonances between the emission of J-aggregate-covered metallic nanorods and spherical cavity modes
- 15.30: Coffee break
- 16.00 17.10 : Thursday late afternoon session—Chair: Antonio I. Fernández-Domínguez
- 16.00: Invited talk: Francisco J. García Vidal, Mapping between quantum optics and nanophotonics
- 16.30: Kavya S. Mony, Electrical conductivity in amorphous polymers under vibrational strong coupling
- 16.50: Dmitry Dovzhenko, Extended lasing states in electrically tuneable liquid-crystal microcavity
- **17.10:** Break
- 17.20 18.00: Awards & discussion







Best student talk competition

The following talks enter the best student talk competition, sponsored by Nanophotonics. One prize of 2500 DKK will be offered after evaluation by a scientific committee. The winner will be announced at the closing discussion on Thursday.

- 1. May Godsi, Revealing the nature of high-order cavity polaritons under the coupling-decoupling transition
- 2. Xabier Arrieta, Ultra-strong coupling between optical phonons and surface plasmon polaritons observed in real space
- 3. Frederik Schröder, Strong coupling between a dielectric nanocavity and a monolayer transition metal dichalcogenide
- 4. Jonas M. Peterson, Direct imaging of polariton relaxation dynamics and interactions
- 5. Michael Papachatzakis, Giant Rabi splitting and polariton photoluminescence in an all solution-deposited dielectric microcavity
- 6. hyperlinkMoutaoukalYassir El Moutaoukal, Chiral strong coupling quantum electrodynamics Hartree-Fock
- 7. Ishita Jena, Quantum dynamics in coupled plasmonic cavities
- 8. Aleesha George, Ultrafast spectroscopy reveals (dis)order-driven polariton dynamics
- 9. Fritz Joshua Schnur, Sub-5-fs pump-probe spectroscopy on strongly coupled exciton-cavity-polaritons
- 10. Akshaya G, Room-temperature polaritons in a tunable open microcavity using 2D hybrid perovskites
- 11. Pietro Tassan, On-chip ultrafast polariton transistors
- 12. Daniel J. Tibben, Extending the self-discharge time of Dicke quantum batteries using molecular triplets
- 13. Marco Marangi, Metasurface-enhanced superradiance in strongly-coupled J-aggregates
- 14. Kavya S. Mony, Electrical conductivity in amorphous polymers under vibrational strong coupling







Poster Session

17 June, $18.00\mathchar`-20.00\mathchar`-Campustorvet, in front of O100$

Please carefully read the following guidelines to prepare for your participation:

Poster specifications: —Portrait orientation, pdf format

Printing of the posters: Please note that due to regulations of the University of Southern Denmark, posters *must* be printed at the University of Southern Denmark. The printing will be free of charge for you, and we will ensure that your poster is printed and ready for the exhibition.

Latest 10 June 10 AM: Upload your poster (pdf format) to the conference system or send the poster to loju@mci.sdu.dk!

When and where: Campustorvet, just in front of O100

Poster Setup: All posters will be placed by the organisers, provided that you have sent them in due time for printing.

Best Poster Award of DKK 2500: 2 poster prizes, sponsored by Nanophotonics, will be offered. A scientific committee will evaluate the posters. Each poster presenter will have an interview with one or more committee members during the poster session. The committee will assess the posters based on content, presentation, and the presenter's ability to communicate their research effectively. The winners of the Best Poster Awards will be announced at the closing discussion on Thursday. Make sure to attend the award ceremony for the announcement and to celebrate with your peers!

Miscellaneous: Your poster can remain exposed until the end of the conference. Please plan to remove your poster by the closing hours of the last day.







List of Posters

- 1. Abdelouadoud Mammeri, Hyperspectral camera for inline spectral analysis of organic solar cells
- 2. Ahmed Gaber Abdelmagid, Organic polaritons enable high-responsivity, angle-independent narrowband microcavity photodiodes
- 3. Alexandra R. Wright, Understanding the role of molecular aggregation in polariton formation
- 4. Ángel Giménez-Mascarell, Hexagonal silica microsphere monolayers supporting surface phonon-polaritons for radiative cooling applications
- 5. Angus Crookes, Multimode strong coupling in plasmonic nanocavities
- 6. Ann-Céline Zimmermann, Terahertz control in a transmission electron microscope
- 7. Arpan Dutta, Organic excitonic nanostructures for weak and strong coupling
- 8. Arun Kumar Kanakati, Simulating nuclear quantum effects in electronic strong coupling
- 9. Avishek Sarbajna, Optoelectronic response of light-matter interaction in ultrathin excitonic systems
- 10. Carlos Bustamante, Topology optimization of optical devices and their potential application in polaritonic chemistry
- 11. P. Elli Stamatopoulou, A multi-scale approach for modeling strong coupling in open nanocavities probed by electrons
- 12. Fathimath Faseela, Polaritonic engineering in non-fullerene acceptor organic photovoltaic devices
- 13. Florian Maurer, Quantum description of plasmon polaritons in the PZW representation
- 14. Frieder Lindel, Strong coupling between periodic arrays of quantum emitters and periodic light
- 15. Gal Sandik, Exploring the multiple scattering of organic exciton-polaritons using interferometric imaging
- 16. Hanlin Fang, Confining excitons in semiconducting moiré superlattices
- 17. Hassan Ali Qureshi, TBA
- 18. Helena Poulose, Towards nonlinear spectroscopic studies of organic molecules under vibrational strong coupling
- 19. Hira Asif, Stark control of plexcitonic states in incoherent quantum systems
- 20. Jacob Horak, Analytic model reveals local molecular polarizability changes induced by collective strong coupling
- 21. Jakub Skórka, Quantisation of electromagnetic field near dispersive resonators
- 22. Jie Ji, Phase topology and group index of THz vibro-polaritons in metasurfaces
- 23. José N. Gama, Surface exciton polaritons from visible to the near-infrared supported by nolecular materials
- 24. R. Kevin Kessing, Cooperative effects in vibrational polaritons require environment-induced nonequilibrium steady states
- 25. Kuljeet Kaur, Charge transport engineering in organic semiconductors through strong coupling in MOSFET-based Fabry-Pérot cavities
- 26. Lukas Freter, Exact numerical solution of the dissipative Tavis–Cummings model in the context of superradiance
- 27. Luke Hands, Causality for the second quantisation of open non-Hermitian nanophotonic systems





- 28. Marcelo Barreiro, Surface and cavity exciton-polaritons in J-aggregate films
- 29. Martijn Wubs, Collective single-photon emission mediated by surface plasmon polaritons and disturbed by phonons
- 30. Michael Reitz, Nonlinear semiclassical spectroscopy of ultrafast molecular polariton dynamics
- 31. Mikkel Have Eriksen, Chiral light-matter dynamics with thermal magnetoplasmons in graphene nanodisks
- 32. Nicola Peruffo, The nature of excited states relaxation in plexcitonic materials
- 33. Nikolaos Kyvelos, Mesoscopic phenomena and fabrication imperfections in plasmonic nanolens: impact on chiral light-matter interactions
- 34. Oleg V. Kotov, Lifshitz-Lorentz theory for cavity-modified ground state of a polaritonic system
- 35. Olli Siltanen, A unified quantum master equation model of cavity-coupled OLEDs
- 36. Pedro Ninhos, Tunable exciton polaritons in band-gap engineered hexagonal boron nitride
- 37. Rahul Bhuyan, The role of the size of the exciton reservoir and polariton states on strong exciton photon coupling dynamics
- 38. Rana Lomlu, Vast polaritonic enhancement of donor-acceptor energy transfer
- 39. Sandeep Singh, Morphology-driven light-matter interactions in semiconductor polaritonic systems—a review
- 40. Santiago A. Gomez, The quantum palette of methylene blue: unveiling vibronic deatures and the influence of dimerization and CB[7] encapsulation towards strong coupling applications
- 41. Sebastian Montillo, Theoretical insights into the resonant suppression effect in vibrational polariton chemistry
- 42. Sebastian Toivonen, Hierarchical pure states approach to non-linear spectroscopy of polaritonic quantum batteries
- 43. Sergii Morozov, An atlas of photonic and plasmonic materials for cathodoluminescence microscopy
- 44. Sinan Bascil, Quantifying the effect of vibrational strong coupling on hydrogen bonding
- 45. Sricharan Raghavan-Chitra, High-Q microresonators unveil quantum rare events
- 46. Sven Ebel, Cathodoluminescence by backscattered and secondary electrons
- 47. Therese Karmstrand, A quantum description of the dissipative properties of cavity polaritons in the ultrastrong coupling regime
- 48. Thomas F. Allard, Mirror-induced effects in cavity polaritonics: influence on edge states
- 49. Timo Leppälä, Organic microcavity polariton dynamics with non-Markovian quantum state diffusion
- 50. Ville Tiainen, Selectively guiding excitation energy by collaborative strong light-matter coupling
- 51. Vladislav Yu. Shishkov, Sympathetic mechanism for vibrational condensation enabled by polariton optomechanical interaction
- 52. Wenhua Zhao, Feedback cooling of fermionic atoms in optical lattices
- 53. Yu-Chen Wei, Enhanced delayed fluorescence in non-local netasurfaces: the role of electronic strong coupling







Abstracts







Monday, 16 June: PhD school







Organic microcavities in the strong light–matter coupling regime

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Presentation type: Invited school lecture

We will give a historical perspective on the physics of organic microcavities in the strong excitonphoton coupling regime. The emphasis will be on the collective strong coupling regime, where many molecules are strongly coupled to a small set of cavity modes. In this regime, new eigenstates of the system called exciton-polaritons emerge, which possess a number of peculiar properties. The description will focus on the macroscopic quantum phenomena that can emerge in the high density regime such as Bose-Einstein condensation and superfluidity.







Electromagnetic field quantization in nanophotonic systems

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Presentation type: Invited school lecture

In this talk, I will review the general problem of quantizing electromagnetic fields in nanophotonic systems. Starting from the framework of macroscopic QED [1], I will discuss its application to systems involving single quantum emitters coupled to dielectric microcavities, plasmonic nanocavities [2], and more complex hybrid metallo-dielectric resonators [3]. I will then address the extension of this formalism to ensembles of quantum emitters [4], highlighting the emergence of photon-mediated interactions among them [5]. Finally, I will explore the preparation and probing of nanophotonic systems using modulated electron beams [6], with particular emphasis on the role of the extended character of electron wavefunctions within the QED framework.

[1] J. Feist et al., Macroscopic QED for quantum nanophotonics: emitter-centered modes as a minimal basis for multiemitter problems, Nanophotonics 10, 477 (2021).

[2] R. Sáez-Blázquez et al., Plexcitonic quantum light emission from nanoparticle-on-Mirror cavities, Nano Lett. 22, 2365 (2022).

[3] I. Medina et al., Few-mode field quantization of arbitrary electromagnetic spectral densities, Phys. Rev. Lett. 126, 093601 (2021).

[4] A. Miguel-Torcal et al., Multiqubit quantum state preparation enabled by topology optimization, Optica Quant. 2, 371 (2024).

[5] J. Abad-Arredondo and A. I. Fernández-Domínguez, *Electron-assisted probing of polaritonic light-matter states*, Nanophotonics 13, 2015 (2024).

[6] J. Abad-Arredondo and A. I. Fernández-Domínguez, *Quantum state preparation and readout with modulated electrons*, Phys. Rev. B 111, 165422 (2025).







Exciton-polaritons in atomically thin materials

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Presentation type: Invited school lecture

Two-dimensional materials and semiconductors are a exciting platform for exploring light-matter interactions in a highly tunable environment. While inorganic 2D semiconductors lie beyond the primary focus of this summer school, they share key characteristics with organic materials. In this lecture, we will introduce the fundamental properties of excitons and polaritons atomically thin semiconductors and their interfaces. This aim is to generate meaningful connections and dialogue between the communities working on organic and inorganic systems.







Quantum-electrodynamical density-functional theory: an introduction to a novel framework to control materials phenomena

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Presentation type: Invited school lecture

In this lecture I will discuss the general framework of quantum-electrodynamical density-functional theory and show applications specifically for equilibrium modifications of material properties in photonic environments. I will first derive from fundamental principles the Pauli-Fierz Hamiltonian, which allows a mathematically well-defined description of non-perturbatively-coupled light-matter systems. I will then show how a density-functional reformulation allows an efficient and accurate way to solve this quantum-field problem non-perturbatively, before highlighting some applications. The lecture will follow the general ideas presented in Ref. [1].

[1] M. Ruggenthaler et al., Understanding polaritonic chemistry from ab initio quantum electrodynamics, Chem. Rev. 123, 11191 (2023).







Fabricating plasmonic platforms for strong coupling: from plasmonics to DNA origami

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Presentation type: Invited school lecture

Strong coupling between light and matter arises when the interaction strength between an optical mode and a quantum emitter exceeds their respective loss rates, leading to the formation of hybrid light-matter states known as polaritons. Achieving this regime requires intense local electromagnetic fields, ultra-small optical mode volumes, and precise spatial alignment between the field and the emitter.

Plasmonic nanostructures are particularly well suited for this purpose due to their ability to confine light far below the diffraction limit and generate strong near fields via surface plasmon resonances. In this talk, the key physical parameters that govern strong coupling will be introduced, and the role of plasmonic cavities in enhancing the local optical environment to enable this interaction will be discussed.

A central challenge in these systems—precise control over the number and position of emitters in the plasmonic hotspot—will be addressed using DNA origami nanotechnology. This programmable molecular platform offers nanometer-scale spatial control over cavity assembly and emitter placement, enabling systematic exploration of strong coupling at the single-emitter level.







Tuesday, 17 June






Manipulating matter by strong coupling to the vacuum electromagnetic field

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Presentation type: Invited keynote talk

Over the past decade, the possibility of manipulating material and chemical properties by using hybrid light-matter states has stimulated considerable interest [1,2]. Recent progress in understanding why vibrational strong coupling has such large impact on molecular properties will be discussed together with the technical challenges in studying chemistry in microfluidic cavities.

[1] F. J. Garcia Vidal et al., *Manipulating matter by strong coupling to vacuum fields*, Science 373, eabd336 (2021).

[2] K. Nagarajan et al., Chemistry under vibrational strong coupling, J. Am. Chem. Soc. 143, 16877 (2021).







Electromagnetic field quantization in nanophotonic systems

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Presentation type: Invited talk

We report an investigation into the de-localisation of excitons in a conjugated polymer under conditions of strength coupling. Conjugated polymers are interesting in this regard because there is a long-standing theoretical prediction of a spectral signature arising from such de-localisation; i.e. that the relative strength of the 0-0 and 0-1 vibrational components in the photoluminescence spectrum will change with the extent of de-localisation. We employ an 'open' system involving conjugated polymers coupled to a surface plasmon mode. Early results will be reported, as part of a 'work in progress'; what remains to be done will be described, and a number of open questions will be identified.







Revealing the nature of high-order cavity polaritons under the coupling–decoupling transition

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Presentation type: Contributed talk

When organic molecules are embedded in an optical cavity, their interaction with light can be significantly enhanced. Once the interaction overcomes the decay processes, the system enters the strong coupling regime, where molecular wavefunctions are reshaped, leading to the formation of new quantum states called polaritons [1,2]. In the realm of strong coupling, it is often assumed that a single (dispersive) photonic mode interacts with the material, which is correct in the case of a half-wavelength cavity. But what happens in larger cavities supporting several longitudinal modes, as illustrated in Fig. 1(a)? How, then, do the molecules interact with multiple photonic modes? Recently, we predicted theoretically [3] and demonstrated experimentally [4] that in this scenario, the interaction can take one of two distinct forms, depending on the system parameters. One mechanism involves the coherent mixing of multiple cavity modes through their mutual interaction with the material, resulting in a mid-polariton branch. In the other mechanism, each cavity mode independently couples to the molecules, leading to separate, decoupled polaritonic branches. Surprisingly, one of these parameters is the cavity size — a factor that is usually ignored in cavity QED but proves crucial here. In our study, we observed the two coupling mechanisms, as well as the transition between them by gradually increasing the cavity size while maintaining a fixed coupling strength (shown in Fig. 1(b)). Moreover, we explored the system's nature through photoluminescence spectroscopy, revealing unexpectedly pronounced upper polariton emission not commonly seen in single-mode cavities (Fig.1(c)). Finally, we investigated the ultrafast dynamics of this system by performing pump-probe spectroscopic measurements, revealing significant changes in the transient spectra throughout the transition. Our findings demonstrate a new level of control over the photophysical properties of strongly coupled systems, with important consequences for longrange intermolecular energy transfer mediated by high-order polaritons in multimode cavities [5].







As such, they hold significant potential for novel organic photovoltaic architectures. Furthermore, controlling exciton-mediated interaction between different photonic modes offers a promising tool for quantum information processing and polariton condensation dynamics in multimode cavities. We stress that the concept we present is not limited to molecular systems but can rather be applied generally in all strongly coupled systems.



FIG. 1. (a) A sketch of a multimode cavity: two 35 nm Ag mirrors with a TDBC-doped polymer spacer. The cavity length supports multiple longitudinal modes, with the first four shown. (b) and (c) Reflectivity and emission measurements respectively, of multimode cavities show mid-polaritons below the transition (628 nm) on the left and independent polaritons above (1615 nm) on the right. TMM simulations (white lines) reveal a Rabi splitting of ~ 128 meV.

 T. W. Ebbesen, Hybrid light-matter states in a molecular and material science perspective", Acc. Chem. Res. 49, 2403 (2016).

M. Hertzog et al., Strong light-matter interactions: a new direction within chemistry, Chem.
Soc. Rev. 48, 937 (2019).

[3] M. Balasubrahmaniyam et al., Coupling and decoupling of polaritonic states in multimode cavities, Phys. Rev. B 103, L241407 (2021).

[4]] M. Godsi et al., Exploring the nature of high-order cavity polaritons under the couplingdecoupling transition, J. Chem. Phys. 159, 134307 (2023).

[5] K. Georgiou et al., Ultralong-range polariton-assisted energy transfer in organic microcavities, Angew. Chem. Int. Ed. 60, 16661 (2021).







On the origin of local chemical modifications under collective strong coupling

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Presentation type: Contributed talk

Despite recent numerical evidence, one of the fundamental theoretical mysteries of polaritonic chemistry is how and if collective strong coupling can induce local changes of the electronic structure to modify chemical properties. First, we present non-perturbative analytic results for a model system consisting of an ensemble of N harmonic molecules under vibrational strong coupling (VSC) that alters our present understanding of this fundamental question [1]. Within the framework of cavity Born–Oppenheimer Hartree–Fock [2], we discover that the electronic molecular polarizabilities are modified even in the case of vanishingly small single-molecule couplings [1]. Consequently, we can demonstrate analytically that this local polarization mechanism persists even in the large-N limit. Our non-perturbative analytic observations demonstrate that hitherto existing collective-scaling arguments are insufficient for polaritonic chemistry [1]. In the dark, i.e., without an externally applied field, things become more subtle and the complex (anharmonic) electronic structure starts to play a significant role. Numerical evidence reveals the emergence of a cavity-induced polarization pattern that persists under collective strong coupling conditions, i.e., a *polarization glass* is born (Fig. 1) [3]. Developing this theoretical concept further has astonishing consequences, when going beyond the dilute gas approximation. In the quasi-dilute limit, we additionally allow for inter-molecular dispersion effects that are introduced by the dipole self-energy [4]. The resulting energy minimization problem is degenerate [3,4,5] and strongly resembles the iconic Sherrington-Kirkpatrick model of a spin glass [4]. This establishes a fundamental theoretical link between the seemingly unrelated fields of polaritonic chemistry and spin glasses. Exploring its profound implications for the theoretical framework of polaritonic chemistry, our mapping suggests a collectively







induced instability in the dressed electronic structure (spontaneous replica symmetry breaking), which could provide the long-sought seed for significant local chemical modifications in polaritonic chemistry (a nucleation-like effect). Moreover, our mapping paves the way to incorporate, adjust, and probe numerous spin glass concepts in polaritonic chemistry, such as frustration, aging dynamics, excess of thermal fluctuations, time-reversal symmetry breaking, or stochastic resonances [4].



FIG. 1. Pictorial representation of a cavity-induced molecular polarization glass from vibrational strong coupling.

[1] J. Horak et al., Analytic model reveals local molecular polarizability changes induced by collective strong coupling in optical cavities, arXiv:2401.16374 (2024).

[2] T. Schnappinger et al., Cavity Born-Oppenheimer Hartree-Fock ansatz: light-matter properties of strongly coupled molecular ensembles, J. Phys. Chem. Lett. 14, 8024 (2023).

[3] D. Sidler et al., Unraveling a cavity-induced molecular polarization mechanism from collective vibrational strong coupling, J. Phys. Chem. Lett. 15, 5208 (2024).

[4] D. Sidler et al., The connection of polaritonic chemistry with the physics of a spin glass, arXiv:2409.08986, (2024).

[5] C. M. Bustamante et al., The relevance of degenerate states in chiral polaritonics, J. Chem.
Phys. 161, 244101 (2024).







Two-dimensional electronic spectroscopy probes strong coupling: what can we learn?

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Presentation type: Invited talk

Many of the exciting properties of novel types of quantum materials, from polariton condensation to light-induced superconductivity arise from strong couplings among electrons, phonons, spins and photons in the material. These couplings give rise to the formation of new hybrid quasi-particles, with optical and electronic properties that can be vastly different from those of the uncoupled excitations. Two-dimensional electronic spectroscopies (2DES), replacing the excitation pulse in common pump-probe spectroscopies by a phase-locked pulse pair, can give unique insights into such couplings. Cross peaks in the 2DES, oscillating as a function of time delay between pump pair probe, provide definite spectroscopic signatures for such couplings, making 2DES with fewfemtosecond time resolution a method of choice for unraveling the microscopic physical processes that govern quasi-particle formation in quantum materials. Recent progress in time resolution, spectral bandwidth and sensitivity [1-3] have allowed to apply this method to strongly coupled polariton system based on organic materials and to probe the ultrafast coherent Rabi oscillations, the hall mark of strong coupling, directly in the time domain [4,5].

In my talk I will give an overview of these results. Specifically, I will discuss the new information that can be gained about the optical nonlinearities of excitons in J-aggregated organic semiconductors that are strongly to surface plasmon polaritons [4,6,7]. 2DES data show how strong coupling transforms the usual diffusive hopping exciton transport in organics into a coherent transport of excitons over mesoscopic distances at room temperature. I will compare these results to first, still unpublished studies of strong couplings between the excitons in a monolayer of WS₂ [8] to a plasmonic nanoresonator. The experimental results show that strong coupling forms new hybrid particles with optical nonlinearities that are vastly enhanced over those of both monolayer excitons and plasmons. 2DES reveals the role of many-body correlations for these enhanced nonlinearities, opening up new pathways for tailoring optical nonlinearities in hybrid systems based on atomically thin semiconductors. All these examples give strong indications that many-body interactions are crucial for understanding the optical nonlinearities of such hybrid systems.







[1] D. Timmer et al., *Phase-cycling and double-quantum two-dimensional electronic spectroscopy using a common-path birefringent interferometer*, Optica 11, 1646 (2024).

[2] D. Timmer et al., Full visible range two-dimensional electronic spectroscopy with high time resolution, Opt. Express 32, 835 (2024).

[3] D. Timmer et al., Disentangling signal contributions in two-dimensional electronic spectroscopy in the pump-probe geometry, J. Chem. Phys. 162, 124202 (2025).

[4] D. Timmer et al., Plasmon mediated coherent population oscillations in molecular aggregates, Nat. Commun. 14, 8035 (2023).

[5] N. Peruffo et al., Coherent dynamics in solutions of colloidal plexcitonic nanohybrids at room temperature, Adv. Opt. Mater. 11, 2203010 (2023).

[6] D. Timmer et al., Charge delocalization and vibronic couplings in quadrupolar squaraine dyes, J. Am. Chem. Soc. 144, 19150 (2022).

[7] T. Quenzel et al., *Plasmon-enhanced exciton delocalization in squaraine-type molecular aggre*gates, Acs Nano 16, 4693 (2022).

[8] D. Timmer et al., Ultrafast coherent exciton couplings and many-body interactions in monolayer
WS₂, Nano Lett. 24, 8117 (2024).







Ultra-strong coupling between optical phonons and surface plasmon polaritons observed in real space

<u>Xabier Arrieta^{1,*}</u>, Edoardo Vicentini², Martin Schnell^{2,3}, Maria Barra Burillo², Andrei Bylinkin², Rainer Hillenbrand^{2,3}, Ruben Esteban^{1,4}, and Javier Aizpurua^{3,4,5}

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Presentation type: Contributed talk

The interaction between light and excitations in a material can be enhanced beyond the dissipation rates, thus reaching the strong coupling regime, which leads to the emergence of hybrid light-matter states known as polaritons. This hybridization of states can modify the properties of materials, and enables exciting potential applications in polaritonic chemistry, quantum technologies and nonlinear optics. Additionally, when the coupling strength becomes comparable to the energy of the excitations, the system enters the ultra-strong coupling (USC) regime, leading to new quantum effects, such as the modification of the ground state [1]. To reach USC in the nanoscale, both plasmonic and phononic resonances have been considered favourable. However, coupling these two types of excitations in the same system is challenging because they are usually characterized by very different frequencies.

In this work, we present the first real-space observation of vibrational USC between Transverse Optical (TO) phonons and surface plasmon polaritons (SPP). Specifically, we analyze a 50 nm-thick SiC layer on an InAs substrate, showing that we can achieve USC with a tiny amount of material, in contrast to the large volumes typically required in microcavities. To probe the polaritons, we combine scattering-type scanning near-field optical microscopy (s-SNOM) with pump-probe Fourier-transform infrared spectroscopy [2] (Fig. 1(a)). The near-infrared pump laser enables dynamic control of the doping level in the InAs, thereby tunning the SPP frequency to infrared frequencies so that it can couple with the TO phonon. Phase spectra measurements of the coupled system (Fig. 1(b,e)) reveal a large splitting between the upper and lower polaritons, which results from the coupling between the InAs SPP (Fig. 1(c,f)) and the SiC TO phonon (Fig. 1(d,g)). The large polaritonic splitting provides experimental evidence that the system is in the USC regime.









FIG. 1. (a) Schematic representation of the experimental pump-probe setup used to measure the polaritons of a 50 nm-thick SiC layer on an InAs substrate. The pump laser can tune the SPP of the InAs to the TO phonon of the SiC, while the probe laser excites the polaritons. (b,c,d) Sketch of the experimental geometry, (e,f,g) phase spectra measurements and (h,i,j) imaginary part of the calculated reflection coefficients. (b,e,h) Correspond to the bare InAs substrate, (c,f,i) to the 50 nm SiC layer, and (d,g,j) to the full system. A significant splitting between the upper and lower polariton is observed for the latter.

To further characterize the system, we complement the experimental measurements with a theoretical analysis of the coupling between the 50 nm-thick SiC layer and the InAs substrate. Calculations of the reflection coefficients (Fig. 1(h,i,j)) are in very good agreement with the phase spectra measurements (Fig. 1(e,f,g)), and corroborate the emergence of the upper and lower polaritons due to the coupling between the SiC TO and InAs SPP. The phonon-plasmon coupling strength $(g \approx 100 \text{ cm}^{-1})$, calculated from a coupled harmonic oscillator model [3], confirms that the system is in the USC regime. Additionally, we find that both the TO phonon and the SPP contribute significantly to the coupled polaritons, with an approximate 40-60 hybridization ratio, as expected in USC. Our findings provide a deeper understanding of the USC phenomenon and may open new possibilities for its applications in nanophotonics and quantum technologies, for example by facilitating the modification of the properties of molecules or a material.

 A. F. Kockum et al., Ultrastrong coupling between light and matter, Nat. Rev. Phys. 1, 19 (2019).

[2] M. Wagner et al., Ultrafast dynamics of surface plasmons in InAs by time-resolved infrared nanospectroscopy, Nano. Lett. 14, 4529 (2014).

[3] U. Muniain et al., Description of ultrastrong light-matter interaction through coupled harmonic oscillator models and their connection with cavity-QED Hamiltonians, Nanophotonics, DOI: 10.1515/nanoph-2024-0528 (2025).







Cavity-Born-Oppenheimer Hartree-Fock: molecules under vibrational-strong-coupling

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Presentation type: Contributed talk

When molecules are placed in a non-classical photonic environment, such as optical or nanoplasmonic cavities, it is possible to form strong light-matter coupled hybrid states called polaritons. Recent experiments show that this strong light-matter coupling is capable of modifying chemical and physical properties and offers a possible novel approach to control chemical reactions. The situation in which the quantized cavity modes are coupled via their characteristic frequency to vibrational degrees of freedom of molecules is called vibrational strong coupling (VSC). In the VSC regime, the chemistry of a single electronic state (mostly the ground state), its vibrational spectroscopy, and properties are influenced by the cavity interaction.

In this contribution, I will discuss how the ab initio Cavity-Born-Oppenheimer-Hartree-Fock (CBO-HF) approach [1] can be used to study the effect of VSC on the molecular properties of single molecules and small ensembles of molecules. Our ab-initio treatment allows us to study the interactions between molecules mediated by the cavity. These interactions give rise to local strong-coupling effects that hint at a possible modification of the chemical reactivity under VSC. Moreover, we observe local changes in both the permanent dipole moment and the static polarizability induced by collective effects under strong-coupling conditions in molecular ensembles.

Extending the CBO-HF framework [2] we developed tools to study how molecular structure changes under strong coupling conditions. By optimizing small molecules resonantly coupled to cavity photon modes, we emphasize the critical importance of reorientation and geometric relaxation. Both effects are currently neglected in most computational studies. Therefore, the interpretation of computational studies based on fixed molecular structures and fixed orientation should be treated with caution. To explain our observations, we present a simple and straightforward concept for estimating the effect of cavity interactions on molecular geometry using static polarizability and dipole moments [2].









FIG. 1. (a) and (b) vibro-polaritonic IR spectra shown in red and (c) and (d) vibro-polaritonic Raman spectra shown in blue of a single formaldehyde molecule coupled to two orthogonal cavity modes for two different cavity frequencies ω_c calculated at the CBO-HF/aug-cc-pVDZ level of theory.

Furthermore, it is possible in the CBO-HF framework to determine vibro-polaritonic IR and Raman spectra in the harmonic approximation [3]. As a demonstration, Fig. 1 shows vibro-polaritonic IR and Raman spectra of a single formaldehyde molecule coupled to two orthogonal cavity modes resonant with either the symmetric or asymmetric CH stretching modes.

Overall, the CBO-HF approach provides a promising and extensible framework for understanding and predicting molecular behavior in photonic environments inspired by the quantum chemistry toolbox.

[1] T. Schnappinger et al., Cavity Born-Oppenheimer Hartree-Fock ansatz: light-matter properties of strongly coupled molecular ensembles, J. Phys. Chem. Lett. 19, 8024 (2023).

[2] T. Schnappinger et al., Do molecular geometries change under vibrational strong coupling?, J. Phys. Chem. Lett. 15, 7700 (2024).

 [3] T. Schnappinger et al., Ab initio vibro-polaritonic spectra in strongly coupled cavity-molecule systems, J. Chem. Theory Comput. 19, 9278 (2023).





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When do molecular polaritons behave like optical filters?

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Presentation type: Invited talk

Molecular polaritons may be regarded as quantum impurity models, where the impurity is a cavity photon mode and the complex anharmonic molecular degrees of freedom serve as a bath. If this bath is large enough $(N \gg 1)$, as in the case of most molecular polariton experiments, the quantum dynamics of such a system becomes very simple to compute, as demonstrated in our recent method, Collective Dynamics using Truncated Equations (CUT-E) [1,2]. An important consequence of the latter is that ultrafast dynamics when N tends to infinity is predicted to be trivially replicated by appropriately shaped pulses outside of the cavity [3], with polaritons serving as optical filters. This negative result forces us to identify situations where polariton formation provides us with results that cannot be reproduced outside of a cavity. I will explain how longer dynamics involved in photoluminescence and other Raman processes provide examples of such situations [4]



FIG. 1. Comparison between out-of- and in-cavity photoinduced molecular processes.

If time permits, I will also briefly mention important conclusions about the role of near-field radiative heat transfer effects in ground state chemical reactivity modified by vibrational strong





coupling [5] as well as a semiclassical method to simulate nonlinear spectroscopy and microscopy of polaritons [6,7].

[1] J. B. Pérez-Sánchez et al., Simulating molecular polaritons in the collective regime using fewmolecule models, Proc. Nat. Acad. Sci. 120, e2219223120 (2023).

[2] J. B. Pérez-Sánchez et al., CUT-E as a 1/N expansion for multiscale molecular polariton dynamics, J. Chem. Phys. 162, 064101 (2025).

[3] K. Schwennicke et al., When do molecular polaritons behave like optical filters?, Chem. Soc. Rev. (2025), in press.

[4] J. B. Pérez-Sánchez and J. Yuen-Zhou, *Radiative pumping vs vibrational relaxation of molecular polaritons: a bosonic mapping approach*, Nat. Commun. 16, 3151 (2025).

[5] Z. Brawley et al., Sub-wavelength chemical imaging of a modified reaction due to vibrational strong coupling, Nat. Chem. 17, 439–447 (2025).

[6] M. Reitz et al., Nonlinear semiclassical spectroscopy of molecular polaritons, Phys. Rev. Lett. 134, 193803 (2025).

[7] P. Fowler-Wright et al., Nonlinear microscopy of polaritons, arXiV:2504.15501 (2025).







Suppression of strong coupling effects by thermal disorder

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Presentation type: Contributed talk

The strong coupling (SC) between molecules and confined light modes of optical cavities to form polaritons has been shown to alter photochemistry, but the origin of this effect is still not fully understood [1]. Although theoretical models suggest suppression of photochemistry due to the formation of new polaritonic potential energy surfaces (PES) [2], many of these models do not account for the energetic disorder among molecules, which is unavoidable under ambient conditions. In addition, the lifetime of the polariton should be sufficiently long for the reactants to evolve over the modified portions of the PES. Despite recent suggestions that the lowest energy polariton state can be very long-lived [3], polariton lifetimes are generally considered to be limited by the lifetime of the cavity photon [4], which is on the order of a few tens of femtoseconds in typical cavities.



FIG. 1. Schematic illustration (not to scale) of (a) an HBQ molecule undergoing ESIPT after obtaining excitation energy via polaritons, and (b) geometry of the utilized metallic Fabry-Pérot cavity. (c) Normalized absorption and emission spectra of HBQ within PMMA film.

To minimize the influence of lifetime and focus on the effect of polariton formation on photoreactivity, we investigate the *excited-state intramolecular proton transfer* (ESIPT) in 10-Hydroxybenzo-[h]quinoline (HBQ) molecule, which occurs faster than the decay of the utilized cavity (Fig. 1(b,c)) [5]. By combining MD simulations with fluorescence and excitation spectra measurements, we demonstrate that despite the observation of Rabi splitting suggesting that the SC regime is reached, this







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reaction is not suppressed. We attribute the lack of an effect to the thermal disorder that prevents the formation of a single lower polariton state. Instead, because all the states acquire a small photonic contribution, the PES remains the same as outside the cavity. The observation that the excitation spectrum of the strongly coupled molecule–cavity system is a product of its absorption and excitation spectrum of bare molecules, suggests that polaritons act as efficient gateways for channeling an excitation into a molecule, which then reacts normally as illustrated in the Fig. 1(a). This hints that SC provides means to tune the action spectrum, rather than to change the reaction.

Recently, we have combined HBQ with an inert molecule with strong absorption at the same energy. Cavities including both molecules undergo collaborative SC where both molecules are strongly coupled with the same cavity mode. This enables increasing the total coupling strength, which allows us to further investigate our earlier observations on the interplay between spectral overlap and thermal disorder in energy channeling [5], as well as to experimentally test the recent theoretical hypotheses [6]. In addition, the difference in chemical reactivity of the molecules is expected to result in a rapid localization of the excitation energy, initially shared among all the molecules, into one of the reactive HBQ molecules [7]. This phenomenon could be utilized for example in the enhanced light-harvesting.

 M. S. Rider and W. L. Barnes, Something from nothing: linking molecules with virtual light, Contemp. Phys. 62, 217 (2022); B. Xiang and W. Xiong, Molecular polaritons for chemistry, photonics and quantum technologies, Chem. Rev. 124, 2512 (2024).

[2] J. Galego et al., Suppressing photochemical reactions with quantized light fields, Nat. Commun.7, 13841 (2016).

[3] F. J. Garcia-Vidal et al., *Manipulating matter by strong coupling to vacuum fields*, Science 373, eabd0336 (2021).

[4] V. M. Agranovich et al., Cavity polaritons in microcavities containing disordered organic semiconductors, Phys. Rev. B 67, 085311 (2003); G. Groenhof et al. Tracking polariton relaxation with multiscale molecular dynamics simulations, J. Chem. Phys. Lett. 10, 5476 (2019).

[5] A. Dutta et al., Thermal disorder prevents the suppression of ultra-fast photochemistry in the strong light-matter coupling regime, Nat. Commun. 15, 6600 (2024).

[6] T. Liu et al., Unlocking delocalization: how much coupling strength is required to overcome energy disorder in molecular polaritons?, Chem. Sci. 16, 4676 (2025).

[7] G. Groenhof and J. J. Toppari, Coherent light harvesting through strong coupling to confined light, J. Phys. Chem. Lett. 9, 4848 (2018).





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Strong coupling with the cavity antiresonances: forming polaritons with imaginary rather than real Rabi splitting

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Presentation type: Contributed talk

Polaritons, hybrid light-matter states, emerge from the strong coupling between confined electromagnetic modes in optical cavities and quantum emitters, enabling control over optical and material properties at the quantum level [1]. Traditionally, polaritons are studied in single-mode resonators, where the photonic spectral density is well-described by simple Lorentzian peaks (Fig. 1(a)). However, advances in nanophotonics have led to a variety of complex cavity architectures, such as plasmonic and hybrid metallodielectric cavities, which enable extreme light confinement and offer various functionalities [2, 3]. These systems exhibit spectral properties that cannot be described by a simple sum of Lorentzian resonances (Fig. 1(b)), including asymmetric lineshapes, Fano resonances, and sharp dips.



FIG. 1. The coupling of a two-level emitter to (a) a single-mode cavity with a Lorentzian spectral density $(J(\omega))$ and (b) a hybrid cavity with an intricate $J(\omega)$. In (a), the emitter is on-resonance with the peak of the Lorentzian in $J(\omega)$, whereas in (b), it is on-resonance with the dip, corresponding to an antiresonance state.

In this work, we focus on the dip structures in the cavity spectral density, which are associated with antiresonance states arising from destructive interference between different photonic modes. Unlike conventional resonances, antiresonances couple to the quantum emitter with complex-valued strengths, reflecting their distinct spectral characteristics. Recently, it has been demonstrated that





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when weakly coupling a two-level emitter to a two-mode hybrid cavity featuring a dip in its photonic spectral density, an ultra-narrow polariton forms [4], enabling single-photon emission through a novel photon blockade mechanism [5]. This polariton forms through strong coupling to the cavity antiresonance, leading to imaginary rather than real Rabi splitting due to the complex coupling strength. Here, we generalize the concept of strong coupling to cavity antiresonances beyond twomode hybrid cavities and a single emitter and discuss the formation of polaritons with imaginary Rabi splitting. Furthermore, we suggest a realistic setup where a collective interaction between the antiresonance and multiple emitters leads to the formation of an ultra-narrow, low-loss polaritonic state. Remarkably, this collective polariton is longer-lived than the dark states in the system.

Our results provide a new perspective on the role of spectral dips in cavity quantum electrodynamics, expanding the understanding of polariton formation and enabling novel strategies for manipulating quantum light–matter interactions with applications ranging from quantum optics to polaritonic chemistry.

 F. J. Garcia-Vidal, et al., Manipulating matter by strong coupling to vacuum fields, Science 373, 6551 (2021).

[2] S. Cui et al., Hybrid plasmonic photonic crystal cavity for enhancing emission from near-surface nitrogen vacancy centers in diamond, ACS Photonics 2, 465 (2015).

[3] B. Gurlek et al., Manipulation of quenching in nanoantenna-emitter systems enabled by external detuned cavities: a path to enhance strong-coupling, ACS Photonics 5, 456 (2018).

[4] Y.-W. Lu et al., Unveiling atom-photon quasi-bound states in hybrid plasmonic-photonic cavity, Nanophotonics 11, 3307 (2022).

 [5] A. Ben-Asher et al., Non-hermitian anharmonicity induces single-photon emission, Phys. Rev. Lett. 130, 243601 (2023).







Theory and quantum dynamics of exciton polariton transport

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Presentation type: Contributed talk

Cavity exciton-polaritons exhibit ballistic transport and can achieve a distance of $100 \,\mu\text{m}$ in one picosecond. This ballistic transport significantly enhances mobility compared to that of bare excitons, which often move diffusively and become the bottleneck for energy conversion and transfer devices. Despite being robustly reproduced in experiments and simulations, there is no comprehensive microscopic theory addressing the group velocity of polariton transport, and its renormalization



FIG. 1. Polariton band structure modification and group velocity renormalization due to polariton-phonon interaction. (a) Modified polariton band structure under different λ . (b) Group velocity of the LP branch $\tilde{v}_{g,-}$ under different λ . (c) Scaling relation of the magnitude of LP group velocity change $|\Delta \tilde{v}_{g,-}|$ with λ . (d) Temperature-dependence of the LP group velocity $\tilde{v}_{g,-}$ at LP energy $\epsilon_{-k} = 1.86 \text{ eV}$ and $\lambda = 6 \text{ meV}$. Theoretical results (solid lines) are compared to quantum dynamics simulations (open circles).







due to phonon scattering while still preserving this ballistic behavior. In this work [1,2], we develop a microscopic theory to describe the group velocity renormalization using a finite-temperature Green's function approach. Utilizing the generalized Holstein–Tavis–Cummings Hamiltonian, we analytically derive an expression for the group velocity renormalization and find that it is caused by phonon-mediated transitions from the lower polariton (LP) states to the dark states, then scattering from dark states back to LP. As such, the dark states do not have to be populated and serve as the virtual state for super-exchange (especially true for a large light–matter detuning). The theory predicts that the magnitude of group velocity renormalization scales linearly with the phonon bath reorganization energy under weak coupling conditions and also linearly depends on the temperature in the high-temperature regime. These predictions are numerically verified using quantum dynamics simulations, demonstrating quantitative agreement. Our findings provide theoretical insights and a predictive analytical framework that advance the understanding and design of cavity-modified semiconductors and molecular ensembles, opening new avenues for engineered polaritonic devices.

[1] W. Ying et al., *Microscopic theory of polariton group velocity renormalization*, arXiv:2411.08288 (2024).

 B. X. K. Chng et al., Quantum dynamics simulations of the polariton transport process, Chem-Rxiv, DOI: 10.26434/chemrxiv-2024-mjd75 (2024).







Strong light–matter coupling: from self-hybridized polaritons to Casimir self-assembly

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Presentation type: Invited talk

Strong light-matter interactions are at the core of many electromagnetic phenomena. In this talk, I will give an overview of several nanophotonic systems which support polaritons — hybrid states of light and matter, as well as try to demonstrate their potential usefulness in applications. I will start with transition metal dichalcogenides (TMDs) and specifically discuss one-dimensional edges in these two-dimensional materials [1-2]. I will also discuss the concept of self-hybridization, a scenario in which both light and matter subparts in a polaritonic system are supported by the same (nano)structured material [1-4]. We have recently demonstrated such self-hybridization in TMD nanostructures [1-4] and levitating water droplets [4-5] across both electronic and vibrational strong coupling. The latter is interesting, due to abundance of water droplets in natural systems, including mists, fogs, and clouds. Furthermore, I will show that Fabry-Pérot resonators, one of the most important workhorses of nanophotonics, can spontaneously form in an aqueous solution of gold nanoflakes [6-8]. This effect is possible due to the intricate balance between attractive Casimir–Lifshitz forces and repulsive electrostatic forces acting between the flakes. There is a hope that this technology is going to be useful for future developments in self-assembly and polaritonics, as well as help develop a unified view of Casimir and strong light–matter coupling phenomena.

 B. Munkhbat et al., Transition metal dichalcogenide metamaterials with atomic precision, Nat. Commun. 11, 4604 (2020).

[2] B. Munkhbat et al., Nanostructured transition metal dichalcogenide multilayers for advanced nanophotonics, Laser Photon. Rev. 17, 2200057 (2022).

[3] G. Zograf et al., Combining ultrahigh index with exceptional nonlinearity in resonant transition metal dichalcogenide nanodisks, Nat. Photon. 18, 751 (2024).

 [4] A. Canales et al., Abundance of cavity-free polaritonic states in resonant materials and nanostructures, J. Chem. Phys. 154, 024701 (2021).





[5] A. Canales et al., Self-hybridized vibrational-Mie polaritons in water droplets, Phys. Rev. Lett. 132, 193804 (2024).

[6] B. Munkhbat et al., Tunable self-assembled Casimir microcavities and polaritons, Nature 597, 214 (2021).

[7] F. Schmidt et al., Tunable critical Casimir forces counteract Casimir-Lifshitz attraction, Nat.
Phys. 19, 271 (2023).

[8] B. Kücüköz et al., Quantum trapping and rotational self-alignment in triangular Casimir microcavities, Sci. Adv. 10, eadn1825 (2024).







Strong coupling between a dielectric nanocavity and a monolayer transition metal dichalcogenide

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Presentation type: Contributed talk

Monolayer transition metal dichalcogenides host excitons with high binding energies, making them compelling platforms for exploring strong–light matter interactions [1]. Novel extreme dielectric confinement (EDC) cavities, for example in InP [2], confine light on sub-wavelength length scales and support quality factors several orders of magnitude higher than their plasmonic counterparts. Combining sub-wavelength confining dielectric nanocavities with excitons in ML TMDCs is predicted to enhance exciton–exciton interactions, enabling single-photon nonlinearities such as a polariton blockade [3].

We demonstrate strong coupling between an EDC cavity and a monolayer (ML) molybdenum ditelluride (MoTe₂), see Fig.1(a) [4]. The cavity has an effective mode volume of $V_{\text{eff}} = 0.07(\lambda/\text{n})^3$, see Fig.1(c) for the normalized field profile $|\tilde{\mathbf{F}}(\mathbf{r})|$ in the plane of the MoTe2 layer. From numerical simulations in the quasi-normal mode framework based on a reaction coordinate formalism [5], we calculate a light–matter interaction strength g of 4.64 meV. Avoided crossing is demonstrated with both photoluminescence (PL) and reflection measurements, see Fig.1(b) and (d). Fitting the PL measurements with a coupled-oscillator model yields light–matter interaction strengths of $5.3 \pm 0.3 \text{ meV}$, in good agreement with the theory. The Rabi splitting E_{Rabi} at T = 40 K, corresponding to a detuning of $-0.2 \pm 0.7 \text{ meV}$, overcomes the averaged losses of the systems by more than a factor of two. In conclusion, the system is deep in the strong coupling regime. Our work creates an ideal testbed for studying sub-wavelength confined light–matter interactions and marks an important step toward single-photon nonlinearities [5].









FIG. 1. (a) Artistic representation of the ML MoTe₂ on top of the EDC cavity. (c) Simulated electric-field distribution in the plane of the ML MoTe₂. (b) PL and (d) reflection spectra as a function of detuning. The solid and dashed black lines depict the exciton and cavity mode reference, respectively. The red lines depict a fit with a coupled-oscillator model, from which the light–matter interaction strength is obtained.

[1] C. Schneider et al., Two-dimensional semiconductors in the regime of strong light-matter coupling, Nat. Commun. 9, 2695 (2018).

[2] M. Xiong et al., Experimental realization of deep sub-wavelength confinement of light in a topology-optimized InP nanocavity, Opt. Mater. Express 14, 397 (2024).

[3] E. Denning et al., Quantum theory of two-dimensional materials coupled to electromagnetic resonators, Phys. Rev. B 105, 85306 (2022).

[4] F. Schröder et al., Strong coupling between a dielectric nanocavity and a monolayer transition metal dichalcogenide, arXiv:2502.06529 (2025).

[5] E. Denning et al., Cavity-induced exciton localization and polariton blockade in two-dimensional semiconductors coupled to an electromagnetic resonator, Phys. Rev. Res. 4, L012020 (2022).







Direct imaging of polariton relaxation dynamics and interactions

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Presentation type: Contributed talk

Recently, transition metal dichalcogenides (TMDCs) have emerged as compelling platforms for exploring exciton-polaritons—quasiparticles formed through the strong coupling between excitons and cavity-confined photons—due to their simple fabrication, rich excitonic behavior, and high tunability. Importantly, their demonstrated ability to host exotic many-body phases such as roomtemperature-stable Bose–Einstein condensates [1] makes them particularly intriguing for investigating fundamental quantum phenomena and developing novel optoelectronic and quantum technologies. However, phenomena such as polariton bottlenecks and quantum blockades, which significantly influence the formation and stability of these quantum states, remain poorly understood, constraining the full realization of their potential.

To address this, we employ ultrafast pump-probe microscopy and time-resolved photoluminescence microscopy to directly image the evolution and interactions of polariton populations in real and momentum space for tungsten- and molybdenum-based systems. Most notably, we observe a clear bottleneck effect—typically detrimental to the formation of zero-momentum condensates that strongly depends on polariton density, cavity detuning, and the relative energetic positions of spin-dark states and phonon modes. Complementing our experimental findings, we perform microscopic theoretical modeling based on the semiclassical Boltzmann equations to examine the complete time- and momentum-resolved exciton-polariton relaxation dynamics [2], incorporating polariton–polariton interactions and nonlinear phenomena.

These results provide critical insights into the mechanisms governing polariton dynamics in TMDC-based systems, paving the way toward improved control and optimization of polaritonic quantum states.







[1] J. Zhao et al., Ultralow threshold polariton condensate in a monolayer semiconductor microcavity at room temperature, Nano Lett. 21, 3331 (2021).

[2] J. M. Fitzgerald et al., Circumventing the polariton bottleneck via dark excitons in 2D semiconductors, Optica 11, 1346 (2024).







Giant Rabi splitting and polariton photoluminescence in an all solution-deposited dielectric microcavity

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Presentation type: Contributed talk

The study of polaritons has attracted significant attention due to their potential applications in optoelectronics, chemistry, and quantum information processing [1]. Recently, polaritons have also been leveraged to enhance the operational lifetime of phosphorescent organic light-emitting diodes (OLEDs), marking a major milestone for their practical implementation [2,3]. However, most polaritonic microcavities rely on vacuum deposition techniques, which are costly, inaccessible to many, and incompatible with large-scale, printed optoelectronics. Solution-processed fabrication of microcavities and distributed Bragg reflectors (DBRs) is therefore critical for achieving scalable, tunable, and environmentally friendly polaritonic devices [4].

Herein, we demonstrate strong light-matter coupling within a fully solution-processed dielectric microcavity. Using an in-house-built automated dip-coating system [5], we fabricated highly reflective DBRs consisting of sub-100 nm films, achieving microcavities with a *Q*-factor exceeding 90. The automated dip-coater enabled precise deposition and annealing in a single processing step, ensuring uniformity and preventing interlayer mixing. This high-quality fabrication was key to observing enhanced reservoir exciton scattering into the lower polariton and studying the effects of strong coupling on bimolecular interactions. The microcavities were designed with Rhodamine 6G in a poly(vinyl alcohol) (PVA) matrix as the active material, deposited via spin-coating with an optimized annealing step to control layer intermixing. Reflectivity measurements revealed Rabi splittings of 130 meV, 264 meV, and 400 meV for microcavities with Rhodamine 6G concentrations of 1 mg/mL, 3 mg/mL, and 5 mg/mL, respectively. Photoluminescence measurements confirmed emission from the lower polariton. Excitation-dependent studies demonstrated enhanced scattering of reservoir excitons into the lower polariton and a more than 10-fold increase in the critical excitation density for bimolecular annihilation compared to bare Rhodamine 6G films. This suppression







of singlet-singlet annihilation could only be partially attributed to the sub-3-fold enhancement in radiative lifetime, emphasizing the critical role of strong coupling in modifying molecular dynamics. Additionally, photoluminescence excitation studies showed uniform emission intensity at low pump fluences, with intensity decreasing at higher concentrations due to aggregation effects.

These results establish solution-processed dielectric microcavities and DBRs as a viable and scalable alternative to metal-clad microcavities, offering superior performance while eliminating the need for physical vapor deposition. Our fabrication method opens new possibilities for polaritonic optoelectronic devices beyond conventional deposition constraints, with potential applications in mitigating efficiency roll-off due to exciton-exciton annihilation in organic photonic devices.



FIG. 1. (a) Illustration of the fully solution processed microcavity structure. (b) Reflectivity, photoluminescence and absorption of the microcavity based on the R6G, clearly showing the upper and lower polariton along with PL emission.

[1] H. Zhao et al., Stable blue phosphorescent organic LEDs that use polariton-enhanced Purcell effects, Nature 626, 300 (2023).

[2] R. Bhuyan, et al., The rise and current status of polaritonic photochemistry and photophysics, Chem. Rev. 123, 10877 (2023).

[3] K. S. Daskalakis et al., Converting an organic light-emitting diode from blue to white with Bragg modes, ACS Photonics 6, 2655 (2019).

[4] A. Strang et al., Simple and versatile platforms for manipulating light with matter: strong lightmatter coupling in fully solution-processed optical microcavities, Adv. Mater. 36, 2212056 (2024).

[5] E. Palo et al., Developing solution-processed distributed Bragg reflectors for microcavity polariton applications, J. Phys. Chem. C 127, 14255 (2023).







Bioinspired free-cavity exciton polaritons with J-aggregates for light harvesting

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Presentation type: Invited talk

Light-harvesting structures in natural photosynthetic organelles, such as those found in purple bacteria, consist of chromophores embedded within densely packed antenna systems, organized into precisely arranged nanostructures. In these systems, the chromophores are often naturally occurring J-aggregates, such as bacteriochlorophylls and carotenoids. Inspired by both the molecular and structural features of these biological systems, we have developed a collection of organic materials and nanostructures based on J-aggregates that exhibit tunable optical properties with strong excitonic resonances spanning from the visible to the infrared range [1].

Inspired by the hierarchical multilayer structures of Begonia pavonina iridoplasts, we fabricated all-organic 2D photonic crystals by embedding J-aggregate mixtures into polymer matrices, forming Bragg-like stacks [2]. These structures combine near-zero-index behavior with biomimetic photonic ordering, resulting in enhanced absorption at selected wavelengths [3]. Interestingly, we observe features such as anticrossing-like behavior that resemble strong coupling signatures. However, these effects arise from the highly dispersive nature of the materials rather than light–matter hybridization, prompting a broader discussion on how to distinguish between dispersive optical responses and genuine strong coupling in organic systems.

In a complementary line of research, we have explored how bioinspired nanoscale architectures can be used to confine and manipulate light at subwavelength scales. Starting from the design of J-aggregate nanorings, inspired by the circular arrangement of chromophores in Rhodospirillum molischianum bacteria, we demonstrated through electromagnetic simulations that these structures act as resonators supporting Local Surface Exciton Resonances [1]. Based on this concept, we have recently developed colloidally synthesized J-aggregate nanoparticles that exhibit clear signatures of strong light–matter coupling. These nanoparticles support free-cavity exciton polaritons, offering a scalable and solution-processable route to organic polaritonic materials, and bridging the gap between biomimetic design and practical implementation.







Altogether, these findings pave the way for bioinspired photonic and metamaterial components operating across the visible to infrared spectrum, all within a fully organic platform. At the same time, they offer a fresh perspective on nanoscale light–matter interactions in photosynthetic systems and open new avenues for the development of bio-inspired organic polaritonic architectures.

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[1] S. T. Holder et al., *Bio-inspired building blocks for all-organic metamaterials from visible to near-infrared*, Nanophotonics 12, 307 (2023).

[2] M. A. Castillo et al., Enhanced light absorption in all-polymer biomimetic photonic structures by near-zero-index organic matter, Adv. Funct. Mater. 32, 2113039 (2022).

[3] M. A. Castillo et. al., *Tunable narrowband excitonic optical Tamm states enabled by a metal-free all-organic structure*, Nanophotonics 11, 4879 (2022).







Wednesday, 18 June







Strong coupling and topological phenomena in plasmonic lattices

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Presentation type: Invited keynote talk

Arrays of plasmonic nanoparticles, so-called plasmonic lattices, when combined with an emitter material (gain medium), provide a versatile platform for studies on light-matter interaction in the nanoscale, including collective coherent phenomena as well as topological photonics. We have experimentally realized a BEC of hybrids of surface plasmons and light in a nanoparticle array, with both at weak coupling and strong coupling (polariton) regime, with unique polarization and coherence properties [1-4]. In the lasing regime, we have observed bound state in continuum (BIC) modes with different topological charges [5,6]. Recently, we studied the quantum geometric tensor in these systems. We experimentally observed non-zero quantum metric and Berry curvature along the diagonals of the Brillouin zone of a square lattice of gold nanoparticles [7]. By a theoretical analysis, we show that the Berry curvature originates solely from non-Hermitian effects [8]. This is the first experimental observation of a non-Hermitian Berry curvature. We have introduced a novel approach to design lasing with exceptionally high topological charges [9]. Finally, we will present new results on the photon statistics of our strongly coupled BEC [10].

 T. K. Hakala et al., Bose-Einstein condensation in a plasmonic lattice, Nat. Phys. 14, 739 (2018).

[2] A. I. Väkeväinen et al., Sub-picosecond thermalization dynamics in condensation of strongly coupled lattice plasmons, Nat. Commun. 11, 3139 (2020).

[3] J. M. Taskinen et al., Polarization and phase textures in lattice plasmon condensates, Nano Lett.21, 5202 (2021).

[4] A. J. Moilanen et al., Spatial and temporal coherence in strongly coupled plasmonic Bose–Einstein condensates Phys. Rev. Lett. 127, 255301 (2021).

[5] R. Heilmann et al., Quasi-BIC mode lasing in a quadrumer plasmonic lattice, ACS Photonics 9, 224 (2022).

[6] G. Salerno et al., Loss-driven topological transitions in lasing, Phys. Rev. Lett. 129, 173901 (2022).







[7] J. Cuerda et al., Observation of quantum metric and non-Hermitian Berry curvature in a plasmonic lattice, Phys. Rev. Res. 6, L022020 (2024).

[8] J. Cuerda et al., *Pseudospin-orbit coupling and non-Hermitian effects in the quantum geometric tensor of a plasmonic lattice* Phys. Rev. B 109, 165439 (2024).

[9] K. Arjas et al., High topological charge lasing in quasicrystals, Nat. Commun. 15, 9544 (2024).

[10] E. A. Mamonov, S. Eyvazi, and P. Törmä, in preparation (2025).







Addressing ultrastrong light–matter interaction of organic molecular layers in optical and infrared cavities

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Presentation type: Invited talk

The excitation of vibrations in single organic molecules and their assemblies can be modified by their coupling to nanoscale visible/infrared resonators. Spectral tailoring of phononic bands and molecular vibrational fingerprints, as well as nonlinear responses derived from the interaction with nanocavities will be reported.

The interaction of vibrations in organic molecules or phonons in solid state layers with optical and infrared resonators can produce a dramatic modification of the energy and decay rates of the states of the coupled system. Nanoscale plasmonic structures, including thin films and nanocavities are particularly suited to enhance the electromagnetic coupling with molecular layers, and even single molecules, located in the proximity. The strength of the coupling depends on the polarization of the molecules, as well as on the local field acting on them. When this coupling strength is larger than the decay rates of the vibrations and that of the electromagnetic resonator, hybridized coupled states of excitation give rise to new polaritonic branches, accessible by infrared spectroscopy.

Direct coupling of vibrations with infrared resonators will be shown to reach the strong and ultrastrong coupling regime of interaction, as in hBN layers filling up an infrared Fabry-Perot cavity [1], where the ultrastrong coupling regime can be relatively easily achieved. Moreover, nanoantennas made of hBN itself can sustain infrared modes which strongly interact with the vibrations of the material, producing polaritonic opening of infrared branches, as directly probed in infrared scattering microscopy [2]. We face now the possibility to tailor infrared polaritonic branches in phononic materials coupled to low carrier concentration plasmonic resonators, opening a new venue to engineer phononic integrated devices. Some examples will be shown.

Furthermore, vibrations in molecules can also be explored and enhanced by inelastic scattering of light in nano- and pico-cavities, in so-called molecular optomechanics [3]. This interaction can produce a non-linear signal of vibrational side bands, as in Surface-Enhanced Raman Spectroscopy







performed in picocavities. The control of the optomechanical interaction by picocavity engineering can lead to reversible vibrational energy shifts and broadenings at the single molecule level. Evidence of this behavior will be shown. Vibrational coupling to nanocavities thus emerge as a powerful alternative paradigm to tailor infrared photonic states.

[1] M. Barra-Burillo et al., *Microcavity phonon polaritons from the weak to the ultrastrong phonon–photon coupling regime*, Nat. Commun. 12, 6206 (2021).

[2] I. Dolado et al., Remote near-field spectroscopy of vibrational strong coupling between organic molecules and phononic nanoresonators, Nat. Commun. 13, 6850 (2022).

[3] F. Benz et al., Single-molecule optomechanics in "picocavities", Science 354, 726 (2016).







Nanophotonics to control mid-infrared light and polariton condensation

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Presentation type: Contributed talk

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Confining light to picoscale volumes within plasmonic nanogaps unlocks new regimes of lightmatter interaction, enabling both strong coupling with single molecules and the emergence of collective optical phenomena [1,2]. When plasmonic nanoparticles self-assemble into extended arrays (Fig. 1(a)), these interactions lead to a previously unobserved form of room-temperature polariton condensation. Each molecular exciton couples tightly to localized plasmonic fields, facilitating longrange interactions across gaps exceeding 100 nm. Under continuous-wave pumping, we observe a threshold in photoluminescence without spectral narrowing, excluding conventional lasing. Instead, emission spreads spatially beyond the pumped region (Fig. 1(b)), indicating a collective mode that is synchronized in space but not in time. Michelson interferometry uncovers long-range coherence, demonstrating a robust emergent condensate (Fig. 1(c)).

The same confinement principles also enable advances in mid-infrared (IR) detection, critical for biomedical diagnostics, environmental sensing, and thermal imaging. By structuring disordered gold nanoparticle layers with precise molecular spacing, we achieve simultaneous mid-IR and visible resonances, allowing ultra-sensitive upconversion of IR to visible light down to the single-molecule level [3]. Ultrafast vibrational spectroscopy confirms that vibrational cascades, central to plasmonic condensation, also drive collective super-radiant states [4] in IR-visible conversion.

Embedding lanthanide-doped nanocrystals within plasmonic nanogaps enables efficient shortwaveinfrared (SWIR) to visible light conversion, enhancing detector performance. Photon upconversion, combining several low-energy photons to generate one high-energy photon is of wide interest for biomedical, catalytic and photonic applications. The excitation wavelength of LnNPs has






FIG. 1. Synchronization of interacting dipoles between plasmonic nanocavities and enhanced short-wave infrared (SWIR) upconversion in nanocavities. (a) Emission from array of nanoparticles pumped by tightlyfocused 633 nm laser exciting core region which feeds surrounding halo. Methylene blue emitter within CB[7] host forms optical dipole (purple) in nanogap between gold nanoparticles. (b) Imaged emission below ($P < P_{th}$) and above ($P > P_{th}$) threshold pump power. (c) Experimental spatio-temporal $g^{(1)}(R, \tau)$ decay for synchronized dipoles. (d) Schematic illustration of the nanoparticle-on-foil nanocavity constructed from gold nanoparticles (AuNP) on top of lanthanide nanoparticle (LnNP) monolayer, and pump power dependence of the SWIR upconversion intensity.

been limited to the second near-infrared window (1000 - 1700 nm). We demonstrate self-sensitized holmium-doped nanoconverters that extend sensitization to $2 \,\mu\text{m}$, achieving up to 15.2% relative upconversion efficiency [5]. Precise dopant control and shelling optimize energy transfer, while integration into plasmonic nanocavities boosts emission intensity up to 32-fold (Fig. 1(d)) by reducing the Ho emission lifetime from 29 μ s to < 1 ns via Purcell enhancement.

These findings bridge fundamental light-matter physics with applied photonics, advancing both the quest for room-temperature polariton condensates and next-generation mid-infrared photodetectors.

[1] R. Arul et al., Giant mid-IR resonant coupling to molecular vibrations in sub-nm gaps of plasmonic multilayer metafilms Light: Sci. Appl. 11, 281 (2022).

[2] S. Hu et al. Robust consistent single quantum dot strong coupling in plasmonic nanocavities, Nat. Commun. 15, 6835 (2024).

[3] R. Chikkaraddy et. al., Single-molecule mid-infrared spectroscopy and detection through vibrationally assisted luminescence, Nat. Photon. 17, 865 (2023).

[4] R. Arul et al., Raman probing the local ultrastrong coupling o vibrational plasmon polaritons on metallic gratings, Phys. Rev. Lett. 13, 126902 (2023).

[5] R. Arul et al., Efficient short-wave infrared upconversion by self-sensitized holmium-doped nanoparticles, arxiv:2411.19949 (2025).







Organic polaritons enable high-responsivity, angle-independent narrowband microcavity photodiodes

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Presentation type: Contributed talk

Plexcitonic nanoparticles exhibiting strong light-matter interactions between plasmonic nanoparticles and J-aggregates, hold promise for various applications, particularly in sensing, which remains largely unexplored. However, their colloidal stability is often compromised by two key factors: (1) the sensitivity of J-aggregates to environmental conditions such as pH, ionic strength, and solvent polarity, and (2) the tendency of J-aggregates to adsorb onto the plasmonic surface, which can destabilize the colloids. To overcome these challenges, we have developed robust excitonic Au@Ag@J-aggregate nanorods encapsulated within a mesoporous silica shell (Au@Ag@Jaggregate@mSiO₂ nanorattles). The light-matter interactions in these hybrids were analyzed using UV-vis and surface-enhanced Raman scattering (SERS) spectroscopy, employing the J-aggregates as Raman reporters. These nanoparticles exhibited high SERS efficiency when excited at both 532 nm and 633 nm laser lines, even achieving single-nanoparticle detection with the 532 nm laser. Finite-difference time-domain (FDTD) calculations further supported these findings, revealing the electric field distribution in the plexcitonic nanoparticle. Near the upper plexciton (532 nm), the electric field is highly confined within the J-aggregate shell due to its near-zero refractive index, maximizing light-matter interactions. At the lower plexciton wavelength (633 nm), the field extends through the J-aggregate/water interface, resembling conventional plasmonic nanoparticles. These results emphasize the significance of optimizing strong coupling effects for efficient SERS, paving the way for ultrasensitive biosensing and bioimaging by fine-tuning the interaction between the lower polariton and the excitation wavelength.









FIG. 1. (A) Experimental extinction spectra of Au@Ag@mSiO₂ nanorattles (black) and plexcitonic nanorattles (red). (B) Transmission Electron Microscopy image of plexcitonic nanorattles. (C-D) Calculated spatial distribution of the electric field enhancement to the fourth power in an X-section crossing at the center of a plexcitonic Au@Ag NR in the XZ plane, for polarizations parallel to the long axis, under 532 nm and 633 nm irradiation.

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 C. Estévez-Varela et al., Plexcitonic nanorattles as highly efficient SERS-encoded tags, Small 20 ,2306045 (2023).







Quantum optics with radiation on the move

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Presentation type: Invited talk

With the scaling of quantum technologies to many separate material quantum components, we may have recourse to couple these systems by quantum radiation of light, microwaves or phonons. In future optical quantum processors, we may, conversely, need to manipulate the quantum states of radiation pulses by their interaction with non-linear stationary quantum components. Several physical processes have been proposed and already demonstrated for these tasks. There are, however, rather fundamental obstacles to the treatment of propagation of radiation in circuits for quantum computing. These obstacles include the general multimode character of propagating fields and the duration and spatial extent of useful light and microwave pulses. The talk will review recent developments of a cascaded master equation approach to deal theoretically with these obstacles, and it will present examples of new, unforeseen, possibilities for easy preparation and manipulation "on the fly" of quantum states of light and matter.

 A. H. Kiilerich and K. Mølmer, Input-output theory with quantum pulses, Phys. Rev. Lett. 123, 123604 (2019).

[2] Y. Fang et al., Deterministic photon sorting in waveguide QED systems, Phys. Rev. Lett. 128, 213603 (2022).

[3] M. M. Lund et al., Subtraction and addition of propagating photons by two-level emitters, Phys. Rev. Lett. 133, 103601 (2024).







Ultrafast spectroscopy reveals (dis)order-driven polariton dynamics

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Presentation type: Contributed talk

We present our latest work on a fundamental quantum optical theory of strongly coupled plasmonic systems. These systems are inherently non-Markovian, exhibiting a dynamic exchange of energy and information between molecules, plasmons, and the electromagnetic field, while simultaneously being subject to significant loss and decoherence through radiation, dissipation, and multi-mode coupling. A conventional open quantum system approach (Fig. 1a) fails to fully capture all of these effects in such low Q environments [1], leading to inaccurate descriptions of energy transfer and coherence. Our goal is to develop a framework that captures all the non-Markovian dynamics in plasmonic systems strongly coupled to organic molecules (Fig. 1b), and demonstrate the implications for quantum information processing, metrology, and photochemistry in plasmonic environments. Furthermore, we highlight how such an understanding of strongly coupled plasmonic systems can change our perspective on the nature of quantum information flow in other open quantum systems.



FIG. 1. Open quantum system (a) versus pseudomode description (b) of plasmonic systems strongly coupled to a quantum (dipole) emitter. The pseudomode description retains all the information about the dynamical evolution of the system.

Traditionally, one describes non-Markovian dynamics by an open quantum system with an irreversible interaction with a memoryless environment (reservoir), into which information is lost. This paradigm is well suited to leaky yet high-finesse, macroscopic cavities where the system-reservoir







boundary is clearly defined by a reflective surface. However, in plasmonic systems no such clear physical boundary exists. Instead, the system and environment are completely defined by the approximations made on the spectral integral that arises from the quantum equations of motion of the full system (e.g. [2]);

$$X_{\rm sys}(t) = -\int_0^t \mathrm{d}\tau \int_0^\infty \mathrm{d}\omega \, D(\omega, \mathbf{r}) \, e^{i(\omega_{\rm X} - \omega)(t - \tau)} X_{\rm sys}(t). \tag{1}$$

Here, is the local density of states, and $X_{\text{sys}}(t)$ could be any system variable of interest such as the state of molecular excitation, or its transition moment. For example, the Born–Markov approximation assumes $D(\omega, r)$ is sufficiently smooth, Wigner–Weiskopff decay assumes it has constant value, while a Lorentzian approximation of $D(\omega)$ is accurate when the system is a series of high-finesse resonances [1].

Surprisingly, we find that the finite geometry of realistic plasmonic and nanophotonic environments gives rise to an exact solution of the spectral integral in equation 1, despite the complexity it brings to the spectral density. We adopt this approach to provide an invertible transformation from the continuous set of true modes of the system to a discrete set of pseudomodes, for which the equations of motion can be solved exactly [3]. We illustrate this for the case of a quantum (dipole) emitter strongly coupled to a nano-sphere, the simplest structure that has finite bounds in all three spatial dimensions. Our treatment captures all dynamics of the system whether Markovian or not, whilst the invertible nature of our pseudomode transformation allows us to reconstruct the full evolution of the quantized electromagnetic field in both the spatial and temporal domains. By examining our exact solutions, we are able to precisely explain the roles of mode regularisation and its link to energy propagation, renormalisation, and the Lamb shift in nanophotonic systems. Finally, we give an overview of our progress to incorporate dispersion and its associated loss into our formalism, and the utility of our method applied to the Heisenberg picture, where a plethora of quantum optical phenomena, such as multi-partite entanglement, dressed states, photon pair production, and squeezed light generation can be readily adopted into the plasmonic domain.

 A. Saharyan et al., Light-matter interaction in open cavities with dielectric stacks, Appl. Phys. Lett. 118, 154002 (2021).

[2] S. M. Barnett and P. M. Radmore, Methods in Theoretical Quantum Optics (Oxford University Press, 2002).

[3] B. Yuen and A. Demetriadou, *Exact quantum electrodynamics of radiative photonic environ*ments, Phys. Rev. Lett. 133, 203604 (2024).







Chiral strong coupling quantum electrodynamics Hartree–Fock

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Presentation type: Contributed talk

Light-matter strong coupling offer an invaluable route for engineering molecular properties by finetuning the electromagnetic fields [1]. Coupling chiral molecules with circularly polarized electromagnetic vacuum fluctuations results in a unique chiral interaction, which offers a viable way for enantiomeric discrimination (see Fig. 1) and enantioselective synthetic pathways [2]. In this talk, I will present the theoretical development of a method able to capture the enantiospecific power of chiral light. The method extends the strong coupling quantum electrodynamics Hartree–Fock method to a chiral discriminating light-matter Hamiltonian [3,4]. To achieve this, the shape of the chiral field is incorporated beyond the dipole approximation by employing the velocity gauge



FIG. 1. Left and right-handed chiral cavities coupled with pure enantiomers of a proline molecule.







and a basis optimization. Furthermore, the use of an orbital specific coherent-state transformation enables to capture electron-photon correlation already at the mean-field level and permit to obtain consistent set molecular orbitals and orbital energies [5,6].

 J. A. Hutchison et al., Modifying chemical landscapes by coupling to vacuum fields, Angew. Chem. Int. Ed. 51, 1592 (2012).

[2] D. G. Baranov et al., Toward molecular chiral polaritons, ACS Photonics 10, 2440 (2023).

[3] R. R. Riso et al., Strong coupling in chiral cavities: nonperturbative framework for enantiomer discrimination, Phys. Rev. X 13, 031002 (2023).

[4] R. R. Riso et al., Chiral polaritonics: cavity-mediated enantioselective excitation condensation, Rep. Prog. Phys. 88, 027901 (2025).

[5] R. R. Riso et al., Molecular orbital theory in cavity QED environments, Nat. Commun. 13, 1368 (2022).

[6] Y. El Moutaoukal et al., Toward polaritonic molecular orbitals for large molecular systems, J. Chem. Theory Comput. 20, 8911 (2024).







Ultrafast imaging of nonlinear exciton polariton interactions

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Presentation type: Invited talk

Excitons—bound electron-hole pairs—are the electronic excited states in many organic and inorganic semiconductors, particularly in low-dimensional materials. Semiconductor microcavities operating in the strong coupling regime, which host exciton-polaritons, have emerged as promising systems for enhancing photon nonlinearities. However, exciton-polaritons have yet to reach the regime of deeply strong interactions at the single-photon or few-photon level.

To address this challenge, I will present our recent efforts to enhance these interactions by forming exciton Fermi polarons in single-layer transition metal dichalcogenides (TMDs). Furthermore, I will discuss results from integrating two-dimensional semiconductors with plasmonic lattices, highlighting their potential for achieving stronger light–matter interactions







Nonlinear polaritons in van der Waals heterostructure metasurfaces

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Presentation type: Contributed talk

Integrating metasurfaces with two-dimensional (2D) materials hold significant promise for creating ultrathin photonic devices, yet merging these two realms remains challenging. Here, we demonstrate van der Waals (vdW) heterostructure metasurfaces, where 2D material stacks are directly embedded with nanophotonic functionalities in a monolithic platform. Leveraging symmetry-protected quasi bound states in the continuum (qBIC) resonances, engineered in hexagonal boron nitride (hBN) thin films, we achieve high quality (Q) factor optical resonances enabling strong light confinement [1]. We then fabricate vdW metasurfaces directly from hBN-encapsulated WS₂ monolayers via common top-down fabrication techniques [2]. Room-temperature strong coupling between qBIC modes and WS₂ excitons is confirmed from both absorption and emission properties of the vdW metasurfaces. Moreover, back focal plane spectroscopy reveals directional polariton dynamics governed by qBIC resonances, while ultralow excitation fluences (< 1 nJ/cm^2) induce significant exciton-polariton nonlinearities, outperforming conventional approaches of about 3 orders of magnitude. This approach overcomes the need for external optical cavities, opening new avenues for ambient operation of low-power polaritonic devices, towards low-threshold lasers, optical switches, and polaritonic condensates.

[1] L. Kühner et al., *High-Q nanophotonics over the full visible spectrum enabled by hexagonal boron nitride metasurfaces*, Adv. Mater. 35, 2209688 (2023).

[2] L. Sortino et al., Van der Waals heterostructure metasurfaces: atomic-layer assembly of ultrathin optical cavities, arXiv:2407.16480 (2024).







Quantum dynamics in coupled plasmonic cavities

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Presentation type: Contributed talk

Plasmonic nanocavities facilitate strong light-matter interactions due to their ability to confine electromagnetic fields into subwavelength volumes. This field confinement arises from the evanescent field of localised surface plasmon polaritons (LSPs) formed on the surface of the metal and has led to several advances in spectroscopy, non-linear optics and metamaterials [1]. In particular, the nanoparticle-on-mirror (NPoM) cavity formed by placing a nanoparticle above a metallic film separated by a dielectric spacer has demonstrated strong coupling with a single molecule even at room temperatures [2]. Such nanocavities hosting multiple quantum emitters (QEs) are also promising candidates to generate subradiant entangled states at ambient conditions. However, there are several challenges to this: (i) plasmonic nanocavities have extremely large losses due to Ohmic losses in the metal and (ii) it is experimentally difficult to initialize the system with only one QE in an excited state. The latter is a key requirement for generating subradiant entanglement and is a challenge due to the close proximity of the QEs which are usually separated by just a few nanometers [3].

In this work, we design a system composed of two coupled NPoM cavities, each hosting a single QE (Fig. 1a). The cavities are coupled through propagating surface plasmon polaritons (SPPs) launched by each nanoparticle during excitation and since the SPPs have a propagation length of several micrometers, the cavities can be placed far away from each other for selective excitation. In addition, the SPPs launched by each nanocavity interfere either constructively or destructively with each other, depending on the separation between the individual nanocavities (Fig. 1b), which allows the coupling to be switched on and off. To determine the quantum dynamics, we use an open quantum system formalism to accurately account for the various loss channels present in the system. Here, the density matrix ρ is described by a Lindblad master equation [4]:

$$\frac{d\rho(t)}{dt} = -i[\mathcal{H}_{sys},\rho] + \sum_{\xi} \Gamma^{\xi}_{out}(2a_{\xi}\rho a^{\dagger}_{\xi} - a^{\dagger}_{\xi}a_{\xi}\rho - \rho a^{\dagger}_{\xi}a_{\xi}) + \sum_{i} \Gamma_{vib}(\sigma^{(i)}_{z}\rho\sigma^{(i)}_{z} - \rho)$$
(1)







dephasing of QEs



$$\mathcal{H}_{sys} = \underbrace{\sum_{\xi} \omega_{\xi} a_{\xi}^{\dagger} a_{\xi}}_{\text{plasmonic modes}} + \underbrace{\sum_{i=1,2} \frac{1}{2} \omega_d^{(i)} \sigma_z^{(i)}}_{\text{QEs}} + \underbrace{\sum_{\xi,i} g_{\xi}(r^{(i)}) (a_{\xi}^{\dagger} \sigma_{-}^{(i)} + a_{\xi} \sigma_{+}^{(i)})}_{\text{modes-QE interaction}}$$

We show how the different SPP interaction regimes, depending on whether the second nanocavity is positioned at a maxima or minima, affect the quantum dynamics of the system. In particular, the eigenmodes of \mathcal{H}_{sys} also reveal an indirect interaction between the close-to-degenerate bonding and antibonding plasmonic modes because of their large coupling strengths and close spectral positions, which can be tuned with the separation of the nanocavities and the dipole moment of the QEs [4]. Considering these factors, we also use tailored broadband and monochromatic pulses to engineer quantum states in the plasmonic system. The ability to separately excite each NPoM cavity presents a pathway toward the generation of subradiant entanglement in plasmonics, which is useful for future light based quantum technologies.



FIG. 1. (a) Two NPoMs with a QE in each nanocavity. (b) Frequency difference between the bonding and anti-bonding modes and the electric field overlap between propagating SPPs as a function of cavity separation. (c) Quantum dynamics at 960 nm separation.

 J. J. Baumberg et. al., Extreme nanophotonics from ultrathin metallic gaps, Nat. Mater. 18, 668 (2019).

[2] R. Chikkaraddy et al., Single-molecule strong coupling at room temperature in plasmonic nanocavities, Nature 535, 127 (2016).

[3] K. Bedingfield et. al., Subradiant entanglement in plasmonic nanocavities, Phys. Rev. B 111, 075420 (2025).

[4] A. Crookes et. al., Collective multimode strong coupling in plasmonic nanocavities, arXiv:2411.07694 (2024).







Ultrafast collapse of molecular polaritons

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The coupling between plasmonic excitations and electronic transitions in molecules, known as molecular polaritons, is gaining a lot of interest from both the condensed matter physics and physical chemistry communities as it can radically affect the molecular electronic structure, thus resulting in modified energy landscape and electron-transfer pathways [1,2]. In this talk, I will present recent studies on the interaction of photochromic spiropyran(SP)-merocyanine(MC) molecules with aluminum plasmonic nanostructures which display two spectrally separated localized surface plasmon resonances (LSPRs). These nanostructures are designed such that the MC molecular absorption is resonant with the long axis LSPR, whereas it is detuned from the short axis LSPR, giving rise to strong and weak coupling regimes, respectively. Pump-probe spectroscopy measurements reveal a sub-ps collapse of polaritons to pure molecular transition triggered by femtosecond-pulse excitation. By supporting our findings with a quantum mechanical model based on the extension of the Tavis–Cummings Hamiltonian [3], we show that the response of the system on the ps timescale is governed by intramolecular dynamics, occurring one order of magnitude faster with respect to the uncoupled excited molecule relaxation to the ground state in both strong and weak coupling regimes [4].

[1] P. Vasa et al., Real-time observation of ultrafast Rabi oscillations between excitons and plasmons in metal nanostructures with J-aggregates, Nat. Photon. 7, 128 (2013).

[2] A. N. Koya et al., Advances in ultrafast plasmonics, Appl. Phys. Rev. 10, 021318 (2023).

 [3] M. Tavis and F. W. Cummings, Exact solution for an N-molecule—radiation-field Hamiltonian, Phys. Rev. 170, 379 (1968).

[4] J. Kuttruff et al., Sub-picosecond collapse of molecular polaritons to pure molecular transition in plasmonic photoswitch-nanoantennas, Nat. Commun. 14, 3875 (2023).







Ultrafast spectroscopy reveals (dis)order-driven polariton dynamics

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Presentation type: Contributed talk

Many pioneering works seeking to understand exciton-polaritons have been conducted on inorganic semiconductors and organic materials like J aggregates which possess narrow linewidths and minimal disorder. However, in recent years, there has been a notable shift in focus towards exploring the role of exciton-polaritons for triplet harvesting, charge transfer processes in photovoltaic devices, energy transfer dynamics, condensation, and nonlinear optics using materials that possess substantial disorder. Despite the fact that most important polariton phenomena are studied using disordered materials, the explicit role of disorder and its characterization have often been overlooked. Recent studies, including our own, emphasized that disorder can result in the mixing of bright and dark states in the polariton manifold leading to the formation of "gray states" that possess non-zero photon character [1,2,3]. The emergence of these gray states could have profound implications for cavity dynamics by introducing entirely new photophysical pathways [2]. Specifically, the presence of a significantly larger number of states with mixed photonic character would create additional channels for energy redistribution, potentially extending the lifetime of polariton states by enabling energy to dissipate more gradually through the system.

Building on our previous work that examined disorder from a static perspective, we now turn to ultrafast time resolved spectroscopy to examine the impact of disorder on real time polariton relaxation. We investigate systems with varying degrees of electronic disorder evident in their steady state absorption spectra with < 10 fs time resolution. Ultimately, we find a slower relaxation time in more disordered systems suggesting that increased disorder impedes the relaxation pathway from the upper polariton to the lower polariton state. Given that disorder can broaden the density of states and facilitate alternative relaxation routes, these findings support the notion that gray states play a significant role in modulating polariton relaxation dynamics. This insight highlights disorder as a potential design parameter for tuning organic polariton photophysics. By strategically introducing disorder, it may be possible to extend polariton lifetimes, which could be advantageous for applications that rely on long-lived polaritonic states, such as quantum optics or polaritonic









FIG. 1. Disorder effects in polariton relaxation: ultrafast transient absorption studies.

A. George et al., Controlling the manifold of polariton states through molecular disorder, Adv.
Opt. Mater. 12, 2302387 (2024).

[2] M. Son et al., Energy cascades in donor-acceptor exciton-polaritons observed by ultrafast twodimensional white-light spectroscopy, Nat. Commun. 13, 7305 (2022).

[3] T. Gera and K. L. Sebastian, *Exact results for the Tavis–Cummings and Hückel Hamiltonians with diagonal disorder*, J. Phys. Chem. A 126, 5449 (2022).





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Sub-5-fs pump-probe spectroscopy on strongly coupled exciton–cavity-polaritons

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Presentation type: Contributed talk

The dynamics of strongly coupled nanosystems have been intensively studied [1-3], but investigations in the few-fs regime are so far not standard practice. However, many of the systems have dynamics calling for sub-10-fs resolution [4]. We investigate the ultrafast dynamics of strongly coupled exciton–cavity polaritons using sub-5-fs visible light pulses at ambient conditions. The expe-



FIG. 1. Transient reflection map of our strongly coupled system with a pump fluence of $1.82 \, mJ/cm^2$.

riments employ a 11.5 nm thick WS_2 multilayer embedded in a Fabry-Pérot microcavity on a silicon substrate. Degenerate pump-probe spectroscopy was used in reflection, incorporating a selfreferenced probe to minimize shot-to-shot noise. The measurements reveal transient reflectivity SDU \bigstar





dynamics on the 10-fs timescale, inaccessible to conventional Ti:Sapphire systems with pulse durations exceeding 30 fs. This unprecedented time resolution provides novel insights (Fig. 1) into the polariton formation process, coupling dynamics and lifetime in these systems. Power dependent investigations reveal the saturation behavior of the polaritons. These findings offer new perspectives on the early-stage evolution of exciton-polariton states in microcavities.

[1] Y. Tang et al., Ultrafast response of a hybrid device based on strongly coupled monolayer WS_2 and photonic crystals: the effect of photoinduced Coulombic screening, Laser Photon. Rev. 14, 1900419 (2020).

[2] W. Du et al., Ultrafast modulation of exciton-plasmon coupling in a monolayer WS_2 -Ag nanodisk hybrid system, ACS Photonics 6, 2832 (2019).

[3] P. Vasa et al., Real-time observation of ultrafast Rabi oscillations between excitons and plasmons in metal nanostructures with J-aggregates, Nat. Photon. 7, 128 (2013).

[4] A. Bisht et al., Collective strong light-matter coupling in hierarchical microcavity-plasmon-exciton systems, Nano Lett. 19, 189 (2019).







Thursday, 19 June







SMILES – a plug and play platform for molecular polariton condensates

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Presentation type: Invited talk

Frenkel excitons in molecular systems facilitate the observation of strong exciton-photon coupling and non-equilibrium Bose Einstein like condensation at room temperature. However, most work in the context of condensation has been carried out using specific molecular systems. Here we report a plug-and-play platform for realizing polariton condensation using most commercially available fluorescent dyes [1, 2]. This is made possible using the "Small Molecular Ionic Isolation Lattice (SMILES)." [3] Realization of lattices of these condensates using lithography and structured illumination will also be presented [4]. Finally, we will discuss the role of polariton condensation on energy transfer dynamics between donor-acceptor systems.

[1] P. Deshmukh et al., *Plug-and-play molecular approach for room temperature polariton conden*sation, ACS Photonics 11, 348 (2024).

[2] D. Choi et al., *Highly coherent room-temperature molecular polariton condensates*, Adv. Opt. Mat 13, 2500086 (2025).

[3] C. R. Benson et al., *Plug-and=play optical materials from fluorescent dyes and macrocycles* Chem 6, 1978 (2020).

[4] R. K. Yadav et al., Direct writing of room temperature polariton condensate lattice, Nano Lett.24, 4945 (2024).







Condensation of cavity exciton-polaritons in perovskite nanocrystals at room-temperature

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Presentation type: Contributed talk

Strong light-matter coupling and exciton-polariton condensates offer a promising approach to introduce robust interactions and nonlinear effects into a wide array of photonic technologies, spanning from high-speed all-optical logic to low-threshold topological lasers. By integrating colloidal semiconductor quantum dots—notable for their pronounced three-dimensional confinement and excellent optical characteristics—as active medium in microcavities, it is possible to boost polaritonic interactions through quantum confinement. Nonetheless, the realization of exciton-polariton condensation in microcavities at room temperature has remained elusive for both epitaxial and colloidal quantum dots.

In this work [1], we demonstrate room-temperature polariton condensation by embedding a thin film of monodisperse colloidal CsPbBr₃ quantum dots within a tunable optical resonator. This resonator incorporates a Gaussian-shaped deformation creating a wavelength-scale potential well for polaritons. Under pulsed optical excitation, we demonstrated the emergence of polariton condensation manifested by a superlinear increase in emission intensity, a narrowing of the emission linewidth, a blueshift (Fig. 1a), and an extension of temporal coherence (Fig. 1b). Our results highlight the potential of perovskite-based colloidal quantum dots, celebrated for their remarkable optical properties and high tunability, as a cutting-edge platform for next-generation polaritonic









FIG. 1. (a) Measurement of the condensation threshold, illustrating how emission intensity (top), emission linewidth (middle), and blueshift (bottom) evolve as excitation fluence increases. The uncondensed groundstate polariton emission is marked by purple circles, the ground-state condensate by orange squares, and the non-condensing first-excited state by gray diamonds. (b) Real-space interferograms of the emitted signal below (top panels) and above (bottom panels) threshold are shown. At $\Delta t = 0$ ps, interference fringes appear in both regimes. Below threshold, these fringes diminish within about 0.1 ps, whereas above threshold, they persist for as long as 2.8 ps.

[1] I. Georgakilas et al., Room-temperature cavity exciton-polariton condensation in perovskite quantum dots, arXiv:2408.10667 (2024).







Room-temperature polaritons in a tunable open microcavity using 2D hybrid perovskites

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Presentation type: Contributed talk

Cavity quantum electrodynamics (cavity QED) refers to the study of the interaction between light and quantum emitters in an optical cavity. The simplest form of an optical cavity is the Fabry-Perot (FP) resonator, which can be used for harnessing strong light matter interactions [1]. Hybrid perovskites have emerged as an interesting class of 2D semiconductors in the recent years. Unlike conventional 2D materials like transition metal dichalcogenides, perovskites can be easily synthesised in large quantities. Their unique naturally existing quantum well structures confines the excitons in two dimensional plane. This gives rise to large excitonic binding energies at room temperature [2].

Previous works have demonstrated strong coupling in 2D hybrid perovskites in a FP cavity where both the reflecting mirrors are planar [3]. At present, we are working with a tunable microcavity that has planar-concave mirrors, since this allows better confinement of light [4]. The open-cavity system that we have fabricated allows us to tune the photonic modes to improve the coupling strength and can also incorporate various samples. We have synthesised perovskite structures that have coupled strongly with photons in a planar-planar cavity. Due to the small mode volume in a planar-concave cavity, the coupling strength is enhanced, while the lateral confinement of the mode allows quantisation of the polariton mode. This platform will be further developed to obtain condensates which will be studied to build next generation nonlinear nanophotonic devices.

 J. Bellessa et al., Materials for excitons-polaritons: exploiting the diversity of semiconductors, MRS Bull. 49, 932 (2008).

[2] L. Yang et al., Manipulation of the structure and optoelectronic properties through bromine inclusion in a layered lead bromide perovskite, Chem. Mater. 35, 3801 (2023).

[3] J. Wang et al., Room temperature coherently coupled exciton-polaritons in two-dimensional organic-inorganic perovskite, ACS Nano 12, 8382 (2018).

[4] D. Hunger et al., A fiber Fabry-Perot cavity with high finesse, New J. Phys. 12, 065038 (2010).







Polariton waveguides for nonlinear optical phenomena

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Presentation type: Invited talk

Polaritons are quasiparticles emerging from the hybridization of confined photons and excitons in semiconductor structures which have shown a plethora of interesting macroscopic quantum phenomena, including superfluidity, quantized circulation, quantum turbulence, and topological effects [1-3].

By structuring polariton waveguides into photonic crystals, we have engineered their dispersion and demonstrated polariton condensation into topologically protected states with significantly reduced dissipation [4].

More recently we demonstrated coherent quantum fluid molecules formed by polariton condensates in bound states in the continuum (BIC) exploiting the effective attractive feature of negative mass dispersion. These artificial polaritonic molecules exhibit coherent hybridization, nontrivial topological characteristics, and can be scaled to form extended mono- and diatomic chains featuring tunable non-Hermitian band structures [4, 5]. Furthermore, we explored the possibility of engineering artificial gauge fields in these waveguides by spatially modulating the grating filling factor, effectively generating a synthetic electric field.

Finally, we discuss the recent experimental realization of emergent supersolidity in multimode polariton waveguides, an exotic quantum phase characterized by the rupture of two independent symmetries, the U(1) typical of condensation and the translational symmetry, achieved via parametric processes. This phenomenon opens promising avenues for exploring novel quantum phases and complex collective behaviors in driven-dissipative systems [6].

 D. Caputo et al., Josephson vortices induced by phase twisting a polariton superfluid, Nat. Phot. 13, 488 (2019).

 [2] R. Panico et al., Onset of vortex clustering and inverse energy cascade in dissipative quantum fluids, Nat. Phot. 17, 451 (2023).

[3] A. Gianfrate et al., Measurement of the quantum geometric tensor and of the anomalous Hall drift, Nature 578, 381 (2020).







[4] V. Ardizzone et al., Polariton Bose–Einstein condensate from a bound state in the continuum, Nature 605, 447 (2022).

[5] A. Gianfrate et al., *Reconfigurable quantum fluid molecules of bound states in the continuum*, Nat. Phys. 20, 61 (2024).

[6] D. Trypogeorgos et al., *Emerging supersolidity in photonic-crystal polariton condensates*, Nature 639, 337 (2025).







Driven-dissipative superfluids: a compact Kardar–Parisi–Zhang dynamics of the phase

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Presentation type: Invited talk

Abstract to be uploaded soon







On-chip ultrafast polariton transistors

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Presentation type: Contributed talk

The speed of electronic circuits has plateaued due to the limitations of transistor scaling, prompting research into all-optical logic as an alternative for high-speed computing [1]. In this direction, recent developments utilizing the strong light-matter interaction regime offered by exciton-polariton microcavities, have led to the realization of transistors at cryogenic temperature [2]. Furthermore, ultrafast transistor action [3] and NOR gate functionalities [4] have been demonstrated at room-temperature with organic semiconductors embedded in DBR vertical cavities, by exploiting polariton condensation through bosonic stimulation. Nonetheless, this vertical geometry, which has the wavevector of the polariton condensate mainly perpendicular to the substrate, necessitates the use of external free-space optics to re-route the emitted light between the individual transistors. This results in propagation delays on the order of hundreds of picoseconds that ultimately prevent the realization of scalable ultrafast circuits.

This work introduces a scalable approach to room-temperature transistor functionalities in a planar architecture by employing integrated high-contrast grating (HCG) microcavities filled with an organic polymer (MeLPPP) [5]. Such a system exhibits strong light-matter coupling, confirmed by polariton condensation, characterized by a nonlinear emission increase, spectral narrowing, and blue shift (Fig. 1(a)). With one HCG cavity generating the pulsed in-plane control signal that constitutes the input "seed" for a second "transistor" cavity (Fig. 1(b)), we demonstrate transistor action on a picosecond time scale (Fig. 1(c)) by precisely defining the excitation scheme through a fine control of the time delay between the excitation pulses of the two cavities [6]. Hence, we observe up to 60x amplification with minimal signal distortion and a switching contrast exceeding 8:1. By leveraging rubust nanofabrication techniques from silicon photonics, this scalable architecture opens new possibilities for integrated all-optical logic circuits thanks to the planar on-chip nature of the signal routing.

[1] D. A. B. Miller, Are optical transistors the logical next step?, Nat. Photon. 4, 3 (2010). [2] D. Ballarini et al., All-optical polariton transistor, Nat. Commun. 4, 1778 (2013). SDU 🏠 OLIMA UNIVERSITY OF SOUTHERN DENMARK





FIG. 1. (a) The top panel shows the emission as a function of excitation fluence. This light-in light-out characteristic exhibits a nonlinear increase above the threshold ($P_{th} = 39 \,\mu \text{J cm}^{-2}$, dashed line). The top left inset shows the emission spectra below threshold (gray data points) and above threshold (black data points), and their fitted spectra (green and orange, respectively). The right insets show the real-space emission images of the cavity when pumped below and above threshold, corresponding to the fluences with the green and orange data points. The bottom panel displays the peak width (blue triangles) and center energy of the emission peak (red squares) as a function of excitation fluence, showing a sudden narrowing at the threshold and a continuous blue-shift above. (b) Scanning electron microscopy image of the seed and transistor cavities without polymer layer deposited with arrows illustrating the excitation beams and the output of the seed cavity flowing to the transistor cavity. (c) Real-space images of the emission with excitation of the transistor cavity at $0.8P_{th}$. In the top green panel, the seed cavity is not excited. In the middle blue panel, it is excited with $1.2P_{th}$ at a time delay of +1 ps after the excitation of the transistor cavity.

[3] A. V. Zasedatelev et al., A room-temperature organic polariton transistor, Nat. Photon. 13, 378 (2019).

[4] D. A. Sannikov et al., Room temperature, cascadable, all-optical polariton universal gates, Nat.
Commun. 15, 5362 (2024).

[5] T. Stöferle et al., Ultracompact silicon/polymer laser with an absorption-insensitive nanophotonic resonator, Nano Lett. 10, 3675 (2010).

[6] P. Tassan et al., Integrated ultrafast all-optical polariton transistors, arXiv:2404.01868 (2024).





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Polariton-assisted hopping in amorphous non-conducting polymers

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Presentation type: Contributed talk

Over the past decade there has been much interest in using the strong coupling regime of cavity quantum electrodynamics as a tool to modify fundamental properties of matter such as the charge transport mechanisms. Charge transport in materials such as polar crystals or polymers crucially depends on the coupling of carriers to vibrational degrees of freedom, which leads to the formation of dressed carriers called polarons. When those vibrations are resonantly coupled to the modes of



FIG. 1. (a) Current-voltage characteristics under vibrational strong coupling of the out-of-plane bending mode of polystyrene to the second-order mode of a metallic cavity at resonance (red), for a detuned cavity (purple), and without cavity (black). (b) Theoretically computed conductance enhancement as a function of the cavity length L for different phonon-photon coupling strengths (blue: $\Omega_0 = 15.7 \text{ cm}^{-1}$, red: $\Omega_0 =$ 31.4 cm^{-1} , black: $\Omega_0 = 47.1 \text{ cm}^{-1}$). Inset: Conductance enhancement for a resonant cavity with length $L = 4.5 \,\mu\text{m}$ as a function of the coupling strength Ω_0 .





a cavity resonator in the infrared frequency range, an interesting question is whether the emergence of vibronic polaritons can modify the polaron properties and thus affect charge transport in the material. Recent results in amorphous non-conducting polymers embedded in a cavity suggest that vibrational strong coupling can indeed lead to a spectacular enhancement of conductance [1] (Fig. 1a). Here we briefly present those experiments and discuss the theoretical approach that was used to describe the results.

[1] Sunil Kumar et al., Extraordinary electrical conductance through amorphous nonconducting polymers under vibrational strong coupling, J. Am. Chem. Soc. 146, 18999 (2024).





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Triplet population via polaritons in an organic microcavity quantum battery

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Presentation type: Contributed talk

Quantum batteries—quantum systems for energy storage—have gained interest due to their potential scalable charging power density. A quantum battery based on the Dicke model has been explored previoulsy using strongly coupled organic microcavities, which enable a cavity-enhanced energy transfer process called superabsorption [1]. However, energy storage lifetime in these devices is limited by fast radiative emission losses, worsened by superradiance.



FIG. 1. (a) We consider a microcavity with a donor and acceptor layer, separated by an inert polymer spacer to avoid direct dipole-dipole couplings between them. The donor is chosen to be a bright, strongly absorbing dye, while the acceptor is chosen to absorb weakly and to have a dark molecular triplet state. (b) Annotated representative absorption dispersion of a device, clearly demonstrating strong coupling. (c) By transferring energy to the dark molecular triplet states of the acceptor layer we aim to prevent the detrimental effects of superradiance, which leads to short self-discharge time due to cavity-enhanced radiative emission losses (γ_r) from the bright donor singlets, usually in the pico to nanosecond timescale. Instead, the triplet non-radiative relaxation (γ_{nr}) can be orders of magnitude slower, leading to microsecond self-discharge times.

Here, we demonstrate a promising approach to extend the energy storage lifetime of the microcavity quantum battery device using molecular triplet states. We examine a type of multi-layer





microcavity where a strongly coupled active absorption layer transfers energy to the molecular triplets of a storage layer, identifying two regimes based on exciton-polariton resonances. We tested one of these mechanisms by fabricating and characterising five devices detuned across an exciton-polariton resonance, extending the storage time in this class of device by three orders of magnitude.

 J. Q. Quach et al., Superabsorption in an organic microcavity: toward a quantum battery, Sci. Adv. 8, eabk3160 (2022).







Device physics of organic solar cells under strong light-matter coupling

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Presentation type: Contributed talk

Light can strongly interact with molecules to create polaritons, significantly altering the excited energy states. This opens up new possibilities for developing light-based technologies, enhancing energy transfer over large distances, and influencing the formation and breaking of chemical bonds [1]. In this talk, I will discuss our recent work focused on investigating the impact of strong light-matter coupling on the physics of organic solar cells.



FIG. 1. Device physics of (a) reference and (b) cavity organic solar cell under strong light–matter coupling regime.

We show that exciton-polaritons can reliably exist in solution-processed bulk-heterojunction OSCs and can influence their device properties. Combining transient absorption measurements for thin films and transient photocurrent/photovoltage decay measurements for devices, we investigated the photo- and device physics of PTB7:PC71BM cavity OSCs. We find two-fold benefits of strong coupling on OSC device physics, as depicted in Fig. 1. Firstly, we observed that charge photogeneration is enhanced compared to the reference OSC devices. Secondly, we found that the energy of the charge transfer (CT) states is blue-shifted, likely allowing access to higher-energy CT states. This is accompanied by the longer charge carrier lifetime observed in the transient photo-voltage decay measurements that we infer because of reduced bimolecular recombination [2].







This study underscores the significant impact of strong light–matter coupling on modifying the device physics in OSCs, paving the way for engineering more efficient OSCs.

[1] D. Tibben et al., Molecular energy transfer under the strong light-matter interaction regime, Chem. Rev. 123, 8044 (2023).

[2] Y. Tang et al, Strong light-matter coupling leads to a longer charge carrier lifetime in cavity organic solar cells, ACS Photonics 11, 1627 (2024).







Phase topology and group index of THz vibro-polaritons in metasurfaces

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Presentation type: Invited talk

Near-field microscopy is a powerful technique for investigating optical modes and the interaction of electromagnetic waves with matter at subwavelength scales. Conventional near-field microscopes typically detect or excite the near field while operating (exciting or detecting) from the far field. We present a double-probe near-field microscope that both excites and detects in the near field, enabling fully local measurements.

The system employs THz micro-photoconductive antennas to generate ultrashort (single-cycle) THz pulses and detect the transient electric field in the time domain. Fourier transformation of these transients yields the complex near field, including amplitude and phase. By precisely controlling the separation between the antennas, we directly access key photonic properties such as the partial local density of states (PLDOS), spatial coherence, and energy transfer.

This approach is particularly suited for probing dark optical modes, such as bound states in the continuum (BICs), that are inaccessible from the far field. The suppressed radiative losses of these modes enhance light-matter interactions and support the strong coupling regime. In this presentation, we will demonstrate near-field mapping and PLDOS measurements of metasurfaces, reveal the mechanism behind the complete far-field suppression in BICs, and show the extreme electromagnetic field confinement of these modes at the surface. This field confinement facilitates strong coupling with molecular vibrations in lactose thin films, leading to the formation of vibropolaritons.







Metasurface-enhanced superradiance in strongly-coupled J-aggregates

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Presentation type: Contributed talk

The phenomenon of superradiance describes the collective and coherent behaviour of an ensemble of N emitters that spontaneously evolve into a macroscopic superradiant state where the emission intensity scales as N^2 and the emission rate as N [1]. J-aggregates, consisting of highly ordered self-assembled chromophores with delocalized excitons, are a platform of choice to observe the emergence of superradiance. However, decoherence effects from disorder and thermal noise restrict this behavior to low temperatures.

In this work, we overcome the limitations set by dephasing events and disorder by coupling an emitter of high oscillator strength (J-aggregate) and a cavity with delocalized field distribution and high quality factor (BIC-supporting silicon metasurface). The latter has the purpose of subjecting spatially separate emitters to a common cavity mode, aiding dipole–dipole synchronization, and inducing strong coupling. Within this regime, we explore the influence Rabi splitting ($\hbar\Omega$) has on the exciton (J-exc) and lower exciton-polariton (LP) emission rates, intensities, and photon statistics.

Our sample consists of multiple $50\mu mx50\mu m$ amorphous silicon (Si) metasurfaces that support a bound state in the continuum resonance with distinct energy detunings from the J-exciton. The



FIG. 1. (a) Schematic of the BIC metasurface covered by the J-aggregate film. Angle-resolved (b) reflectance and (c) photoluminescence highlighting the formation of exciton-polaritons.







sample is coated by a thin film (25 nm) of a cyanine-based J-aggregate (TDBC). Three metasurfaces, with distinct energy detunings from the exciton, strongly couple to the exciton forming exciton-polaritons with Rabi splittings of $\hbar\Omega(LP-3) = 320$ meV (Fig. 1(b) (c)), $\hbar\Omega(LP-2) = 283$ meV, and $\hbar\Omega(LP-1)=214$ meV.

Within the strong coupling regime we observe strong emission from the BIC LP (Fig. 2(a)), which when studied as a function of excitation fluence and Rabi splitting shows two linear regimes with fluence thresholds that are inversely proportional to the Rabi splitting (Fig. 2(b)). Additionally, the photon statistics of the BIC LP and J-exc are compared via measurement of the intensity correlation function, $g^{(2)}(\tau)$ (Fig. 2(c)). Here, photon bunching is shown to increase with excitation fluence, and the same fluence threshold is found to determine a regime of enhanced superbunching. These results hint at the onset of inter-aggregate superradiance, where strong coupling induces larger coherence to the exciton via vibronic decoupling [2].



FIG. 2. (a) Photoluminescence spectra of three distinct metasurfaces and their BIC LP with different Rabi splittings. (b) Comparison between the photoluminescence intensity of the J-exc and the BIC LPs as a function of excitation fluence and Rabi splitting. (c) Fluence-dependent study of the photon statistics of the J-exc and BIC LPs. The same fluence threshold associated to the onset of the inter-aggregate emission is observed in (b) and (c).

In conclusion, our results highlight the potential of dielectric BIC metasurfaces for the development of bright and power-efficient superbunched light sources at room temperature.

R. H. Dicke, Coherence in spontaneous radiation processes, Phys. Rev. 93, 99 (1954).
F. C. Spano, Optical microcavities enhance the exciton coherence length and eliminate vibronic coupling in J-aggregates, J. Chem. Phys. 142, 184707 (2015).






Generating phase singularities using surface exciton polaritons in an organic natural hyperbolic material

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Presentation type: Contributed talk

Surface exciton polaritons (SEPs) are electromagnetic waves bound to a surface through the interaction with excitons in the surface material. SEPs provide an alternative to surface plasmon polaritons (SPPs) that could form the basis for all-organic nanophotonics [1]. We perform prism coupling experiments to study surface exciton polaritons excited at visible wavelengths in a thin film of the J-aggregate TDBC. Our experiments were performed using ellipsometry [2], which allows one to simultaneously characterise the intensity (Fig. 1, left panel) and phase (Fig. 1, right panel) response of SEPs. We find that SEPs in TDBC can be of higher-quality than SPPs in Au and Ag films. We further show that SEPs are capable of completely suppressing reflection, resulting in the generation of phase singularities. Our analysis reveals that TDBC is a natural hyperbolic material with giant optical anisotropy [3]. This work confirms that SEPs are a viable alternative to SPPs and suggests that J-aggregates may find wider use in enabling all-organic nanophotonics.



FIG. 1. SEP prism coupling experiment dispersions constructed using the ellipsometric intensity (Ψ , left-hand plot) and phase (Δ , right-hand plot) parameters.

[1] M. G. Gentile, et al., Optical field-enhancement and subwavelength field-confinement using excitonic nanostructures, Nano Lett. 14, 2339 (2014).







[2] P. A. Thomas et al., A new signature for strong light-matter coupling using spectroscopic ellipsometry, Nano Lett. 20, 6412 (2020).

[3] E. S. H. Kang et al., Organic anisotropic excitonic optical nanoantennas, Adv. Sci. 9, 2201907 (2022).







Fano resonances between the emission of J-aggregate-covered metallic nanorods and spherical cavity modes

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Hybrid plasmonic-photonic resonators can enhance the performance of quantum emitters, such as J-aggregates of dye molecules or quantum dots, in terms of both intensity and tunability. This enhancement is based on the cooperative Purcell effect arising from both plasmonic and microcavity (MC) confinement of the interacting electromagnetic (EM) field and elementary excitations responsible for the emission [1]. For instance, plasmonic modes of metallic nanoparticles deposited on a MC can couple to optical whispering-gallery modes (WGMs) to maximize the system's absorbance [2]. On the other hand, fluorescence spectra and lasing behavior of dye molecules or J-aggregates adsorbed on a dielectric sphere are modified clearly showing the influence of the WGMs [3,4].



FIG. 1. Left panel: Scheme of the experiment: (i) J-aggregate-covered colloidal Ag nanorods are dispersed on the surface of a $7 \mu m$ silica sphere; (ii) The system is excited with a laser beam focused as shown in the figure and its photoluminescence (PL) is recorded. **Right panel**: Experimental PL emission spectrum under 532 nm laser excitation. Vertical lines are calculated eigenmodes of the sphere for various values of the mode numbers n (superscript) and l (subscript).







frequently observed feature in such systems is an asymmetric profile of either emission [1] or absorption [2] lineshape, known as Fano resonance. However, only a quasi-periodic structure of narrow photoluminescence (PL) peaks was observed in Ref. [4], presumably arising from the coupling between the emission of J-aggregates and WGMs.

In this work, we investigated PL spectra of PIC J-aggregates covering silver nanorods (Ag NRs), deposited on the surface of a spherical MC (see Fig. 1). The spectra show a series of Fano-shape resonances in the broad spectral range of 550 - 650 nm. The configuration shown in Fig. 1 is typically used for the WGM's excitation [5] and the PL spectrum features indeed show perfect correlation with the frequencies of specific EM modes with high angular momenta, l, calculated using the Mie theory. We are not aware of previous observations of Fano resonances in the emission of J-aggregates. By comparing these results to Ref. [4], it can be concluded that Ag NRs are an essential ingredient for their observation. Two more experimental facts corroborate this conclusion:

(1) The emission spectrum of the J-aggregates, quite narrow when they are free in solution, broadens considerably when they are adsorbed on Ag NRs suspended in the same solution and covers almost the whole spectral range shown in Fig. 1;

(2) The absorption spectrum of such a solution, generally little affected by the presence of the Jaggregates (that is, the absorption is mostly due to localised surface plasmons in the NRs), contains a Fano-type feature related to the absorption peak of the J-aggregates.

We propose a theoretical attempt of putting these pieces of puzzle together by (i) considering Fano resonances originating from a weak coupling between localised surface plasmons in Ag NRs and the WGMs, and (ii) calculating the hybrid local density of states in the cavity, near its surface, that modulates the J-aggregate emission spectrum.

[1] H. M. Doeleman et al., Observation of cooperative Purcell enhancements in antenna-cavity hybrids, ACS Nano 14, 12027 (2020).

 [2] N. Thakkar et al., Sculpting Fano resonances to control photonic-plasmonic hybridization, Nano Lett. 17, 6927 (2017).

[3] H. Yukawa et al., Microcavity effect of dyes adsorbed on a levitated droplet, Phys. Rev. A 60, 2491 (1999).

[4] D. Melnikau et al., Whispering gallery mode resonators with J-aggregates, Opt. Express 19, 22280 (2011).

[5] X. Zambrana-Puyalto et al., *Excitation mechanisms of whispering gallery modes with direct light scattering*, Laser Photon. Rev. 15, 8 (2021).







Mapping between quantum optics and nanophotonics

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Presentation type: Invited talk

In this talk I will present the main findings of our efforts to developing an accurate theoretical formalism, able to capture the complex and quantum nature of the electromagnetic (EM) fields associated with nanophotonic structures, but simple enough to be used utilizing the well-established tools of quantum optics. We will start by introducing the general framework of Macroscopic QED, highlighting the key role played by the spectral density when analyzing the dynamics of quantum emitters interacting with nanophotonic systems. Then we will present the main idea of our formalism, which is to replace the infinite cloud of medium-assisted EM field by a finite network of coupled EM modes. In this way, what is needed is to find network parameters that reproduce the nanophotonic spectral density closely enough. We will discuss how this formalism can be extended to deal with multiple emitters, ultra-strong coupling and periodic dielectric media. I will end by discussing how our method can be also utilized to have access to the spatially resolved photon statistics of light emerging from quantum emitters interacting with nanophotonic structures.







Electrical conductivity in amorphous polymers under vibrational strong coupling

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Presentation type: Contributed talk

It is established that electrical conductivity of organic molecules can be boosted by electronic strong coupling [1-4]. We have shown recently that vibrational strong coupling of intrinsically non-conducting and amorphous polymers such as polystyrene, deuterated polystyrene, and poly (benzyl methacrylate) enhances the electrical conductivity by at least six orders of magnitude compared to the uncoupled polymers [5]. Further, we explored the role of VSC in improving the conductivity in other aromatic and aliphatic polymers. Remarkably, the enhanced conductance is vibrational mode selective and occurs only under the VSC of the aromatic C-H(D) out-of-plane bending modes of the polymers. Polymers like poly (methyl methacrylate), poly (vinyl alcohol) which does not contain the aromatic groups, remain insulating under VSC. I will also discuss our progress in using VSC to connect the spatially separated conducting polymers.

 E. Orgiu et al., Conductivity in organic semiconductors hybridized with the vacuum field, Nat. Mater. 14, 1123 (2015).

[2] N. Krainova et al., Polaron photoconductivity in the weak and strong light-matter coupling regime,Phys. Rev. Lett. 124, 177401 (2020).

[3] P. Bhatt et al., Enhanced charge transport in two-dimensional materials through light-matter strong coupling, ACS Nano 15, 13616 (2021).

[4] K. Kaur et al., Controlling electron mobility of strongly coupled organic semiconductors in mirrorless cavities, Adv. Funct. Mater. 33, 2306058 (2023).

[5] S. Kumar et al., *Extraordinary electrical conductance through amorphous nonconducting polymers under vibrational strong coupling*, J. Am. Chem. Soc. 146, 18999 (2024).







Extended lasing states in electrically tuneable liquid-crystal microcavity

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Presentation type: Contributed talk

Non-equilibrium Bose–Einstein condensates (BEC) in photonic systems are promising for exploring and engineering phases of matter under extreme conditions and can be used to perform analogue simulations at both cryogenic and room temperatures. In order to establish a robust and scaleable platform for computing based on non-equilibrium photonic condensates problems of tuneability and reconfigurability should be addressed. Here we demonstrate the realisation of coherent coupling between individually pumped lasing states and formation of spatially extended macroscopically occupied states in a microcavity filled with liquid crystals (LC) and P580 dye (dve-doped LCMC) at room temperature. The saturation of the optical transition in P580 laser dye leads to the blueshift of the photoluminescence aboth the lasing threshold similar to the mechanism of the emission blueshift reported for strongly coupled organic microcavities [1]. It results in a ring-shaped emission profile in the momentum space and sufficient in-plane momentum of the coherent emission leading to coherent energy exchange between spatially separated individually pumped states, similar to the well known behaviour of polaritonic BEC in GaAs systems at cryogenic temperatures. We bring optical reconfigurability by controllable driving of each lasing state with a focused non-resonant optical excitation and utilise it to form a coupled state in a dyad, 1D chain, and a 2D lattice of condensates with the coupling strength defined by the distance between the pumping spots, pump power, and polarisation of the pump. We demonstrate the possibility of going beyond the nearestneighbour limit of coupling in a chain of coherently coupled emitters by controlling the polarisation of the pump. Furthermore, we show electrical control over interaction between coupled lasing states







by applying external voltage. High birefringence of the LC provides wide range tuneability of the dispersions for the microcavity optical modes with different polarisation and allows for control over coupling efficiency by changing the in-plane component of the emission. In Fig. 1, we demonstrate 3 distinctive regimes of interaction.



FIG. 1. (a) **Electrically tuneable coupling in a dyad configuration**. Experimental images of the lasing state emission in (a-c) real-space, (d-f) momentum space for two pump spots at (a,d) 0 V, (b,e) 1.6 V, and (c,f) 1.84 V, showing (a) coupled supermode lasing state, (b) uncoupled separate lasing spots, and (c) extended lasing state in Rashba–Dresselhaus regime correspondingly.

We are able to switch on (Fig. 1a,d) and off (Fig. 1b,e) the coupling in a dyad configuration by tuning the external voltage, as well as to bring the LCMC into the Rashba–Dresselhaus spinorbit coupling regime (Fig. 1c,f) [2], where the emission is separated in the momentum space and becomes circularly polarized. The latter regime allows for the realisation of momentum-space separated chiral lasing from the coupled spatially extended lasing states at room temperature and possibility for electro-optically controlled propagation of the spin information in coupled arrays of coherent emitters.

 T. Yagafarov et al., Mechanisms of blueshifts in organic polariton condensates, Commun. Phys. 3, 18 (2020).

[2] M. Król et. al., Realizing optical persistent spin helix and Stern-Gerlach deflection in an anisotropic liquid crystal microcavity, Phys. Rev. Lett. 127, 190401 (2021).







Tuesday, 17 June: Poster session







Hyperspectral camera for inline spectral analysis of organic solar cells

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Presentation type: Poster

The slot-die roll-to-roll (R2R) coating technique is widely recognized for its effectiveness in the scalable production of organic photovoltaic (OPV) devices. This method enables high-speed deposition while maintaining precise control over film thickness and uniformity [1]. In this study, we introduce a hyperspectral camera system designed for inline, spatially resolved monitoring of single and multilayer coatings in organic solar cells. The system features a highresolution sensor with 2160 x 2560 pixels, where 2160 pixels are dedicated to spatial tracking and 2560 pixels to spectral acquisition. After binning, the spatial resolution reaches 143 microns per pixel, with a spectral resolution of less than 1 nm. The system operates within a spectral range of 453 to 718 nm.

Using these parameters, we successfully performed inline measurements and analysis of the transmission spectra of poly(3-hexylthiophene-2,5-diyl) P3HT and OIDTBR* blends, achieving spatially resolved deconvolution of absorption peaks. We applied the multivariate curve resolution method to determine the spatial distribution of P3HT and OIDTBR concentrations in the blend film. Additionally, the system was capable of detecting defects and drying patterns. For thicker semitransparent films, we demonstrated through inline measurements and offline analysis the thickness of Polyethylene terephthalate (PET), Poly(3,4-ethylenedioxythiophene):polystyrene sulfonate (PE-DOT:PSS) wet films, and Zinc Oxide (ZnO) sputtered films. Due to the thin nature of the active layers, we cannot measure their absolute thickness, however, Fig. 1 shows the inline measurements of the standard deviation map of thickness of a 1:1 (w:w) P3HT:OIDTBR sample deposited by slot die coating.







The hyperspectral camera system offers spatially resolved inline analysis across the full coated strip width of up to 15 mm, enabling the creation and detailed analysis of real-time property maps of the films. This system is a promising tool for optimizing the slot-die coating process, with the potential to enhance research, innovation, and scalability in the field of organic photovoltaics. The high spectral resolution provides the capability for spatial mapping of optical properties, including molecular resonances as realized in cavity structures enabling strong light–matter coupling.



FIG. 1. Standard deviation map over several hundreds of frames.

[1] L. Zhu et al., Achieving 20.8% organic solar cells via additive-assisted layer-by-layer fabrication with bulk p-i-n structure and improved optical management, Joule 8, 3153 (2024).

* OIDTBR full chemical formula: (5Z)-3-ethyl-5-[[4-[15-[7-[(Z)-(3-ethyl-4-oxo-2-sulfanylidene-1,3 thiazolidin-5-ylidene)methyl]-2,1,3-benzothiadiazol-4-yl]-9,9,18,18-tetra(nonyl)-5, 14dithiapentacyclo[10.6.0.03,10.04,8.013,17]octadeca 1(12),2,4(8),6,10,13(17),15-heptaen-6-yl]- 2,1,3-benzothiadiazol-7 yl]methylidene]-2-sulfanylidene-1,3-thiazolidin-4-one.







Organic polaritons enable high-responsivity, angle-independent narrowband microcavity photodiodes

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Presentation type: Poster

Microcavities have become a widely adopted strategy for enabling narrowband spectral selectivity, particularly in organic photodiodes (OPDs). However, their utility is hindered by pronounced angular dispersio—shifts exceeding 50 nm at 45° tilt—which compromises angle-independent operation. To address this limitation, we engineer microcavities that strongly couple photonic resonances with molecular transitions in the material, forming hybrid light-matter polaritons. These states inherit suppressed angular dispersion from their material component while retaining photonic spectral selectivity, offering a transformative platform for practical narrowband detection.

In this work, we demonstrate polariton-enhanced OPDs (Fig. 1a) that simultaneously achieve narrowband responsivity and exceptional angular stability. The optimized devices exhibit a spectral full-width at half-maximum (FWHM) of < 30 nm and a record responsivity up to 0.24 A/W at 965 nm under -2 V bias. Crucially, the polariton photoresponse shows negligible angular dispersion across a $\pm 45^{\circ}$ angular range, enabling robust operation under oblique light incidence (Fig. 1b). Remarkably, this performance exceeds existing state-of-the-art spectral-narrowing technologies (Fig. 1c). By unifying high responsivity, spectral precision, and angle insensitivity, polariton OPDs represent a critical advancement toward deployable narrowband optoelectronics for imaging, sensing, and spectroscopic systems. This work establishes polaritons not only as a versatile design paradigm for mitigating angular dispersion in cavity-integrated devices but also that







OPDs represent an exceptionally promising device configuration where polaritonic effects can lead to substantial and undisputed improvements in performance.



FIG. 1. (a) Simplified scheme of the polariton OPD with a sketch of the interaction between the cavity resonance and the exciton. (b) Angle-resolved responsivity maps from 0° to 45° with an interval of 15° for the polariton OPDs with device resonance of 945 nm (top) and 965 nm (bottom). *c) Comparison of the responsivities of narrowband OPDs with different spectral narrowing techniques at 0 V with peak wavelengths between 940 nm and 990 nm. CCN: Charge Collection Narrowing, EDN: Exciton Diffusion Narrowing, MC: Microcavity Narrowing.

[1] A. G. Abdelmagid et. al., Polaritons in non-fullerene acceptors for high responsivity angleindependent organic narrowband infrared photodiodes arxiv:2412.06741 (2024).

[2] Y. Wang et al., Narrowband organic photodetectors-towards miniaturized, spectroscopic sensing, Mater. Horiz. 9, 220 (2022).

[3] B. Siegmund et al., Organic narrowband near-infrared photodetectors based on intermolecular charge-transfer absorption, Nat. Commun. 8, 15421 (2017).







Understanding the role of molecular aggregation in polariton formation

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Presentation type: Poster

J-aggregates, organic chromophores that self-assemble into supramolecular structures with large transition dipole moments and ultra-narrow line widths [1], are commonly used to enter the strong coupling regime [2-6]. Despite their widespread use in the field, experimental results are often interpreted within the framework of the Tavis–Cummings Hamiltonian [7], which neglects dipolar coupling between monomeric units in J-aggregates. However, recent theoretical work by Bradbury et al. demonstrates that excitonic coupling within the cavity significantly influences polariton properties, particularly in the presence of strong disorder [8]. Here, we elucidate the role of aggregation on polaritonic spectral signatures by incorporating thin films of monomeric and aggregated cyanine dyes into multimode Fabry-Pérot cavities with fixed coupling strength [Fig. 1(a)]. Our preliminary work demonstrates that the monomeric cavity is not strongly coupled [Fig. 1(b)], while we expect that the J-aggregate-containing cavity will easily afford polaritons. Not only does our work highlight the importance of including dipolar coupling in theoretical descriptions of strongly coupled aggregates but also demonstrates a facile method to deposit solution processable monomeric films of cyanine dyes at relatively high concentrations.

[1] J. L. Bricks et al., *Fluorescent J-aggregates of cyanine dyes: basic research and applications review*, Meth. Appl. Fluor. 6, 012001 (2018).

[2] M. Rödel et al., Anisotropic photophysical properties of plexcitons in strongly coupled metalorganic thin films, ACS Photonics 12, 107 (2025).

[3] M. Russo et al., Direct evidence of ultrafast energy delocalization between optically hybridized J-aggregates in a strongly coupled microcavity, Adv. Opt. Mater. 12, 2400821 (2024).

[4] E. Hulkko et al., Effect of molecular Stokes shift on polariton dynamics, J. Chem. Phys. 154, 154303 (2021).









FIG. 1. (a) Upper: Cartoon depicting proposed cavity experiments containing monomeric or J-aggregated cyanine dyes. Monomeric units are shown as bricks. Lower: Absorption spectrum of monomeric and J-aggregated thin films, with chemical structure of chosen dye shown in the inset. (b) Angle-resolved transmission spectra of cavities containing monomeric cavities spin coated at 5200 rpm (upper) and 6700 rpm (lower).

[5] D. G. Lidzey et al., Observation of strong exciton-photon coupling in semiconductor microcavities containing organic dyes and J-aggregates, Opt. Mater. 12, 243 (1999).

[6] D. G. Lidzey et al., Room temperature polariton emission from strongly coupled organic semiconductor microcavities, Phys. Rev. Lett. 82, 3316 (1999).

 M. Tavis and F. W. Cummings, Exact solution for an N-molecule-radiation-field Hamitonian, Phs Rev. 170, 379 (1968).

[8] N. C. Bradbury et al., Stochastic methodology shows molecular interactions protect twodimensional polaritons, Phys. Rev. B 109, L241303 (2024).







Hexagonal silica microsphere monolayers supporting surface phonon-polaritons for radiative cooling applications

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Presentation type: Poster

Silica exhibits remarkable optical properties in the mid-infrared (MIR) spectrum (1.a). As a polar material, silica presents transverse optical (TO) phonon modes in the MIR, leading to a high reflectance in a spectral region known as the Restrahlen band. Within this band, TO phonon can be strongly coupled with photons, leading to the appearance of Surface Phonon-Polaritons (SPhPs) [1]. These SPhPs increase the density of optical states at the material's surface, i.e., they enhance near-field confinement. By structuring a periodic structure on the material, it is possible to match these modes with far-field diffraction modes, enhancing the material's radiative emission.

In the case of silica, the Restrahlen band lies within the so-called Atmospheric Transmission Window (ATW), a spectral region where the atmosphere is transparent (1.b). This spectral coincidence can be exploited by designing silica microstructures to enhance thermal radiation from Earth-temperature bodies, facilitating heat dissipation into outer space. Recently, we demonstrated that a close-packed hexagonal monolayer of 8μ m silica microparticles, forming diffraction gratings, can be used for radiative cooling applications [2].

In this work, we have prepared large areas of close-packed hexagonal monolayers of silica microparticles with diameters ranging from 2μ m to 10μ m (1.c). This fabrication method is based on a self-assembly approach where a dispersion of microparticles is deposited onto a substrate through meniscus-assisted assembly. The optical response of the monolayers in the ATW was studied using FTIR a function of the microparticle size. We compare the observed optical response with the presence of SPhPs in the microstructures by modelling their electromagnetic response. The objective is to determine the optimal particle size for radiative cooling applications and correlate with the SPhPs supported by microstructure.









FIG. 1. (a) Optical constants of silica in the MIR region with the Restrahlen band marked in red. (b) Atmospheric transmission. (c) Scanning Electron Microscope images of the prepared monolayers with silica particle diameters of i) 2μ m, ii) 4μ m, iii) 6μ m and iv) 8μ m.

T. G. Folland et al., Probing polaritons in the mid- to far-infrared., J. Appl. Phys. 125, 191102.
 (2019).

[2] J. Jaramillo-Fernandez et al., A self-assembled 2D thermofunctional material for radiative cooling, Small 15, 1905290 (2019).







Multimode strong coupling in plasmonic nanocavities

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Presentation type: Poster

Plasmonic nanocavities are an exciting platform to access the quantum properties of light and matter [1]. In particular, they offer a unique pathway to the strong coupling regime —where light and matter reversibly exchange energy— due to their deep subwavelength mode volumes [2]. Most descriptions of plasmonic strong coupling use a single-mode approximation —following the success of single mode models in dielectric cavities. However, plasmonic nanocavities are unique since they support a dense collection of modes that have large coupling strengths [3], and in general, the origin and impact of these modes on the quantum dynamics is not known.

In this work [4] we show that in contrast to most other cavities, off-resonant plasmonic modes play a crucial role in strong coupling. In particular, we show that n strongly coupled plasmonic modes introduce up to n(n+1)/2 oscillation frequencies in the quantum dynamics. The amplitudes and frequencies of these oscillations depend on the coupling strengths and detunings of each mode and therefore, we also identify three distinct regions depending on those parameters: (i) single mode, (ii) multimode, and (iii) collective multimode strong coupling. In the single mode coupling regime, the quantum dynamics are governed by a single resonant mode. In the multimode coupling regime, the off-resonant modes become significant and produce multiple distinct oscillations frequencies, and in the collective coupling regime (which occurs past a critical dipole moment) the light–matter interaction is dominated by a single ultrafast oscillation. This ultrafast oscillation is due to the collective coupling with multiple modes, and is over one order of magnitude faster than if a single mode approximation is made.

In Fig. 1(a) we show a single quantum emitter (QE) interacting with a common plasmonic system, the nanoparticle on mirror (NPoM) cavity. The electric field profile of the quasinormal modes (QNMs) in the gap centre are shown in Fig. 1(b) and are labelled with the index $\xi = (\ell 0)$. To determine their effect on strong coupling, we first consider a single QE with transition frequency $\omega_0 = \omega_{(10)} = 283$ THz and dipole moment $\mu = 72$ $\hat{\mathbf{z}}$ D. In Fig. 1(c) we show the quantum dynamics when including interacting modes up to $\xi = (\ell_{\text{max}} 0)$. For a single mode ($\ell_{\text{max}} = 1$) the QE exchanges







FIG. 1. Multimode strong coupling (a) Schematic of a nanoparticle on mirror (NPoM) cavity; a single QE is placed at the gap centre. (b) Electric field $E_z(x, y, 0)$ of the first nine $\xi = (\ell 0)$ quasinormal modes (QNM's). (c) Evolution of the QEs excited state population n(t) when including interacting modes up to $\xi = (\ell_{\text{max}}0)$ for $\ell_{\text{max}} = 1, 2, 3, 7, 9$ where the QE has frequency $\omega_0 = \omega_{(10)} = 283$ THz and dipole moment $\mu = 72$ \hat{z} D.

energy with the cavity at a single (Rabi) frequency. However, as more modes are included in the interaction, the quantum dynamics become more complicated. In particular, when $\ell_{\text{max}} = 9$ the evolution is dominated by a single ultra-fast oscillation; this is collective multimode strong coupling.

These insights advance the understanding of quantum dynamics in realistic plasmonic systems, and highlight their potential for enabling ultra-fast energy transfer in quantum technologies.

[1] M. S. Tame et al., Quantum plasmonics, Nat. Phys. 9, 329 (2013).

[2] R. Chikkaraddy et al., Single-molecule strong coupling at room temperature in plasmonic nanocavities, Nature 535, 127 (2016).

[3] N. Kongsuwan et al., *Plasmonic nanocavity modes: from near-field to far-field radiation*, ACS Photonics 7, 463 (2020).

[4] A. Crookes et al., Collective multimode strong coupling in plasmonic nanocavities, arXiv:2411.07694 (2024).







Terahertz control in a transmission electron microscope

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Presentation type: Poster

Ultrafast transmission electron microscopy enables the measurement of fundamental processes in space and time due to its extremely high spatial and temporal resolution. We demonstrate the first-ever generation of isolated 19 fs electron pulses while simultaneously preserving the extremely high spatial resolution in a transmission electron microscope [1]. This represents a breakthrough for studying faster physical processes such as phonon and plasmon dynamics, phase transitions and ultrafast electronics on ultimate spatial and temporal scales, as previously generated isolated electron pulses in a transmission electron microscope were limited to 200 fs [2]. Fig. 1 shows a schematic sketch or our ultrafast transmission electron microscope. Ultra-short laser pulses first generate electron pulses with a pulse duration of up to 200 fs by photoemission. To attain enhanced temporal resolution, these electron pulses must be subsequently compressed in time. For this purpose we use a terahertz waveguide that is provided with a hole. This allows electrons to interact with the electric field of the terahertz pulses and thus be modulated in energy and time.

Using the terahertz waveguide, we demonstrate isolated electron pulse compression down to 19 fs, while maintaining flat pulse fronts, which is a 10-fold improvement to previous works. By adapting the waveguide we can further show electron deflection in space and electron acceleration of up to 100 eV. These results prove that the terahertz waveguide is a very effective all-optical tool to control electrons within a transmission electron microscope, making it a ground breaking device for nanophotonics, metamaterials, material science, electron quantum optics and other related research









FIG. 1. Experimental setup for electron pulse compression in a transmission electron microscope. Ultrashort laser pulses (green) generate electron pulses via photoemission. Interaction between the terahertz electric field (violet) and the electron pulses (blue) within the metallic waveguide (orange) leads to compression of the electron pulses at the sample stage. This enables pump probe experiments utilizing a pump laser (red) and the compressed electron probe pulses. On the right, we display the phase space diagram of the electron pulses in the energy and time domain.

 J. Kuttruff et al., Terahertz control and timing correlations in a transmission electron microscope, Sci. Adv. 10, eadl6543 (2024).

[2] A. Feist et al., Ultrafast transmission electron microscopy using a laser-driven field emitter: femtosecond resolution with a high coherence electron beam, Ultramicroscopy 176, 63 (2017).







Organic excitonic nanostructures for weak and strong coupling

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Presentation type: Poster

Organic thin-film-based excitonic nanostructures are a promising alternative for plasmonic systems. Such organic films exhibit Frenkel excitons at room temperature with high absorption coefficient. These excitonic materials are derived from common organic dye-doped polymer films by increasing the doping concentration so much, that a real part of the permittivity goes negative, $\mathcal{R}\{\epsilon(\omega)\} < 0$, within an energy range just above their material absorption. Within this narrow band of negativereal-permittivity, the organic film supports surface exciton (SE) modes analogous to the surface plasmon (SP) modes in metals. These modes exist as propagating ones on top of the film or/and localized SE modes in organic excitonic nanostructures [1,2]. Like SP modes, SE modes can be exploited in refractive index sensing and near-field enhanced spectroscopy in the weak coupling regime [3] and as an optical cavity in the strong coupling regime [4].

In this work, we report how to enhance the optical performance of the SE modes by optimizing the choice of excitonic dyes as well as their doping concentration, and show how they can be utilized as the resonator mode for the strong coupling applications. In the case of weak coupling, we study the effect of molecular concentration in terms of oscillator strength and Lorentzian broadening on various SE modes when employed in sensing and spectroscopy. The optical performance of the SE modes is evaluated in terms of sensing, like sensitivity and figure of merit, as well as near-field enhancement, like enhancement factor and field confinement. In the case of strong coupling, we study whether SE modes can strongly couple to the photoactive molecules and SP modes or not, and whether the polariton modes are visible.

Our simulations reveal that, in the weak coupling regime, in general, an increase in oscillator strength of the excitonic dye enhances the performance of the SE modes while a broadening in the dye absorption linewidth degrades that as a counteracting effect. This demonstrates that the optical performance of an excitonic system is tunable via molecular concentration, which is a clear advantage over the plasmonic systems. In addition, different SE modes show different degrees of tunability and equivalency in performance when compared to SP modes in silver and gold.







In the strong coupling regime, our numerical findings reveal that SE modes can facilitate strong coupling by sustaining the energy-splitting-induced transparency. However, the polaritons may not be visible in the absorption since they can easily be located outside of the narrow energy-range of negative-real-permittivity. Also it seems that the SE modes cannot couple strongly with the SP modes.

Our findings shed light on the weak and strong coupling properties of SE modes and provide crucial information for developing and optimizing novel excitonic nanodevices for contemporary organic nanophotonics.

[1] L. Gu et al., Quest for organic plasmonics, Appl. Phys. Lett. 103, 021104 (2013).

[2] M. J. Gentile et al., Optical field-enhancement and subwavelength field-confinement using excitonic nanostructures, Nano Lett. 14, 2339 (2014).

[3] A. Dutta and J. J. Toppari, Effect of molecular concentration on excitonic nanostructure based refractive index sensing and near-field enhanced spectroscopy, Opt. Mater. Express 13, 2426 (2023).
[4] A. Dutta and J. J. Toppari, Weak and strong coupling properties of surface excitons, Phys. Rev. B 109, 165117 (2024).







Simulating nuclear quantum effects in electronic strong coupling

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Presentation type: Poster

Experiments suggests that the intrinsic properties of organic photo-active materials can change when placed inside an optical cavities, including their electrical and energy conductivity, photoemission quantum yield, lasing threshold, or even chemical reactivity. While these changes have been attributed to the hybridization between excitations in the material and the confined light modes of the cavity into polaritons, the current theoretical understanding of these phenomena is too limited to systematically control chemistry with cavities. To bridge this gap, semi-classical molecular dynamics models have been developed that not only treat the collective nature of the coupling in the aforementioned experiments, with very many molecules coupled to the cavity, but also maintain chemical accuracy to describe reactions. However, the semi-classical approximation precludes the investigation of nuclear quantum effects, which could play a key role in relaxation, lasing or reactivity. To investigate the limitations of semi-classical MD for polaritonic chemistry, we systematically compared polariton dynamics between semi-classical and fully quantum mechanical simulations of realistic model systems, using with the multi-configuration time-dependent Hartree (MCTDH) method. Our comparison suggests that for small molecules, the results are qualitatively in good agreement, and hence that the semi-classical approach provides a good compromise between accuracy and computational efficiency for investigating the effects of strong light mater coupling on photo-chemistry. Nevertheless, although minor, nuclear quantum effects play a role, which will be discussed in the presentation.

[1] M. Tavis and F. W. Cummings, Approximate solutions for an N-molecule-radiation-field Hamiltonian, Phys. Rev. 188, 692 (1969).

[2] O. Vendrell, Collective Jahn-Teller interactions through light-matter coupling in a cavity, Phys.
 Rev. Let. 121, 253001 (2018).







[3] R. H. Tichauer et al., Multi-scale dynamics simulations of molecular polaritons: the effect of multiple cavity modes on polariton relaxation, J. Chem. Phys. 154, 104112 (2021).
[4] I. S. Ulusoy et al., Modifying the nonradiative decay dynamics through conical intersections via collective coupling to a cavity mode, J. Phys. Chem. A 123, 8832 (2019).







Optoelectronic response of light–matter interaction in ultrathin excitonic systems

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Presentation type: Poster

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Strong coupling occurs when the interaction between light and matter—such as the Fabry–Pérot (FP) resonance and excitons in a semiconductor—becomes so strong that the two can no longer be considered separate, leading to the formation of hybrid states known as polaritons. On the other hand, non-trivial phase shifts in an ultrathin, lossy material on a reflective substrate also produce prominent resonances [1], and when these resonances interact with excitons, they yield splitting in the reflectance similar to that seen in strong coupling. Although such phenomena have been suggested to form exciton-polaritons [2], our photocurrent measurements demonstrate that the interaction between absorption resonance and excitons does not alter the optoelectronic properties of the system or generate polaritons. We further examine how phase shifts in the resonant optical mode affect the hybridized system and its optoelectronic behavior—a facet that has not yet been fully explored.

A thin flake on Au substrate acts as a lossy Gires–Tournois interferometer with an absorption resonance (Fig. 1(a)), whereas a thicker flake exhibits FP resonance without the need for a reflective substrate (Fig. 1(b)). Figure 1(c) presents individual normalized external quantum efficiency (NEQE, calculated from photocurrent measurements) and absorptance plots for 12 nm and 80 nm thick flakes. For the 80 nm flake, results from both the experiments match nicely showing prominent Rabi splitting around A-exciton and confirming polariton formation via FP–exciton coupling and its impact on optoelectronic properties. For the 12 nm flake, although we observe splitting in absorptance measurement, NEQE shows only distinct A- and B-exciton peaks, indicating that absorption resonance—exciton interaction does not generate polaritons. Simulations across varying flake thickness also show clear absorptance splittings for both thin and thick flakes (Fig. 1(d), regions (1) and (2)). Moreover, both in simulation and experiments, region (3) shows hybridization







FIG. 1. (a) Schematic illustrating the destructive interference in thinner flakes responsible for an absorption resonance. (b) Schematic depicting the FP resonance supported by thicker WS₂ flakes. (c) Experimental absorptance derived from reflectance measurements (brown) and NEQE (blue) spectra for WS₂ flakes with thicknesses of t = 12 nm and t = 80 nm are plotted together for comparison (vertical dashed lines indicate A- and B-exciton wavelengths). (d) Simulated 2D map of absorptance (z axis, in arb.u.) across varying flake thicknesses alongside experimental results, where the blue dices indicate the spectral positions of maxima from NEQE measurements and the brown dots mark the maxima from absorptance measurements. (e) NEQE and reflectance measurements for a 64 nm thick flake stacked on different substrates.

with the weaker B-exciton, and region (4) reveals higher-order hybridization where the FP mode interacts with both A- and B-excitons. Finally, Fig. 1(e) compares results for a 64 nm thick flake, showing that when the substrate is changed from Au to less reflective SiO₂-on-Si, the NEQE (and reflectance) maxima (and minima) exhibit a red shift (marked by arrows (1) and (2)). This shift is attributed to the absence of the additional phase induced by gold's reflective surface, causing the flake to behave as if it were slightly thicker on SiO₂-on-Si than on gold. For Au substrate, we also observe blue shift of B-exciton (arrow 3) wavelength because of higher-order coupling.

This study advances our understanding of light-matter interactions and the corresponding optoelectronic responses in multilayer lossy excitonic systems.

 M. Kats et al., Nanometre optical coatings based on strong interference effects in highly absorbing media, Nat. Mater. 12, 20 (2013).

 [2] H. Zhang et al., Hybrid exciton-plasmon-polaritons in van der Waals semiconductor gratings, Nat. Commun. 11, 3552 (2020).







Topology optimization of optical devices and their potential application in polaritonic chemistry

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Presentation type: Poster

Topology optimization (TO) is a powerful inverse design methodology widely used in classical optics [1]. This allows the fabrication of optical devices tailored towards specific characteristics by the intuitive modification of continuum parameters. In this work, we present our TO implementation that works with Maxwell equations in frequency domain and in a 2D grid. This implementation allows us to design devices that shape the radiated field emitted by classical sources, thereby enhancing phenomena such as the energy transfer between two or more points and the Purcell effect. The resulting designs are applied then to enhance the interaction between light and molecules. For this, we include the devices in Finite-Difference Time-Domain (FDTD) simulations [2,3], replacing classical emitters with realistic molecules modeled by the quantum mechanical simulation software DFTB+ [4]. The resulting framework allows us to simulate enhanced energy transfer processes among molecules, accelerated radiative decay through the Purcell effect, and the formation of polaritons. Our results demonstrate the potential of optical devices, designed with TO, to specifically control processes within polaritonic chemistry. Additionally, our findings illustrate the advantages and potential of semiclassical methodologies in simulating light–matter interaction in strong-coupling regimes at low computational costs and under realistic conditions.

[1] A. M. Hammond et al., *High-performance hybrid time/frequency-domain topology optimization* for large-scale photonics inverse design, Opt. Express 30, 4467 (2022).

 M. Sukharev et al., Dissociation slowdown by collective optical response under strong coupling conditions, J. Chem. Phys. 158, 084104 (2023).







[3] M. Sukharev, Efficient parallel strategy for molecular plasmonics — A numerical tool for integrating Maxwell-Schrödinger equations in three dimensions, J. Comput. Phys. 477, 111920 (2023).
[4] B. Hourahine, DFTB+, a software package for efficient approximate density functional theory based atomistic simulations, J. Chem. Phys. 152, 124101 (2020).







A multi-scale approach for modeling strong coupling in open nanocavities probed by electrons

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Composite nanostructures combining components that support confined electromagnetic modes with electronic transitions under certain conditions can enter the strong coupling regime, manifested by the emergence of hybrid light-matter states. The intriguing optical properties of these polaritonic states are promising for a wide range of applications, including polaritonic chemistry, polaritonic lasing, and Bose–Einstein condensation [1].



FIG. 1. (a,b) Electron energy loss (EEL) probability for an electron traveling parallel to the axis of a silver wire coated with a coaxial layer of thickness 20 nm made of (a) J-aggregates, and (b) ZnMOF for varying core radius. (c) EEL probability of the uncoated silver wire. In all panels, we consider an electron of kinetic energy 200 keV, passing at distance 5 nm from the surface of the wire. The vertical red lines trace the energy of the electronic transitions in the organic molecules.

Achieving a large coupling strength requires two key ingredients: a nanocavity that sustains tightly confined modes and an electronic transition associated with a strong dipole moment. Plasmonic nanocavities embedded in excitonic materials comprising organic molecules, such as metal–organic frameworks (MOF) and J-aggregates, are ideal platforms. In that regard, fast electron beams are excellent probes to study this light–matter interaction, as they can excite very tightly







confined modes with high quality-factors that are otherwise inaccessible with conventional lightbased methods [2].

The theoretical modeling of molecular materials requires quantum chemistry calculations, most commonly based on the time-dependent density functional theory (TDDFT). However, such computations are feasible only at molecular material sizes up to 1000 atoms, whereas larger nanoscale designs require a macroscopic approach. This work uses a multi-scale approach for studying strong coupling in open nanocavities probed by fast electron beams. Starting from TDDFT, we derive the macroscopic effective optical parameters of the excitonic material via a homogenization method and simulate the optical response of the composite plasmonic–excitonic nanostructure using the T-matrix formalism [3]. We apply the method to study the coupling between silver nanowires covered in MOFs and J-aggregates coatings, as illustrated in Fig. 1, and the resulting polaritonic branches. Our results reveal large energy splittings —signatures of strong coupling—, which are tunable through the geometry of the structure and the electron beam parameters.

[1] P. Törmä and W. L. Barnes, Strong coupling between surface plasmon polaritons and emitters: a review, Rep. Prog. Phys. 78, 013901 (2015).

[2] F. J. García de Abajo, Optical excitations in electron microscopy, Rev. Mod. Phys. 82, 209 (2010).

[3] B. Zerulla et al., A multi-scale approach for modeling the optical response of molecular materials inside cavities, Adv. Mater. 34, 2200350 (2022).







Polaritonic engineering in non-fullerene acceptor organic photovoltaic devices

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Enhancing the quantum efficiency of organic photovoltaic (OPV) devices by leveraging the strong light-matter interactions provided by polaritonic states is an emerging research area. This approach enables modification and improvement of the electrical properties of the solar cell, e.g., through enhanced charge carrier mobility and energy transfer, without altering the chemical structure [1]. While coupling of cavity photons with electron donor type excitons has been explored [2,3], extending this concept to non-fullerene acceptors, which have been key in raising the performance of organic photovoltaic devices in recent years, remains underexplored.

In this work, we design a Fabry-Perot cavity for an organic solar cell to investigate the effects of strong light-matter coupling on charge generation and transfer in the device. Specifically, we fabricate a bulk heterojunction OPV using PM6 as the donor and Y7-12 as the non-fullerene acceptor and introduce an optical cavity structure by depositing thin Ag films onto the ITO transparent electrode. Current density-voltage (J-V) characteristics and charge generation measurements are performed to analyze device performance, and thus the influence of strong-light matter coupling phenomena.

External quantum efficiency (EQE) measurements reveal a significant redshift and a steepened EQE edge, indicating a reduction in optical bandgap energy (E_{opt}) and potentially minimized energy loss in this region [3,4]. These features, along with the apparent splitting of absorption peaks, serve as a strong indication of polariton formation [4] in the OPV cells. The influence of these effects on the photovoltaic parameters such as open circuit voltage (V_{oc}) , Fill Factor (FF), **SDU**







FIG. 1. (a) Structure of the Fabry Perot cavity OPV device used in the study. (b) Normalized EQE spectra of the Fabry Perot cavity device, and the reference device without a cavity.

and Short-Circuit Current (J_{sc}) , along with the positive impact on the internal quantum efficiency of the cells, will be presented and discussed.

These findings offer new insights into studying and optimizing OPV performance through strong light–matter coupling, paving the way for novel approaches in exciton management and energy harvesting.

 M. Hertzog et al., Strong light-matter interactions: a new direction within chemistry, Chem. Soc. Rev. 48, 937 (2019).

[2] Y. Tang et al, Strong light-matter coupling leads to a longer charge carrier lifetime in cavity organic solar cells, ACS Photonics 11, 1627 (2024).

 [3] L. M. A. de Jong et al., Enhancement of the internal quantum efficiency in strongly coupled P3HT-C60 organic photovoltaic cells using Fabry-Perot cavities with varied cavity confinement, Nanophotonics 13, 2531 (2024).

[4] V. C. Nikolis et al., Strong light-matter coupling for reduced photon energy losses in organic photovoltaics, Nat. Commun. 10, 3706 (2019).







Quantum description of plasmon polaritons in the PZW representation

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Extreme regimes of cavity quantum electrodynamics, where electronic degrees of freedom in a quantum material strongly interact with the electromagnetic modes of a cavity resonator, have been recently investigated to engineer new phases of matter. In particular, plasmonic resonators, which rely on collective electronic modes, allow achieving subwavelength confinement of the electromagnetic field, which leads to a large enhancement of the light-matter coupling. This coupling gives rise to hybrid excitations called plasmon polaritons.

Developing a quantum description of plasmon polaritons has aroused interest for the purpose of studying their coupling to quantum emitters, such as excitons in quantum confined structures. We provide a general quantum description of plasmon polaritons, allowing us to describe both localized surface plasmons, that arise, e.g., in a metallic nanosphere, and propagating surface plasmon polaritons (SPPs) at a planar interface between a dielectric and a metal. We work in the Power–Zienau–Woolley (PZW) representation, that provides a description in terms of the polarization and electric displacement fields. We show that our description of SPPs in the PZW representation is more intuitive and straightforward than the one proposed in the p \cdot A representation [1], and is free of ambiguities that existed in a recent attempt using the PZW representation [2].

F. Alpeggiani and L. C. Andreani, *Quantum theory of surface plasmon polaritons: planar and spherical geometries*, Plasmonics 9, 965 (2014).

[2] Y. Todorov, Dipolar quantum electrodynamics theory of the three-dimensional electron gas, Phys. Rev. B 89, 075115 (2014).







Strong coupling between periodic arrays of quantum emitters and periodic light

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Strong light-matter coupling encountered in cavity quantum electrodynamics has enabled controlled manipulations of quantum states at the few-excitation level, building one of the main workhorses for future quantum technologies. Periodically structured surfaces (metasurfaces), such as metamaterials or photonic crystals, on the other hand, have evolved as a powerful tool to manipulate classical electromagnetic fields. Metasurfaces were further shown to support a variety of highly tuneable, strongly confined electromagnetic modes to which nearby quantum emitters can strongly couple [1,2,3].

Here, we explore the potential of metasurfaces for cavity QED applications. Specifically, we propose a framework for strong light-matter coupling in which collective excitations of quantum emitter arrays are strongly coupled to a metasurface [4], see Fig. 1. We derive a cavity QED Hamiltonian for such systems that allows for the efficient application of standard theoretical tools. Our framework accounts for general lattice and nanoparticle geometries, as well as dispersion, absorption, and retardation effects. The wide range of possible geometrical parameters and material properties of the metasurface makes this a highly tuneable strong-coupling quantum optics platform, which we demonstrate at the example of quantum light generation: entangled photon pairs can be generated that are entangled in the spatial and/or frequency domain and that are directionally emitted.

[1] T. T. H. Do et al., Room-temperature strong coupling in a single-photon emitter-metasurface system, Nat. Commun. 15, 2281 (2024).

[2] M. Ramezani et al., *Plasmon-exciton-polariton lasing*, Optica 4, 31 (2017). SDU 🏠 UNIVERSITY OF SOUTHERN DENMARK







FIG. 1. Setup of the proposed cavity QED scheme. An emitter array is strongly coupled to a periodically structured metasurface. This results in polariton formation that can be used, e.g., for quantum optical applications.

[3] T. K. Hakala et al., Bose-Einstein condensation in a plasmonic lattice, Nat. Phys. 14, 739 (2018).

[4] F. Lindel et al., Strong coupling between periodic arrays of quantum emitters and metasurfaces, in preparation (2025).






Exploring the multiple scattering of organic exciton-polaritons using interferometric imaging

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Presentation type: Poster

Molecular exciton-polaritons, having a dual photonic and material nature, are not localized like excited molecular states – they are instead delocalized over length-scales comparable to the optical wavelength [1-3]. This delocalized nature of polaritons has major implications to the design of organic solar cells, which are largely limited in their efficiency by the mobility of the generated excitons. Additionally, this partially excitonic nature of polaritons indicates that they can interact with each other, enabling the design of photonic analogous of electronic components.

While the delocalized and coherent nature of polaritons was demonstrated more than a decade ago [1], recent experimental and theoretical works [3,4] have revealed that polaritons have intricate propagation dynamics, undergoing a transition from ballistic propagation —movement at a constant velocity— to a diffusive type of movement, dominated by multiple scattering.



FIG. 1. (a) Schematic of the optical setup for interferometric imaging of polaritons, or 'INTERPOL' for short. (b) Demonstration of interferometric imaging capabilities. Top panel shows molecular (TDBC) emission, middle shows the propagation of polaritons, and the bottom panel is an image of polariton propagation with interference fringes introduced.







In this work, we further examine the delocalized nature of polaritons and their long-range coherence properties, in order to understand the importance of incoherent (phase-destructive) scattering vs. coherent (phase-preserving) scattering in the two transport regimes. In our studies, we employ wide-field interferometric imaging (Fig. 1) to measure the mutual coherence of polariton emission at distant points along the polariton propagation and for varying photonic weights. Interestingly, we find that not only does the fringe visibility depend on the separation between the measured points, but also on where along the propagation they are located. By providing information on the loss of coherence, such experiments can shed further light on the nature of the scattering processes involved in polaritonic transport and on the transition between ballistic motion and diffusion.

[1] S. Aberra Guebrou et al., Coherent emission from a disordered organic semiconductor induced by strong coupling with surface plasmons, Phys. Rev. Lett. 108, 066401 (2012).

[2] S. Hou et al., Ultralong-range energy transport in a disordered organic semiconductor at room temperature via coherent exciton-polariton propagation, Adv. Mater. 32, 2002127 (2020).

[3] M. Balasubrahmaniyam et al., From enhanced diffusion to ultrafast ballistic motion of hybrid light-matter excitations, Nat. Mater. 22, 338 (2023).

[4] I. Sokolovskii et al., Multi-scale molecular dynamics simulations of enhanced energy transfer in organic molecules under strong coupling, Nat. Commun. 14, 6613 (2023).







Confining excitons in semiconducting moiré superlattices

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Excitons are widely found in semiconductors and play an important role in their electrical and optical properties. More specifically, excitons in 2D semiconducting transition metal dichalcogenides (TMDs) exhibit large binding energies, excellent tunability, and compatibility with various photonic platforms [1]. Confining excitons enables the creation of single-photon emitters, which are crucial for studying cavity quantum electrodynamics and optical quantum technologies [2]. Here, we report the exciton localization from cryogenic temperature [3] to room temperature [4] using moiré superlattices (see Fig. 1). The remarkable impact of the moiré effect on excitons is attributed



FIG. 1. A schematic illumination of excitons trapped in a moiré superlattice. The moiré superlattice is created by stacking two different TMD monolayers, leading to the formation of moiré potential that captures the excitons generated by optical excitations. The depth and periodicity of the potential trap can be modulated by the twist angle.

to atomic reconstruction, which is visualized using an atomic force microscope. Such moiré-trapped excitons are predicted to form arrays of indistinguishable single-photon emitters [5]. Their strong







interaction with an optical cavity can lead to exciton-polaritons [6], opening new opportunities for exploring cavity quantum electrodynamics.

[1] F. Xia et al., Two-dimensional material nanophotonics, Nat. Photon. 8, 899 (2014).

[2] M. Esmann et al., Solid-state single-photon sources: recent advances for novel quantum materials,

Adv. Funct. Mater. 34, 2315936 (2024).

 [3] H. Fang et al., Localization and interaction of interlayer excitons in MoSe₂/WSe₂ heterobilayers, Nat. Commun. 14, 6910 (2023).

 [4] Q. Lin et al., Moiré-engineered light-matter interactions in MoS₂/WSe₂ heterobilayers at room temperature, Nat. Commun. 15, 8762 (2024).

[5] H. Yu et al., Moiré excitons: from programmable quantum emitter arrays to spin-orbit-coupled artificial lattices, Sci. Adv. 3, e1701696 (2017).

[6] L. Zhang et al., Van der Waals heterostructure polaritons with moiré-induced nonlinearity, Nature 591, 61 (2021).







The origin of delayed electroluminescence in polariton light emitting diodes

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The field of strong light-matter coupling has become an area of significant search for chemistry and organic optoelectronics research due to its large range of interesting applications such as modification of chemical reactions, enhanced rates of energy transfer and Bose–Einstein condensates. In recent years, there have been multiple studies on harvesting triplet excitations through reverse intersystem crossing (RISC) and triplet-triplet annihilation (TTA) [1]. However, the comprehensive understanding and effectiveness of such processes are still debatable. Furthermore, these systems have been studied only under optical excitations where delayed photoluminescence (PL) identifies the presence of such processes. In this study, we fabricated a simple polariton organic light emitting diode (OLED) to study the effect of strong coupling on RISC and TTA through time-resolved electroluminescence. Our polariton OLED devices consists of a single emitting layer of 2,7-Bis[9,9-di(4methylphenyl)-fluoren-2-yl]-9,9-di(4-methylphenyl) fluorene (TDAF) sandwiched between a lithium fluoride (LiF)/aluminum-mirror anode and a molybdenum oxide (MoO₃)/aluminum-mirror cathode. Unlike previous polariton studies on thermally-activated and phosphorescent emitters, TDAF is primarily a fluorescent material with large energy gap between S1 and T1 of $\sim 0.8 \,\mathrm{eV}$ which is essential in this study as it hinders RISC from T1 to S1 within the molecule and would allow us to observe drastic effects of polariton in the dynamics of the system. To collect the time resolved electroluminescence, we excite our samples using square electrical pulses with rise and fall times of sub-9 ns and collected the signal using a custom-built k-space and time-correlated single photon counting (TCSPC) spectroscopy setup. We supplemented our experiments by incorporating theoretical fits from the coupled rate equations model. The model considers all significant mechanisms leading to the delayed electroluminescence in organic emitters. After a comprehensive analysis, our







results indicate that delayed electroluminescence in our devices was primarily driven by emission from trapped charges and the mechanism remains unaffected in the presence of strong coupling [2,3].

 R. Bhuyan et al., The rise and current status of polaritonic photochemistry and photophysics, Chem. Rev. 123, 10877 (2023).

[2] A. G. Abdelmagid et al., *Identifying the origin of delayed electroluminescence in a polariton organic light-emitting diode*, Nanophotonics 13, 2565 (2024).

[3] T. Leppälä et al., Linear optical properties of organic microcavity polaritons with non-Markovian quantum state diffusion, Nanophotonics 13, 2479 (2024).







Towards nonlinear spectroscopic studies of organic molecules under vibrational strong coupling

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Polariton chemistry is an emerging route to manipulate chemical reactions [1]. However the underlying fundamental mechanisms of how a reaction coordinate is altered by the coupling of molecular vibrations with a vacuum field are still to be understood. Applying ultrafast coherent multidimensional (2DIR) spectroscopy to study strong coupling of molecular systems could provide insights to how the energy distribution changes when these delocalized hybrid states are formed [2,3]. Experimental investigation of the vibrational dynamics can provide insights into the fundamental mechanisms of how vibropolaritons might modulate chemical reactions.



FIG. 1. Steady state IR spectrum for Benzaldehyde in a cavity as a function of angle of incidence

Here we report on our (a) cavity design and characterization, (b) static polariton spectra supported by theory and (c) first attempts in performing nonlinear IR spectra of organic compounds,





like Benzaldehyde (C=O), under VSC in cavities. We aim to investigate vibrational lifetimes and energy transfer processes and examine how these depend on cavity and molecular properties.

[1] A. Thomas et al., *Tilting a ground-state reactivity landscape by vibrational strong coupling*, Science 363 615 (2019).

[2] R. Duan et al., Isolating polaritonic 2D-IR transmission spectra, J. Phys. Chem. Lett. 12, 11406 (2021).

[3] B. Xiang et al., Intermolecular vibrational energy transfer enabled by microcavity strong lightmatter coupling, Science 368, 665 (2020).







Stark control of plexcitonic states in incoherent quantum systems

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Active control of quantum states in incoherent quantum system is crucial for in situ programmable and multifunctional photonic integrated circuits (PICs) [1]. Quantum plasmonics provides an efficient pathway to achieve this control by leveraging the quantum properties of intense plasmon modes and the excitonic states of quantum emitters (QEs). The coherent interaction between a plasmonic cavity and an exciton generates hybrid quantum states, known as plexcitonic states. Tuning these states in a resonantly coupled system by varying the interaction strength (f) from the weak to strong coupling regimes reveals two distinct phenomenon: Fano resonance (FR) and vacuum Rabi splitting (RS). Both mechanisms enable coherent oscillations and facilitates the quantum superposition of states, which are critical for entanglement and quantum information processing [2].



FIG. 1. (a) Hybrid quantum plasmonic system of Au bow-tie nanoantenna and voltage-tunable QE. (b) Stark-induced SIT in the off-resonant coupled system. (c) Stark-induced Rabi splitting in resonantly coupled plexcitonic states. (d) PL spectra of Stark tuned plexcitons as a function of detuning for different strengths of Stark field in the weak (f : $0.02\omega_0$) and (e) strong coupling (f : $0.05\omega_0$)

However, achieving coherent control in an off-resonant quantum system through external fields is particularly important for developing actively tunable and programmable photonic components with scalable functionalities [3]. In this study, we theoretically demonstrate the active tuning of plexcitonic modes in both (i) off-resonant and (ii) resonantly coupled plasmon-emitter systems through optical Stark effect (OSE) [4]. Using the Heisenberg-Langevin approach, we evaluate the **SDU**





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dynamics of the intense plasmonic field generated by a bow-tie nanoantenna coupled to QE. In the off-resonant systems, the Stark field shifts the degeneracy of a three-level QE and coherently drives the off-resonant plexcitonic states closer to resonance, leading to a path interference effect and the formation of a transparency window, which we refer as Stark-induced transparency (SIT). Furthermore, even small perturbations in the Stark field yield significant changes in Rabi splitting, reaching values up to $\Omega \leq 350$ meV. These pronounced resonant shifts in the excitonic levels also enables the tunable photoluminescence (PL) in the visible regime, with on/off modulation of PL intensity. In resonantly coupled systems, the Stark field lifts the degeneracy by splitting the excited state of a two-level QE, inducing path interference in the hybrid plexcitonic modes. The Stark-induced splitting also shows the signature of Mollow triplets in the plexcitonic modes, with a maximum energy splitting up to 491 meV between the upper (UP) and lower (LP) plexcitons. We validate these plexcitonic shifts through PL spectra and evaluate the optical response of the system for varying Stark field strengths (see Fig. 1). With increasing field strength, the splitting between UP and LP polaritonic states increases, while the PL intensity gradually decreases, indicating a transition of spontaneous photon emission from high (on) to low (off) as a function of probe field. Our proposed method of Stark tuning of plexcitonic modes provides a pathway for coherent control of quantum devices, offering a means to mitigate decoherence in quantum systems. Additionally, it demonstrates the active tuning of spontaneous photon emission in the visible regime, paving the way for advanced functionalities in quantum photonic applications.

 T. Giordani et al., Integrated photonics in quantum technologies, Riv. Nuovo Cim 46, 71 (2023).
P. Vasa and C. Lienau, Strong light-matter interaction in quantum emitter/metal hybrid nanostructures, ACS Photonics 5, 2 (2018).

[3] H. Asif et al., Voltage-controlled extraordinary optical transmission in the visible regime, Phys. Rev. B. 109, 125425 (2024).

[4] H. Asif and R. Sahin, Stark control of plexcitonic states in incoherent quantum system, Phys. Rev. A. 110, 023713 (2024).







Analytic model reveals local molecular polarizability changes induced by collective strong coupling

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Presentation type: Poster

Despite recent numerical evidence, one of the fundamental theoretical mysteries of polaritonic chemistry is how and if collective strong coupling can induce local changes of the electronic structure to modify chemical properties.

First, we present non-perturbative analytic results for a model system consisting of an ensemble of N harmonic molecules, see Fig. 1, under vibrational strong coupling (VSC) that alters our present understanding of this fundamental question [1]. We discover that the electronic molecular polarizabilities are modified even in the case of vanishingly small single-molecule couplings. Consequently, this non-perturbative local polarization mechanism persists even in the large-N limit. In contrast, a perturbative calculation of the polarizabilities leads to a qualitatively erroneous scaling behavior with vanishing effects in the large-N limit. Our fundamental theoretical observations demonstrate that hitherto existing collective-scaling arguments are insufficient for polaritonic chemistry/physics.

Second, we look at reaction rates inside cavities of more complex, i.e., anharmonic, molecules while allowing for non-equilibrium dynamics. When describing the cavity mediated interactions self-consistently, there could appear a non-conservative force due to the time-correlations (i.e., long-living polarization correlations) in the molecular dynamics [2]. Thus, methods based on modified transition state theories and common rare event methods (like reactive flux) that assume a canonical phase space distribution would be inadequate. Here, we use forward-flux sampling to calculate reaction rates of Shin-Metiu molecules inside a cavity. Besides consequences of the local polarization mechanism, allowing for non-canonical distributions opens the door to connect to stochastic resonance phenomena [3].









FIG. 1. Real enough to capture a collective strong coupling effect on the polarizability but simple enough for analytical solution: our model system of one-electron molecules.

[1] J. Horak et al., Analytic model reveals local molecular polarizability changes induced by collective strong coupling in optical cavities, arXiv:2401.16374 (2024).

[2] D. Sidler et al., Unraveling a cavity-induced molecular polarization mechanism from collective vibrational strong coupling, J. Phys. Chem. Lett. 15, 5208 (2024).

[3] D. Sidler et al., A perspective on ab initio modeling of polaritonic chemistry: the role of nonequilibrium effects and quantum collectivity, J. Chem. Phys. 156, 230901 (2022).







Quantisation of electromagnetic field near dispersive resonators

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Strong coupling between plasmons and quantum emitters has been the subject of recent experimental study, promising applications in quantum technologies and polaritonic chemistry. Theoretical understanding, however, is hindered by the open nature of such systems as well as material dispersion and losses. As a step towards rigorous theory of quantum plasmonics, we present a modes-of-the-universe approach to quantising electromagnetic fields in the presence of dispersive, lossless resonators.



FIG. 1. (a) The system considered consists of an arbitrarily shaped dielectric surrounded by perfectly conducting walls. The dielectric medium is described by the Hopfield model, as an ensemble of uncoupled harmonic oscillators, each with a dipole moment proportional to its displacement. (b) The corresponding frequency dependent relative electric permittivity exhibits a resonance at the natural frequency of the oscillators.

Collective oscillations of conducting electrons within metallic nanostructures—localised surface plasmon polaritons—are associated with strong electromagnetic near-fields, enabling localisation of light well below the diffraction limit. There has been much interest in exploiting this effect to achieve strong coupling of light with quantum emitters [1], motivated by the possibility of observing quantum effects at room temperature [2] and applications in polaritonic chemistry.







Due to material dispersion and Ohmic losses associated with metals and the radiative nature of open resonators, existing approaches to the problem are often phenomenological, *e.g.*, cavity QED type models. However, recently a new scheme has been proposed [3], utilising contour integration to capture the response in terms of a discrete set of damped pseudomodes. Here, we extend this method to dispersive photonic environments.

We present a macroscopic quantum description of the electromagnetic field in the presence of an arbitrarily shaped Hopfield-type dielectric [4,5] (see Fig. 1 for detailed description of the system). We employ the Lagrangian

$$L = \int_{V_1} d^3 \mathbf{r} \left(\frac{\varepsilon_0}{2} \mathbf{E}^2 - \frac{1}{2\mu_0} \mathbf{B}^2 + \frac{\rho}{2} (\partial_t \mathbf{X})^2 - \frac{\rho \omega_X^2}{2} \mathbf{X}^2 - \alpha \left(\mathbf{A} \cdot \partial_t \mathbf{X} + U \nabla \cdot \mathbf{X} \right) \right) + \int_{V_2} d^3 \mathbf{r} \left(\frac{\varepsilon_0}{2} \mathbf{E}^2 - \frac{1}{2\mu_0} \mathbf{B}^2 \right) + \oint_S d^2 S \,\alpha U \mathbf{n} \cdot \mathbf{X},$$
(1)

where **E** and **B** are the electric and magnetic fields, U and **A** are the scalar and vector potentials, **X** is the displacement field of the harmonic oscillators used to model the medium, ρ and α are the mass and negative charge densities associated with the oscillators and ω_X is their natural frequency.

The pseudomode approach [3] is based on normal mode expansion, with the (non-dispersive) resonator enclosed by a perfectly conducting boundary, that is eventually taken to infinity. From the description in terms of a continuum of quantised normal modes, a new picture involving a discrete set of quantised decaying pseudomodes is obtained, providing a convenient description of the effects of the photonic environment. Here, we present an extension of the first part of this procedure, a complete and rigorous modes-of-the-universe quantisation scheme for dispersive and open photonic systems.

[1] P. Törmä and W. L. Barnes, Strong coupling between surface plasmon polaritons and emitters: a review, Rep. Prog. Phys, 78, 013901 (2015).

[2] R. Chikkaraddy et al., Single-molecule strong coupling at room temperature in plasmonic nanocavities, Nature 535, 126 (2016).

[3] B. Yuen and A. Demetriadou, *Exact quantum electrodynamics of radiative photonic environ*ments, Phys. Rev. Lett. 133, 203604 (2024).

[4] J. J. Hopfield, Theory of the contribution of excitons to the complex dielectric constant of crystals, Phys. Rev. 112, 1555 (1958).

[5] B. Huttner and S. M. Barnett, Quantization of the electromagnetic field in dielectrics, Phys. Rev. A 46, 4306 (1992).







Phase topology and group index of THz vibro-polaritons in metasurfaces

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To investigate strong light-matter coupling in optical cavities, current experimental studies mainly focus on intensity information in the spectra using different techniques, such as absorption spectroscopy [1], photoluminescence spectroscopy [2], and Raman spectroscopy [3]. Intensity spectra can directly reflect information on energy levels of polaritons and retrieve the Rabi splitting. However, optical interference and absorption of the upper polariton often complicate the analysis. In this respect, studies on the behavior of phase characteristics of electromagnetic wave propagating through or scattering with strongly coupled systems remain comparatively limited [4]. Terahertz time-domain spectroscopy (THz-TDS) is a promising tool due to its ability to provide amplitude (intensity) and phase information simultaneously by measuring the THz electric field in the time domain.

In this work, we use THz-TDS to investigate the strong coupling of intermolecular vibrations with optical modes of metasurfaces. By exploiting the amplitude and phase information of broadband THz pulses transmitted through the samples, we observe the formation of vibro-polaritons and the change in phase topology when the samples are in the strong light-matter coupling regime: A loop is formed in the transmittance complex plane with a length proportional to the Rabi splitting. The group delay and group index of the THz wave propagating through the samples can be retrieved from the frequency-dependent phase of the transmission. The hybridization of the optical mode with the molecular vibrations leads to the reduction of the group delay and the absolute value of the group index, which has similar values for the upper and lower vibro-polaritons. These results reveal the significance of the phase information provided by THz-TDS in the description of light-matter









FIG. 1. (a) Illustration of the THz-TDS transmission measurement on the coupled system. (b) Measured upper and lower vibro-polariton energies as a function of the lateral separation between the rods in one unit cell, which tunes the metasurface resonances. The horizontal dotted line indicates the intermolecular vibration frequency of α -lactose, and the solid black line represents the simulated resonance of the metasurface covered with a 10 μ m-thick layer with a dielectric constant $\epsilon = 2.5$ representing the α -lactose film in the absence of intermolecular vibrations. (c) Imaginary component of the complex transmission as a function of the real component for the metasurfaces with rods with tuned lateral separations. (d) Simulated (dashed curves) and measured (solid curves) frequency-dependent group index n_g of the investigated systems under the effect of strong coupling and no coupling. The vertical lines indicate the frequencies of the intermolecular vibration of α -lactose and the low-frequency mode of the non-coupled metasurface.

[1] G. Zengin et al., Evaluating conditions for strong coupling between nanoparticle plasmons and organic dyes using scattering and absorption spectroscopy, J. Phys. Chem. C 120, 20588 (2016).

[2] K. D. Park et al., *Tip-enhanced strong coupling spectroscopy, imaging, and control of a single quantum emitter*, Sci. Adv. 5, eaav5931 (2019).

[3] A. Shalabney et al., Enhanced Raman scattering from vibro=polariton hybrid states, Ang. Chem.
Intl. Ed. 54, 7971 (2015).

[4] P. A. Thomas et al., A new signature for strong light-matter coupling using spectroscopic ellipsometry, Nano Lett. 20, 6412 (2020).







Surface exciton polaritons from visible to the near-infrared supported by nolecular materials

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The field of polaritonics has seen significant advancements in the past decades due to the utilization of plasmonic materials like gold and silver [1] in areas such as biosensing, energy and information technologies [2]. On the other hand, 2D materials such as transition metal dichalcogenide monolayers (TMDCs) are providing a novel platform to create and manipulate surface polaritons at cryogenic temperatures [3].

In this work, we bring an alternative by venturing into the realm of organic polaritonics by investigating surface exciton polaritons (SEPs) in molecular materials at room temperature. This alternative is based on carbocyanines which are well known for their chemical stability, water solubility and the large library available with controllable response from the visible to the telecom wavelengths [4]. These molecules have electronic properties that make them ideal for self-assembling into polycrystalline supramolecular structures that promote in-lattice delocalization of Frenkel excitons which bring them with strong sharp excitonic resonances [5]. The presence of these strong resonances is responsible for reaching negative values in the real part of the permittivity, supporting SEPs at the material's interface.

However, the control of conformation of supramolecular structures for the growth of solid films with optical quality is a challenging task. We solved this problem by following a Layerby-Layer (LbL) assembly [6] on flat substrates. First, due to unique electronic properties of cyanine monomers, we were able to stabilize and control the dispersity of the J-aggregates by tuning the chemical properties of water as a solvent and stirring over a large period, respectively. Later, we deposited the self-assembled J-aggregates by dipping alternately a functionalized substrate in the Jaggregate solution and in an aqueous polymer with positive charge – poly(diallyldimethylammonium







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chloride) (PDDA). Taking advantage of the negatively charged functional groups of cyanines, their corresponding J-aggregates were consistently absorbed subsequently with the polyelectrolyte into the surface of the film, creating uniform multi-layers. Using this approach, we fabricated molecular films with narrow-band optical properties from five different J-aggregates with spectral ranges varying from 560 nm to 1000 nm with accurate control over their thickness. Then, using spectroscopic ellipsometry we estimated each of their dielectric functions, seeking for negative values in the real part. Finally, we corroborated the SEP coupling, after successfully exciting and detecting the confined SEP mode living at the interface between the J-aggregate films and the air medium [7], using advanced Fourier imaging spectroscopy combined with a typical Kretschman prism-coupling configuration.

Our work paves a new way for the creation of tailored Vis-NIR polaritonic materials with the potential for nano-structuring controlling distances between different species. Such advancements will enable the exploration of more sophisticated light–matter interaction mechanisms, opening doors for innovative polaritonic devices functionalities.

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 D. Sanvitto and S. Kéna-Cohen, The road towards polaritonic devices, Nat. Mater. 15, 1061 (2016).

[2] L. Wang et al., Optical properties and applications of plasmonic-metal nanoparticles, Adv. Funct. Mater. 30, 2005400 (2020).

[3] T. Low et al., Polaritons in layered two-dimensional materials, Nat. Mater. 16, 182 (2017).

[4] H. Shindy, Fundamentals in the chemistry of cyanine dyes: a review Dyes and pigments 145, 505 (2017).

[5] J. Bricks et al., Fluorescent J-aggregates of cyanine dyes: basic research and applications review, Meth. Appl. Fluoresc. 6, 012001 (2018).

[6] R. Iler, Multilayers of colloidal particles, J. Coll. Interf. Sci. 21, 569–594 (1966).

[7] M. Gentile et al., Optical field-enhancement and subwavelength field-confinement using excitonic nanostructures, Nano Lett. 14, 2339 (2014).







Cooperative effects in vibrational polaritons require environment-induced nonequilibrium steady states

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Presentation type: Poster

Polariton-induced modifications to reactivity have been experimentally demonstrated in various systems, including enzyme-catalyzed reactions [1] and organic reactions [2]. As an extension thereof, "cooperative" coupling mechanisms, in which a solvent molecule undergoes vibrational strong coupling, which then in turn modifies the reactivity of a reactant species, has also been proposed and demonstrated [3]. The conventional treatment of such a polaritonic system would focus on the



FIG. 1. The modifications to chemical reactivity that were demonstrated in [3] are only reproducible using nonperturbative approaches such as pseudomodes [4] and not using standard master equations such as the Lindblad equation. Remarkably, this means implies that cooperative effects cannot occur if the bath has a thermalizing effect on the system, i.e., cooperative modifications to reactivity require non-equilibrium steady







electromagnetic modes and the resonant matter modes, reducing the system to a tractable number of degrees of freedom. However, given that such systems are almost always in the condensed phase and, therefore, interact strongly with many further degrees of freedom, we investigate the effect of these additional "bath" degrees of freedom and find that the non-negligible system–bath coupling leads to significant deviations from the basic polaritonic dynamics, for example in the form of nonequilibrium (nonthermal) steady states. In particular, by comparing perturbative/Markovian approaches with non-perturbative, exact pseudomode approaches [4,5], we elucidate that the nature and modeling of the system–bath interaction "make or break" effects such as modifications to chemical reactivity.

[1] R. M. A. Vergauwe et al., *Modification of enzyme activity by vibrational strong coupling of water*, Angew. Chem. Int. Ed. 58, 15324 (2019).

[2] A. Thomas et al., Ground-state chemical reactivity under vibrational coupling to the vacuum electromagnetic field Angew. Chem. Int. Ed. 128, 11634 (2016).

[3] J. Lather et al., Cavity catalysis by cooperative vibrational strong coupling of reactant and solvent molecules, Angew. Chem. Int. Ed. 58, 10635 (2019).

[4] D. Tamascelli et al., Nonperturbative treatment of non-Markovian dynamics of open quantum systems, Phys. Rev. Lett. 120, 030402 (2018).

[5] A. Lemmer et al., A trapped-ion simulator for spin-boson models with structured environments, New J. Phys. 20, 073002 (2018).







Charge transport engineering in organic semiconductors through strong coupling in MOSFET-based Fabry-Pérot cavities

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Presentation type: Poster

Modifying material properties by strong light-matter coupling of molecular excitation with confined electromagnetic field is a groundbreaking approach offering unprecedented opportunities for designing and improving novel photonic and electronic devices. This phenomenon is intriguing, where the photon exchange rate between light and matter outpaces dissipation processes, resulting in hybrid light-matter states (called polaritons), showing characteristics of both photon and exciton. The new polaritonic states provide an additional transport channel to the flow of electrons, modifying the charge transport in semiconductors. Organic semiconductors (OSCs) are gaining much interest because of their light weight, flexibility, low-cost manufacturing, easy solution processability, etc. However, they still face challenges in improving charge transport properties and overall performance compared to inorganic semiconductors. Efforts are made to improve their performance by conventional methods like molecular design and organic synthesis. Recent breakthroughs have demonstrated that the phenomenon of light-matter strong coupling can potentially tailor the charge transport properties of the semiconductors without any physical or chemical modification [1,2].

We investigated the room temperature engineering of charge transport in OSCs using MOSFET (Metal Oxide Semiconductor Field Effect Transistor) based Fabry-Pérot cavities. This cavity configuration allows simultaneous investigation of the optical and electrical response of a thin film OSC. By utilizing the fact that the electromagnetic field can be confined by structuring the refractive index of the medium, we proposed a mirrorless Fabry-Pérot configuration (Air/OSC layer/SiO₂/Si) using MOSFET devices (Fig. 1(a)). The leaky modes are formed due to refractive index mismatch between the various layers while avoiding the use of any metallic mirror. The frequency of the leaky cavity modes is tuned by varying the thickness of the OSC film. Mobility is high at lower thicknesses due to better packing of OSC dye molecules, which becomes almost constant at higher thicknesses in the absence of coupling. With the variation of thickness, the frequency of the confined mode









FIG. 1. (a) Schematic for mirrorless MOSFET Fabry-Pérot cavity configuration. (b) Charge carrier mobility variation due to the effect of morphology and strong coupling phenomenon, shown by blue and red regions, respectively.

resonates with the molecular transition, and the system slowly enters the strong coupling regime, resulting in a significant enhancement in charge carrier mobility (Fig. 1(b)) [3]. The optical studies and TMM simulations further confirm the resonance effect. Unlike the conventional Fabry-Pérot cavities, the proposed configuration eliminates the leaking issues in electrical response caused by metallic mirrors. It provides the simplest way of tailoring the properties of functional materials. These mirrorless cavity configurations have the potential in experimental science to pave the way for fast polariton-based devices that may have direct applications in quantum sensing and communication.

 E. Orgiu et al., Conductivity in organic semiconductors hybridized with the vacuum field, Nat. Mater. 14, 1123 (2015).

[2] K. Nagarajan et al., Conductivity and photoconductivity of a p-type organic semiconductor under ultrastrong coupling, ACS Nano 14, 10219 (2020).

[3] K. Kaur et al., Controlling electron mobility of strongly coupled organic semiconductors in mirrorless cavities, Adv. Funct. Mater. 33, 2306058 (2023).







Exact numerical solution of the dissipative Tavis–Cummings model in the context of superradiance

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Presentation type: Poster

The Tavis–Cummings (TC) model [1], describing the interaction of a single cavity mode with N spins, is ubiquitous in the field of cavity QED. In order to describe real experiments, it is vital to solve this model in an open quantum systems setting, i.e. by letting the system degrees of freedom couple to an environment, which induces losses and dephasing. A standard way to incorporate such loss terms into the system dynamics is by means of a master equation for the density matrix ρ

$$\partial_t \rho = -\mathbf{i}[H,\rho] + \kappa \mathcal{L}[a] + \gamma \sum_{n=1}^N \mathcal{L}[\sigma_n^-] + \gamma_\phi \sum_{n=1}^N \mathcal{L}[\sigma_n^z] \tag{1}$$

with $\mathcal{L}[X] = X\rho X^{\dagger} - \frac{1}{2}(X^{\dagger}X\rho + \rho X^{\dagger}X)$. The rates κ , γ and γ_{ϕ} describe cavity loss, individual spin losses and individual spin dephasing, respectively. H is the TC Hamiltonian, a is the bosonic annihilation operator of the cavity mode and σ_n^- , σ_n^z are the Pauli operators for the *n*-th spin.

The main difficulty in solving Eq. (1) is the size of the Hilbert space, which grows exponentially with N. For problems with only collective loss/dephasing, one can use a collective spin representation, but this is not possible here. However, a known symmetry of the model is the permutation of pairs of spins, which reduces the exponential scaling to a polynomial scaling in N [2, 3]. In this work, we go further and identify a weak U(1) symmetry of the model for an initial state with zero photons and all spins in the excited state (fully inverted state). This reduces the scaling to N^4 allowing for numerical calculations up to $N \sim 150$ spins, which is far beyond achievable N in previous methods. If the initial state has zero photons and all spins are excited, we call it superfluorescent. In contrast, if there is some small initial symmetry-breaking in the spin states, we call the state superradiant, as there is some initial coherence in the spins.





We use the developed code to compare the exact dynamics of observables, such as the mean photon number in the cavity, to approximate dynamics obtained from a second-order cumulant expansion for different system parameters and initial conditions. We find that the deviation between exact and cumulant solution is generally lower for superradiance initial conditions compared to superfluorescence initial conditions, where in the former case, symmetry-breaking cumulant equations must be used.

 M. Tavis and F. W. Cummings, Exact solution for an N-molecule—radiation-field Hamiltonian, Phys. Rev. 170, 279 (1968).

[2] P. Kirton and J. Keeling, Suppressing and restoring the Dicke superradiance transition by dephasing and decay, Phys. Rev. Lett. 118, 123602 (2017).

[3] N. Shammah et al., Open quantum systems with local and collective incoherent processes: Efficient numerical simulations using permutational invariance, Phys. Rev. A 98, 063815 (2018).







Causality for the second quantisation of open non-Hermitian nanophotonic systems

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Recent rapid experimental developments have allowed for the coupling of quantum emitters with open nanophotonic environments that radiate efficiently to the far-field, holding significant promise towards developing new quantum and polaritonic devices. Quantising such systems is a significant challenge hindering our understanding and interpretation of current experimental results. Here, we present an exact and complete second-quantisation of open nanophotonic systems, focusing on causality and the often overlooked Markovian dynamics of such systems.



FIG. 1. (a) The set up of the system of interest, a dipole placed at some distance r and detector at r' away from the centre of a dielectric nanoparticle. We choose to enclose the system in a spherical boundary with radius sent to infinity. (b) The Quantum Dynamics of a silicon spherical particle when $r = rp = a = 1\mu m$ and r = rp = 1.1a for dipole moment d = 10 D. Note the strong Rabi oscillations and the presence of Markovian dynamics in the top figure and the lack of such features due to a resonance shift in the lower. The initial rapid decay (prior to the red dashed line) happens before causality allows the pseudomodes to take effect.

Improvements in experimental capabilities mean that quantum emitters can be coupled to increasingly complex photonic systems. The study of quantum phenomena in such systems is often done assuming closed environments (such as Fabry–Pérot cavities) or phenomenologically (cQED). However it is crucial, to obtain a full picture, connecting both the near and far field dynamics of







these systems. Due to the a lack of obvious boundary conditions nd the lossy (non-Hermitian) nature of such systems, their quantisation is challenging. Current methods to deal with these challenges usually involve approximations when solving governing integro-differential equations, but such approaches lead to the loss of important (e.g. Markovian) dynamics. Furthermore, issues such as mode divergence, a disconnect between near and far field, and difficulty converting back to natural modes cause a disconnect between the quantum and classical world. To overcome this a new method for the second quantisation of non-Hermitian open quantum systems in an *exact* and *complete* way has been proposed [1]. Here, we advanced this method for arbitrary positioning of a quantum emitter and focus on the manifestation of causality.

This new pseudomode method performs second quantisation on the normal modes of a photonic system enclosed within a geometry matching shell (Fig. 1(a)). Upon infinitely increasing the size of this boundary, we arrive at a set of coupled integro-differential equations that fully describe the (open) system's quantum-dynamics. Their solution is obtained through mapping the continuum of normal modes to a discrete basis of complex pseudomodes [1,2] that themselves are inherently non-hermitian and, by their complex nature transform integral to sum leading to a solution for the quantum-dynamics of the system:

$$\int_{0}^{\infty} k dk \rho(k) \mathbf{u}_{\xi}(k, \mathbf{r}) \mathbf{u}_{\xi}(k, \mathbf{r}') e^{-ick\tau} = \sum_{n} z_{\xi n} \mathbf{v}_{\xi, n}(\mathbf{r}) \mathbf{v}_{\xi, n}(\mathbf{r}') \Theta(\tau - \Delta(\mathbf{r}, \mathbf{r}')) e^{-icz_{\xi n}\tau}$$
(1)

It is within the step-function of the pseudomode that causality is manifested. Through consideration of functional convergence in the complex plane it can be demonstrated that Θ will take the form of the spacetime interval as causality dictates. Fig. 1(b) demonstrates dynamics that show such an effect with natural decay until light has had chance to reach the detector, at which point the pseudomodes dynamics take precedence. Exact calculation of these terms can be used in the renormalisation of the pseudomode expansion allowing the study of the connection between near and far field dynamics.

In conclusion, we present a novel method for the exact and complete quantisation of open non-Hermitian nanophotonic systems and demonstrate the natural emergence of causality and the rise of Markovian dynamics that are often overlooked.

[1] B. Yuen and A. Demetriadou, *Exact quantum electrodynamics of radiative photonic environments*, Phys. Rev. Lett. 6, 203604 (2025).

[2] B. M. Garraway, Nonperturbative decay of an atomic system in a cavity Phys. Rev. A 55, 2290 (1997).







Surface and cavity exciton-polaritons in J-aggregate films

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Presentation type: Poster

Surface polaritons, hybrid excitations arising from the strong coupling between electromagnetic waves and collective material excitations, are a central topic in the study of light-matter interactions. These evanescent waves can confine intense electromagnetic radiation at the sub-wavelength scale, leading to strong local field enhancements. In particular, surface exciton polaritons (SEPs) can form at the surface of materials with a strong excitonic response, and they have been experimentally observed in both inorganic and organic semiconductors [1]. However, the nature of the excitons involved determines the temperature range where SEPs can exist. Inorganic crystals host Wannier–Mott excitons with relatively small binding energies, which only exist at low temperatures, in contrast with organic materials hosting robust Frenkel excitons observed at room temperature [2]. Thus, molecular excitonic materials are a promising alternative to conventional inorganic systems for practical polaritonic applications. Beyond surface modes, exciton-polaritons can also be confined within closed (e.g. Fabry-Pérot, FP) optical cavities, where strong light-matter coupling with cavity electromagnetic modes enables the tailoring of polariton dispersion properties.

Among organic materials, J-aggregates of organic dyes stand out due to their strong excitonic response, exhibiting narrow absorption and emission bands [3]. However, accurately modeling their optical properties remains challenging to this day due to orientation and size disorder of the oligomers composing a J-aggregate layer, possible anisotropy, and interfacial effects such as roughness.

In this work, we investigate surface and cavity exciton-polaritons in J-aggregate films while accounting for some of these complexities. We model their optical response by considering Jaggregates as either 3D-isotropic or composed of 2D sublayers with a uniaxial dielectric response, both resulting from randomly oriented oligomers. We explore the effect of the dielectric response anisotropy and investigate the impact of interface roughness on the SEP dispersion and reflectance spectra, aimed at the most accurate model describing the optical behavior of J-aggregates for photonic applications.









FIG. 1. (Left): Schematic representation of a Fabry-Pérot cavity formed by two parallel mirrors at z = 0and z = L. A J-aggregate film with permittivity $\varepsilon_{\rm J}$ is positioned adjacent to one of the mirrors. The remaining cavity space is filled with a dielectric medium of permittivity ε_1 . (Right panel): P-polarisation dispersion curves calculated for the system displayed in the left panel. Dotted lines are empty cavity modes.

Embedding a J-aggregate layer in a FP optical cavity (Fig. 1) results in a rich dispersion relation containing, beyond the usually considered anti-crossing upper and lower polariton modes [4] (green and dark blue curves in Fig. 1), a guided wave (red curve) and an almost dispersion-less mode following the J-aggregate exciton energy. The latter may look puzzling at a first sight, but we verified its correctness by calculating the field profiles across the structure. Interestingly, such a dispersion-less mode, not seen in the reflectivity spectra, was observed in photoluminescence spectra along with the "conventional" emission from the lower polariton branch, only for cavities with organic emitters [5], and its physical nature has not been completely clarified.

 M. R. Philpott et al., A new optical phenomenon: exciton surface polaritons at room temperature, Mol. Cryst. Liq. Cryst. 50, 139 (1979).

[2] K. Takatori et al., Surface exciton polaritons supported by a J-aggregate-dye/air interface at room temperature, Opt. Lett. 14, 3876 (2017).

[3] J. L. Bricks et al., *Fluorescent J-aggregates of cyanine dyes: basic research and applications review*, Methods Appl. Fluoresc. 6, 012001 (2017).

[4] V. M. Agranovich et al., *Cavity polaritons in microcavities containing disordered organic semi*conductors, Phys. Rev. B 67, 085311 (2003).

 [5] J. Bellessa et al., Strong coupling between surface plasmons and excitons in an organic semiconductor, Phys. Rev. Lett. 93, 036404 (2004).







Collective single-photon emission mediated by surface plasmon polaritons and disturbed by phonons

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It is well known how the dielectric environment of a quantum emitter affects its spontaneousemission rate, but collective emission is affected differently. Here we present how the nanophotonic environment of two identical quantum emitters affects their collective emission of a single shared photon.

First, we consider a slab of hexagonal boton nitride with two quantum emitters at the top surface [1]. We compare a slab in air (see Fig. 1a) with a slab resting on a silver substrate (see Fig. 1b). In the latter case, emission rates and inter-emitter interactions depend strongly on the surface plasmon polaritons (SPP) of the multi-layer geometry. The super- and subradiant decay rates vary in an oscillatory fashion as a function of their horizontal distance ρ , see Fig. 1c. While the non-plasmonic geometries show progressively more strongly damped decay rates as a function of distance, the plasmonic geometries show smaller maximal ampitudes near $\rho/\lambda = 0.3$ than at $\rho/\lambda = 0.6$. On the poster, we will explain this as destructive interference between surface-plasmon mediated and non-SPP-mediated inter-emitter interactions. Similar destructive interference also occurs in the cross spectral density. We conclude that sometimes the SPP mode interferes destructively with the non-guided part of the inter-emitter interaction, on a distance that is important for collective emission in layered geometries.

Besides the influence of the complex nanophotonic environment on single-photon superradiance by two solid-state quantum emitters, we also study the effect of phonon interactions on collective emission [2]. Phonons affect quantum emitters in two ways: polaron formation after optical excitation is described by ultrafast non-Markovian dynamics, while slower dephasing is well described by exponential decay. The disturbing effect of the former on the collective emission and the entanglement decay of emitters is usually not modeled, and also the latter is sometimes neglected. Here









FIG. 1. (a) hBN slab in air, (b) same hBN slab resting on a silver substrate, (c) Super- (+, solid lines) and subradiant (-, dashed lines) collective decay rates γ^{\pm} , scaled by the total single-emitter decay rate γ_{Tot} for two quantum emitters with dipoles both pointing in the *x*-direction. Two-emitter decay rates are shown as a function of their horizontal distance ρ , the latter scaled by the emission wavelength λ . Blue lines correspond to the slab waveguide (WG) configuration of panel (a), red lines to panel (b) when neglecting loss in the silver. Black lines result when not neglecting losses in silver. For comparison, the grey lines depict the collective emission rates in homogeneous hBN. Figure adapted from Ref. [1].

we compare two methods that are efficient also for several emitters: the first method concatenates the fast and slow phonon dynamics, and the second is the polaron method. For a linear array of two or more quantum dots in GaAs, we study both the spectra and the entanglement dynamics corresponding to the single-photon decay. The concatenation method is numerically the most efficient of the two, taking phonon effects into account at the same computational cost as modeling only the emitter-photon interaction in the standard optical master equation [2].

 M. A. Jørgensen et al., Collective single-photon emission and energy transfer in thin-layer dielectric and plasmonic systems, Nanophotonics, in press (2025); DOI: 10.1515/nanoph-2024-0524.
D. Pandey and M. Wubs, Collective photon emission in solid state environments: Concatenating non-Markovian and Markovian dynamics, Phys. Rev. Res. 6, 033044 (2024).







Nonlinear semiclassical spectroscopy of ultrafast molecular polariton dynamics

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We introduce a theoretical framework that allows for the systematic and efficient description of the ultrafast nonlinear response of molecular polaritons, i.e., hybrid light-matter states, in the collective regime of large numbers of molecules \mathcal{N} coupled to the cavity photon mode [1]. Our approach is based on a semiclassical, mean-field evolution of the molecular Hamiltonian and the cavity field, which is complemented by a perturbative expansion of both light and matter counterparts in the input fields entering the cavity. In addition, expansion in terms of the pulse phases enables us to disentangle different excitation pathways in Liouville space, thereby distinguishing contributions to the nonlinear response. The formalism extends traditional free-space nonlinear spectroscopy by incorporating the feedback of matter onto the light field via the induced polarization. We demonstrate the utility of the framework by applying it to the calculation of pump-probe polariton spectra (see sketch in Fig. 1) and show how, by storing the pulses, the cavity facilitates additional excitation pathways, which can be used to isolate purely bright state contributions. Further, we illustrate applications in describing polariton transport by extending the formalism to multimode



FIG. 1. Sketch of an optical cavity which is driven by pump and probe input pulses.





cavities [2] as well as in multidimensional polariton spectroscopy. Our method, which does not scale with \mathcal{N} , is broadly applicable and can be extended to model a wide range of current experiments investigating the dynamical nonlinear response of hybrid light–matter states.

[1] M. Reitz et al., Nonlinear semiclassical spectroscopy of ultrafast molecular polariton dynamics, arXiv:2410.16630 (2024).

[2] P. Fowler-Wright et al., *Mapping molecular polariton transport via pump-probe microscopy*, in preparation (2025).







Chiral light–matter dynamics with thermal magnetoplasmons in graphene nanodisks

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Chiral photonic structures are essential within fields such as quantum information and communication processing, as well as within the detection of chiral molecules like DNA in biochemistry. The strong and tunable optical response of graphene presents unique opportunities to induce significant nonlinear phenomena and chiral light-matter interactions through its plasmonic resonances and near-field interactions with nearby quantum light emitters [1,2]. We propose the formation of self-hybridized thermal magnetoplasmons in doped graphene nanodisks at finite temperatures when subjected to an external static magnetic field [2]. The graphene nanodisks, as schematically shown in Fig. 1(a) support thermal magnetoplasmons that arise from the hybridization between localized magnetoplasmon resonances and thermally allowed Landau level (LL) transitions in graphene. The interband LL transitions are shown in Fig. 1(b), along with the extinction cross section for doped graphene nanodisks at different temperatures when varying the photon energy and magnetic field amplitude. The thermal magnetoplasmons are seen in the anti-crossing between the magnetoplasmonic resonances and the LL transitions at larger temperatures, where the elevated temperature ensures a population difference in the LLs which results in the LL transitions not being Pauli-blocked. These modes combine the exceptional magneto-optical response of graphene with the strong field enhancement of plasmons, making them excellent tools for achieving large chiral light-matter interactions, which are tunable through the carrier concentration, magnetic field, and temperature of the graphene nanodisks. By incorporating the graphene nanodisks into a periodic array used in a Salisbury screen, we demonstrate how such stratified structures can achieve perfect chiral absorption and tunable chiral thermal emission. The results presented in this work not only deepen our understanding of the optical properties of graphene nanostructures, but may also be used for future advanced photonic devices and technologies.









FIG. 1. (a) Schematic of a graphene nanodisk with diameter D under an external magnetic field and with incident left-hand circularly polarized (LCP) and right-hand circularly polarized (RCP) plane wave light. (b) Extinction cross section of a graphene nanodisk at temperatures T = 300 K and T = 600 K while varying the magnetic field amplitude and frequency of light. The diameter of the disk is D = 120 nm and the Fermi energy is $E_{\rm F} = 0.2$ eV.

 M. H. Eriksen et al., Optoelectronic control of atomic bistability with graphene, Phys. Rev. Lett. 129, 253602 (2022).

[2] M. H. Eriksen et al., Chiral light-matter interactions with thermal magnetoplasmons in graphene nanodisks, Nano Lett. 25, 313 (2024).







The nature of excited states relaxation in plexcitonic materials

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Strong light-matter coupling tailors the properties of the molecules by coupling them strongly to light. This phenomenon generates an upper and a lower polariton (UP and LP, respectively), and a bunch of dark states (DS). To occur, strong coupling requires "cavities", materials that amplify light and electromagnetic fields. The hybrid light-molecule properties of strongly coupled materials are appealing for several technological fields such as optoelectronics and quantum technologies. However, it is still not clear how the structure of the cavities influences the overall properties, starting from the energy relaxation of their excited states.



FIG. 1. (a) The VAS mechanism. The relaxation from the UP or DS is mediated through the activation of vibration with energy $\hbar\nu$. (b) Design of a colloidal plexcitonic material composed of a plasmonic nanoparticle, a stabilizing capping layer, and dye molecules. (c) The SERS mechanism. The light scatters from the UP or DS, activating a vibration of the ground state with energy $\hbar\nu$.

Most of our knowledge about relaxation comes from microcavities-based strongly coupled materials, where vibrational-assisted scattering (VAS) is one of the main responsible phenomena (Fig. 1(a)). Similarly to microcavities, strong coupling can occur employing plasmonic nanoparticles as cavities, resulting in the so-called plexcitonic materials (Fig. 1(b)). However, much less is known about plasmonic cavities. On the one hand, their relaxation could be demanded by VAS, as for **SDU**







microcavities. On the other, plasmonic effects must be considered including the surface-enhanced Raman scattering (SERS, Fig. 1(c)).

In this contribution, I will present colloidal plexcitonic materials prepared with nanoparticles and I will compare them with microcavity-based analogous. The study of their emission allowed the comparison between microcavity-based and nanoparticle-based relaxation phenomena. This inspection will reveal that the nature of the emission (plasmonic or microcavity-like) mainly depends on the excited state [1].

[1] Peruffo et al., The role of vibrational-assisted scattering and surface-enhanced Raman scattering in colloidal plexcitonic materials, ACS Nano, under revision (2025).






Mesoscopic phenomena and fabrication imperfections in plasmonic nanolens: impact on chiral light–matter interactions

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We theoretically examine the optical response of a trimeric self-similar sodium plasmonic nanolens under linearly and circularly polarized light. In particular, we explore the impact of nanofabrication imperfections and mesoscopic phenomena, such as electron spill-out, showing that near-field enhancement is not suppressed by quantum corrections. Furthermore, it is emphasized that attention should be paid in the chiroptical response of the system due to the influence of nonlocality, spill-out and Landau damping on gap plasmons.



FIG. 1. Near field enhancement in a perturbed self-similar plasmonic nanolens.

One of the most promising ways of enhancing light-matter interactions is through self-similar plasmonic nanolenses made of three spheres of decreasing sizes and separations [1]. However, this plasmonic trimer is expected to be influenced by nonlocality and quantum effects, such as electron density spill-out and Landau damping, leading to modification in its far- and near-field optical response [2,3]. Here we study a nanochain consisting of three sodium nanospheres (see Fig. 1), to focus on electron spill-out, using different nonlocal models to describe the optical response **SDU**





of a statistical collection of such nanosystems. Through a statistical analysis of thousands of trimer arrangements, we demonstrate that inevitable deviations from the perfect nanolens do not impede plasmonic enhancement in the nanocavities. Moreover, we show that the planar chirality of imperfect nanolenses, originating from nanoparticle displacements, together with the incident light, forms a chiral system that triggers strong chiral light–matter interactions. Finally, we highlight the chiral nature of the scattered fields under the influence of electron spill-out, allowing control over optical chirality density and circular dichroism by tuning the planar chirality of the system.

Our work brings to light the significance of the nanolensing effect in chiral light-matter interactions and suggests that imperfect plasmonic trimers can be exploited to induce considerable circular dichroism while simultaneously control the optical chirality density [4]. Nonetheless, we stress the crucial role of mesoscopic physics in the chiroptical response as ignoring quantum corrections can lead to a misleading quantification of quantities.

 K. Li et al., Self-similar chain of metal nanospheres as an efficient nanolens, Phys. Rev. Lett. 91, 227402 (2003).

[2] N. A. Mortensen, Mesoscopic electrodynamics at metal surfaces, Nanophotonics 10, 2563 (2021).
[3] C. Tserkezis et al., Circular dichroism in nanoparticle helices as a template for assessing quantum-informed models in plasmonics ACS Photonics 5, 5017 (2018).

[4] N. Kyvelos et al., Self-similar plasmonic nanolenses: mesoscopic ensemble averaging and chiral light-matter interactions J. Phys. Chem. C 129, 3635 (2025).







Lifshitz–Lorentz theory for cavity-modified ground state of a polaritonic system

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In recent years, vacuum-induced modifications of molecular properties in a "dark" cavity have attracted considerable attention due to the development of polaritonic chemistry. It is claimed that under ultrastrong coupling of matter to individual cavity eigenmodes, even without external pumping, the electromagnetic (EM) vacuum can modify a range of material properties [1]. The first theoretical models [2] used the single-mode cavity QED approaches to calculate the ground state, while they work well only for effects of photochemistry and photophysics related to excited states. Recent theoretical works have increasingly focused on the influence of non-polaritonic effects [3] and multimode coupling [4]. However, to avoid divergencies, most of them use only a few cavity modes, ignoring the coupling to an infinite number of other vacuum modes, each containing a continuum of wave vectors. Moreover, for calculating the ground state, such approaches that directly sum the zero-point energies of polaritons work best in the single-cavity mode approximation, since taking into account multiple modes can lead to even greater error [5]. For ground state calculations, the logic of light or dark states is not applicable; instead, one must consider the coupling to an infinite set of vacuum modes with a continuum of their wave vectors. This problem has been known at least since 1947 from Bethe theory for the Lamb shift or from the famous work by Casimir and Polder. They developed a renormalization technique, which allows getting a finite result including the infinite number of vacuum EM modes by the subtraction of the corresponding infinite free-space contribution. This method was later applied to cavity geometry by Casimir and then generalized by Lifshitz and his co-authors.

Here we present a fundamental theory for ground state modifications of an ensemble of harmonic oscillators in a dark cavity. Based on the Lifshitz theory for vacuum energy in a cavity and employing the Lorentz model for the permittivity of the oscillator system, we build a non-perturbative macroscopic QED model that accounts for an infinite number of cavity modes with a continuum of







their wave vectors. We reveal qualitative differences from the commonly used single(multiple)-mode models and demonstrate the non-resonant role of polaritons. Our theory allows for a simple inclusion of losses and temperature effects. We discuss the crucial role of temperature in large cavities where the classical Casimir limit occurs. We also provide a comparison with perturbative calculations within the Casimir-Polder theory and a method for experimental verification of the obtained results. Our theory also serves as a bridge between the polaritonic and Casimir communities.



FIG. 1. Sketch of the system consisting of ensemble of harmonic oscillators in a PEC cavity (left) and the ground-state energy change due to the coupling of oscillators to the vacuum EM modes, calculated using our Lifshitz-Lorentz theory and a single-mode Hopfield-like model (right). This is one of the examples showing the qualitative differences between single-mode approximation and exact infinite-mode model.

[1] F. J. García-Vidal et al., *Manipulating matter by strong coupling to vacuum fields*, Science 373, eabd0336 (2021).

[2] C. Ciuti et al., Quantum vacuum properties of the intersubband cavity polariton field, Phys. Rev. B 72, 115303 (2005).

[3] P. Thomas et al., Non-polaritonic effects in cavity-modified photochemistry, Adv. Mater. 36, 2309393 (2024).

[4] F. Herrera and W. Barnes, *Multiple interacting photonic modes in strongly coupled organic microcavities*, Phil. Trans. R. Soc. A. 382 20230343 (2024).

[5] A. Mandal et al., Microscopic theory of multimode polariton dispersion in multilayered materials, Nano Lett. 23, 4082 (2023).







A unified quantum master equation model of cavity-coupled OLEDs

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The primary function of organic light-emitting diodes (OLEDs) is to convert injected electrons into photons. Yet, only 25% of the electronic states (singlets) in electrically excited, fluorescent molecules can emit light, which is why triplet harvesting has attracted significant attention [1–3]. In particular, one often aims to maximize the rate of reverse inter-system crossing (RISC) and depopulate the singlets fast enough, i.e., before they convert back to triplets or interact with other excited states, potentially breaking molecular bonds [4].

Planar microcavities provide a viable architecture to address these issues. By confining the emitters within planar microcavities one can couple the excitons with cavity modes and engineer the population dynamics to one's liking. While the weak-coupling regime is renowned for Purcell-enhanced emission [5], strongly coupled excitons and photons hybridize to form polaritons [6].

To fully understand and optimize exciton-photon interactions and light-emission mechanisms across various coupling regimes, a unified theory of cavity-coupled OLEDs is needed. Here, we introduce a quantum master equation model spanning the zero-, weak-, and strong-coupling regimes [7]. We derive the rates of all the relevant processes [see Fig. 1(a)] using Fermi's golden rule [8] and Marcus theory [9], solve the master equation in all the coupling regimes, and finally evaluate the internal quantum efficiencies [see Fig. 1(b)].

While our model can be used to optimize already accessible device architectures, it also motivates further studies on few-molecule strong coupling in OLEDs.

[1] L. A. Martínez-Martínez et al., *Triplet harvesting in the polaritonic regime: a variational polaron approach*, J. Chem. Phys. 151, 054106 (2019).

[2] A. G. Abdelmagid et al., *Identifying the origin of delayed electroluminescence in a polariton organic light-emitting diode*, Nanophotonics 13, 2565 (2024).







FIG. 1. (a) The system of interest: an organic molecule inside an optical cavity, embedded in a phonon bath and experiencing electrical excitation, polariton transitions, dephasing, ISC, RISC, emission, and nonradiative losses. Although a single, strongly coupled molecule is shown, we consider an ensemble of Nmolecules across all the coupling regimes: no coupling, weak coupling, and strong coupling. (b) Angledependent internal quantum efficiency (ADIQE) of the example stack Al-3DPA3CN-Al shown for different cavity thicknesses and $N = 10^6$. The black solid curves separate the weak (W) and strong-coupling (S) regimes.

[3] O. Siltanen et al., Enhancing the efficiency of polariton OLEDs in and beyond the singleexcitation subspace, Adv. Opt. Mater. 13, 2403046 (2025).

[4] E. Tankelevičiūtė et al., The blue problem: OLED stability and degradation mechanisms, J. Phys. Chem. Lett. 15, 1034 (2024).

[5] K. Vahala, Optical microcavities, Nature 424, 839 (2003).

[6] R. Bhuyan et al., The rise and current status of polaritonic photochemistry and photophysics, Chem. Rev. 123, 10877 (2023).

[7] O. Siltanen et al., Exciton dynamics and quantum efficiencies in optically coupled OLEDs: a unified quantum master equation approach, arXiv:2501.01800 (2025).

[8] E. Fermi, Nuclear Physics (University of Chicago Press, 1950).

[9] R. A. Marcus, On the theory of oxidation-reduction reactions involving electron transfer. I, J. Chem. Phys. 24, 966 (1956).







Tunable exciton polaritons in band-gap engineered hexagonal boron nitride

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Monolayer hexagonal boron nitride (hBN), is one of the most promising 2D materials for optoelectronics, since as a large band gap semiconductor, it displays an intense absorption in the ultraviolet range. Due to the reduced thickness of 2D materials, they show a reduced dielectric screening, which makes it possible for formation of excitons with bigger excitonic binding energies compared to 3D semiconductors. Studies on the optical properties of hBN can be found in the literature, but



FIG. 1. Schematic of the proposed setup in this work. A hBN monolayer is deposited on a quartz substrate, to which an electrostatic 1D periodic potential is applied along the x direction. We compute the optical response of this nanostructured system, when a plane wave with a pure harmonic frequency is impinged normally to the monolayer plane.







there is the need for ways of controlling the its excitonic properties. In this work [1], we suggest a form of tuning the excitonic binding energies by applying a one-dimensional periodic potential on top of hBN, just like depicted in Fig. 1. In this way, we form a superlattice structure that has distinct electronic properties from the original lattice. Namely, introducing the one-dimensional potential leads to the renormalization of the gap and of the electron and hole effective masses. We have seen that increasing the period of the potential increases the anisotropy of the dispersion, which results in a red-shift of the excitonic levels. We computed as well the optical conductivity, and have observed that the intensity of the peaks in the conductivity increases as the frequency of the peaks red-shift. Finally, we determine the dispersion relation of the exciton polaritons the light–matter hybrid modes that arise from the coupling of the exciton with light. Our results show that as the period of the potential increases and the energies of the excitonic states red-shift, the exciton polaritons become less confined.

 P. Ninhos et al., Tunable exciton polaritons in band-gap engineered hexagonal boron nitride, ACS Nano 18, 20751 (2024).







The role of the size of the exciton reservoir and polariton states on strong exciton photon coupling dynamics

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Strong exciton-photon coupling occurs when molecular transition dipole moment and electromagnetic field interaction energy exceeds energy dissipation in an optical cavity, spectroscopically observed as excited-state splitting into polaritons. For N molecules strongly coupled to a cavity mode, this creates N+1 hybrid states: two polaritons (upper and lower) acquire all oscillator strength, while the remaining N-1 states form an optically inactive "exciton reservoir" at the molecular energy level, critical for relaxation dynamics [1]. The relaxation from the exciton reservoir to the lower polariton is slow, making the dynamics of polaritonic relaxation primarily dependent on this relaxation rate. The rate of relaxation from the exciton reservoir to the lower polariton is mainly influenced by the size of the exciton reservoir. Therefore, understanding the effect of the size of the exciton reservoir on polaritonic photophysics is crucial.

In my presentation, I will demonstrate that polaritonic emission is inversely proportional to the size of the exciton reservoir [2]. Furthermore, I will present a kinetic model I developed to analyze angle-dependent lower polariton emission quantum yields. This model allows me to measure the relative size of the exciton reservoir and the lower polaritonic state with cavity detuning [3]. My research reveals that the size of the exciton reservoir decreases as the overlap between the absorption of the molecular transition and the lower polariton increases. Conversely, the size of the lower polariton increases with this overlap. Additionally, I will demonstrate how to use the source term method to quantitatively model the lower polariton emission in both transverse electric and transverse magnetic polarizations [4].

 R. Bhuyan et al., The rise and current status of polaritonic photochemistry and photophysics, Chem. Rev. 123, 10877 (2023).







[2] R. Bhuyan et al., The effect of the relative size of the exciton reservoir on polariton photophysics, Adv. Opt. Mater. 12, 2301383 (2024).

[3]R. Bhuyan and K. Börjesson, *The effect of cavity detuning on the size of the exciton reservoir* (unpublished).

[4] R. Bhuyan et al., *Quantitative modeling of polaritonic emission using the source term method* (unpublished).







Vast polaritonic enhancement of donor-acceptor energy transfer

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Presentation type: Poster

Through the process of Förster Resonance Energy Transfer (FRET), energy can be transferred nonradiatively from excited donor molecules to acceptor species over relatively short distances (<10nm). Owing to their delocalization, exciton-polaritons have been shown to extend this transfer radius to distances reaching up to $2\,\mu\mathrm{m}$ through the simultaneous coupling of donor and acceptor molecules to a common cavity mode [1,2,3]. However, the exact mechanism and dynamics of polariton-assisted energy transfer remain unclear. In this work, we prepare solution-processed Fabry-Perot microcavities involving a donor-acceptor pair with a design that enables the fine tuning of multiple parameters such as loading ratio, detuning, and Rabi splitting. Using these microcavities, we systematically investigate the mechanism of polariton-assisted energy transfer through a combination of photoluminescence and transient absorption spectroscopy. The results demonstrate that the intracavity dark state population in donor-acceptor microcavities is depleted markedly faster – an unexpectedly profound change in molecular photophysics. Our rate model reveals a polaritonic energy transfer rate that is 100,000 times higher than the rate in bare films. It also indicates that the cavity dynamics are dominated by the rapid equilibrium between the dark states and the lower polariton state, and energy transfer to the acceptors breaks this equilibrium. Consequently, we discover that this platform can be a valuable tool which provides insights into the general framework of polariton dynamics and offers new guidance on how to achieve polaritonic modification of molecular photochemistry.

[1] D. M. Coles et al., Polariton-mediated energy transfer between organic dyes in a strongly coupled optical microcavity, Nat. Mater. 13, 712 (2014).

[2] X. Zhong et al., Energy transfer between spatially separated entangled molecules, Angew. Chem. Int. Ed. 56, 9034 (2017).

[3] K. Georgiou et al., Ultralong-range polariton-assisted energy transfer in organic microcavities, Angew. Chem. Int. Ed. 60, 16661 (2021).







Morphology-driven light-matter interactions in semiconductor polaritonic systems—a review

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Exploring the impact of morphology is crucial for optimizing light-matter coupling and tailoring material properties for advanced photonic and quantum applications. Strong light-matter coupling occurs if the photon exchange between light and matter is faster than any other dissipation process, forming two new hybrid states called polaritons, as illustrated in Fig. 1 [1]. These polaritons exhibit characteristics of both photon and exciton and can be tuned by controlling cavity design, material composition, and exciton density. Further, the extent of light-matter interaction can also be tuned by varying the shape, size, and dimensionality of nanoparticles placed inside the cavity through their effects on the oscillator strength, local electromagnetic field, and spectral properties. Morphological variations influence quantum confinement effects, impacting key polaritonic parameters such as Rabi splitting, exciton binding energy, transition dipole moment, and dispersion characteristics [2]. Anisotropic nanostructures also introduce directional coupling effects, modifying polariton dispersion and polarization properties. Beyond individual nanoparticles, the surrounding dielectric environment and cavity structure further shape the energy landscape of polaritonic systems. However, excessive confinement may introduce additional losses, like nonradiative losses and increased scattering, affecting the stability and linewidth of the polaritonic states.



FIG. 1. Light-matter coupling in optical microcavities.









FIG. 2. Effects of light-matter strong coupling.

Advanced fabrication techniques, such as lithography-assisted nanopatterning, microemulsion, hydrothermal, and colloidal synthesis, are being explored to enhance reproducibility [3]. Experimental studies highlight the influence of morphology on light–matter interactions, revealing challenges in achieving precise control over nanoparticle shape for uniform and reproducible polaritonic effects. Overcoming these challenges is essential for integrating semiconductor nanomaterials into microcavities without altering their intrinsic properties.

Depending on embedded nanomaterial characteristics, optical microcavities, and nanostructured environments can amplify or suppress strong coupling effects. Morphology-driven modifications in polaritonic systems offer significant potential by tuning properties for applications in enhanced light harvesting, nonlinear optics, and efficient energy transfer [4] (Fig. 2). Spectroscopic investigations provide insights into morphology-dependent polaritonic behavior, offering pathways for tailoring optical and quantum properties through precise material engineering. The aim of this review is to highlight recent advancements in morphology-dependent polaritonic effects and their implications for photonic and quantum technologies. It emphasizes the interplay between nanoparticle shape, cavity design, and light-matter interactions, offering a foundation for optimizing polaritonic systems through material engineering.

 M. Hertzog et al., Strong light-matter interactions: a new direction within chemistry, Chem. Soc. Rev. 48, 937 (2019).

[2] S. Ghosh et al., Microcavity exciton polaritons at room temperature, Phot. Insights 1, R04 (2022).
[3] N. Baig et al., Nanomaterials: a review of synthesis methods, properties, recent progress, and challenges, Mater. Adv. 2, 1821 (2021).

[4] D. S. Dovzhenko et al., Light-matter interaction in the strong coupling regime: configurations, conditions, and applications, Nanoscale 10, 3589 (2018).







The quantum palette of methylene blue: unveiling vibronic deatures and the influence of dimerization and CB[7] encapsulation towards strong coupling applications

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Presentation type: Poster

The complex of Methylene Blue (MeB) with Cucurbit[7]uril (CB[7]) is a promising system for exploring strong light-matter interactions in polariton chemistry. CB[7] can attache a gold nanoparticle to a gold surface with a 0.9 nm gap. Due to the extreme confinement of the electromagnetic field within this gap, this self-assembled system can act as a plasmonic nanocavity for dyes that form a host-guest complex with the cucurbituril binding inside its barrel. In fact, by encapsulating MeB within CB[7] in such a nanoparticle nanocavity on a mirror, Baumberg and coworkers [1] were able to achieve strong single-molecule coupling and observe the characteristic Rabi splitting. The conclusion that the nanocavity contained a single MeB molecule was based on the absence of a shoulder in the absorption spectrum of MeB in the presence of CB[7] due to suppressed dimerization. However, the nature of the shoulder in the absorption spectrum has long been controversial. While some authors attribute this peak to a vibronic origin, others consider it to result from dimer formation. In this study, using atomistic computer simulations, we show that the shoulder consists of contributions from both dimerization and vibronic progression. After a single molecule of MeB binds to CB[7], the dimerization contribution is eliminated from the shoulder, while the vibronic progression remains. These findings contradict the absence of a shoulder observed in the spectra reported by Baumberg and coworkers. To verify our predictions, we conducted further optical absorption measurements, which confirmed the presence of the shoulder in the measured spectra. Our results thus not only help resolving the controversy regarding the nature of the shoulder, but also call for a critical reassessment of the single-molecule strong coupling regime in the nano-particleon-mirror cavity.

[1] R. Chikkaraddy et al., Single-molecule strong coupling at room temperature in plasmonic nanocavities, Nature 535, 127 (2016).







Theoretical insights into the resonant suppression effect in vibrational polariton chemistry

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Recent experiments demonstrated the possibilities of modifying ground-state chemical reaction rates by placing an ensemble of molecules in an optical microcavity through a resonant coupling between the cavity mode and molecular vibrations. Typical VSC experiments operate in the absence of any light source. The VSC induced rate constant suppression occurs under the resonance condition when cavity frequency matches the molecular vibrational frequency, and only at the normal incidence when considering in-plane momentum inside a Fabry-Pérot cavity. In this work, we use



FIG. 1. Normalized rate constant profile k/k_0 (with respect to the outside cavity case k_0) as a function of cavity frequency ω_c and various Rabi splittings (Ω_R).







quantum dynamics simulations and analytic theories to provide valuable insights into observed VSC phenomena, including the non-linear change of the rate constant when increasing Rabi splitting, modification of both reactive enthalpy and entropy, and a reason why, with a very low barrier, there is a lack of the cavity modification. The analytic theory also exhibits the normal incidence condition, as well as collective coupling effects.







Hierarchical pure states approach to non-linear spectroscopy of polaritonic quantum batteries

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Polaritons, hybrid quasi-particles from strong light–matter coupling, offer a promising platform for quantum batteries, where cooperative effects enhance energy storage and transfer. Understanding their dynamics requires accounting for phonon interactions, which can significantly alter energy absorption and emission.

In our recent work [1], we used the hierarchy of pure states (HOPS) method to study phononinduced effects in the absorption and resonance fluorescence spectra of a quantum dot, capturing non-Markovian and temperature-dependent effects. By extending this approach, we aim to model pump-probe spectroscopy of a polaritonic quantum battery to investigate nonlinear effects and charging dynamics under strong coupling.

Future work will focus on developing HOPS-based simulation methods for these processes, with the goal of providing a framework to explore phonon effects in polaritonic quantum batteries and their potential for efficient energy applications.

[1] S. Toivonen and K. Luoma, *Phonon-induced effects in quantum dot absorption and resonance fluorescence with hierarchy of pure states*, arXiv:2412.20598 (2024), submitted to PRB.







An atlas of photonic and plasmonic materials for cathodoluminescence microscopy

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Cathodoluminescence (CL) microscopy has emerged as a powerful tool for investigating the optical properties of materials at the nanoscale, offering unique insights into the behavior of photonic and plasmonic materials under electron excitation. We introduce an atlas of bulk CL spectra and intensity for a broad range of materials used in photonics and plasmonics (Fig. 1). Through a



FIG. 1. An atlas of investigated photonic and plasmonic materials.





combination of experimental CL spectroscopy and Monte Carlo simulations, we characterize electron penetration depth and energy deposition, offering a foundational reference for interpreting CL spectra and understanding material behavior under electron excitation [1]. Our atlas captures CL signals across a wide range of materials, offering valuable insight into intrinsic emission properties for informed material selection and device design in photonics and plasmonics.

[1] S. Ebel et al., An atlas of photonic and plasmonic materials for cathodoluminescence microscopy, arXiv:2411.08738 (2024).







Quantifying the effect of vibrational strong coupling on hydrogen bonding

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Presentation type: Poster

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Chemical systems assembled through non-covalent interactions (supramolecular chemistry) have been shown to undergo modifications under vibrational strong coupling (VSC) [1-4]. However, owing to the infancy of the field, the mechanism driving these changes remains elusive. To address this knowledge gap, NMR-compatible optical cavities were recently developed [5]. Using a molecular balance, van der Waals dispersion forces were found to be reduced in strength.

Building upon these findings, we synthesized a molecular balance to assess and quantify changes in hydrogen bonding under VSC by coupling vibrational bands of the solvents and the balance [6]. Preliminary results indicate that hydrogen bonding is weakened under VSC. Interestingly, when the hydrogen bond acceptor within the molecular balance is coupled, intramolecular hydrogen bonding becomes weaker. In contrast, coupling the hydrogen bond acceptor solvent weakens intermolecular hydrogen bonding, thereby strengthening the intramolecular hydrogen bond. So far, the magnitude of the VSC-induced effects is comparable to the changes observed when switching to a different









solvent, which highlights their potential significance in the context of chemical reactivity. Since hydrogen bonding interactions differ fundamentally from dispersion forces, they offer a complimentary insight into how VSC influences chemistry.

[1] K. Hirai et al., *Selective crystallization via vibrational strong coupling*, Chem. Sci. 12, 11986 (2021).

[2] K. Joseph et al., Supramolecular assembly of conjugated polymers under vibrational strong coupling, Angew. Chem. Int. Ed. 60, 19665 (2021).

[3] K. Sandeep et al., Manipulating the self-assembly of phenyleneethynylenes under vibrational strong coupling, J. Phys. Chem. Lett. 13, 1209 (2022).

[4] K. Joseph et al., Consequences of vibrational strong coupling on supramolecular polymerization of porphyrins, J. Am. Chem. Soc. 146, 12130 (2024).

[5] B. Patrahau et al., Direct observation of polaritonic chemistry by nuclear magnetic resonance spectroscopy, Angew. Chem. Int. Ed. 63, e202401368 (2024).

[6] N. Y. Meredith et al., Dissecting solvent effects on hydrogen bonding, Angew. Chem. Int. Ed. 61, e202206604 (2022).







High-Q microresonators unveil quantum rare events

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Presentation type: Poster

Classical linear optics asserts that for sufficiently weak intensity, light propagation through dielectric media is entirely governed by linear susceptibilities of the latter. We show that this paradigm becomes inadequate when light is confined long enough to engage in rare events mediated by the fluctuation in the quantum vacuum; this regime is achievable in high-Q microresonators, where photons can complete 10^8 to 10^{10} roundtrips before escaping. We reveal that these quantum-vacuum mediated optical processes directly imprint molecular Raman vibrational fingerprints onto the resonator's linear transmission spectrum—despite their absence in linear susceptibilities. We show that by increasing the number of deposited molecules, these otherwise weak Raman fingerprints can be amplified to levels well above the noise floor, rendering them accessible with emerging photonic architectures and detection schemes. This novel weak-coupling cavity quantum electrodynamical effect not only paves the way for new technology for chemical and biological sensing and Raman



FIG. 1. Schematic illustration of vacuum-mediated Raman processes in the linear optics of a microresonator system, highlighting the Stokes and anti-Stokes components.





spectroscopy but also pioneers a novel frontier in leveraging rare quantum vacuum fluctuations for photonic sensing and spectroscopy by harnessing the extended photon lifetimes and confinements in high-Q microresonators.

[1] S. Mukamel, Principles of Nonlinear Optical Spectroscopy (Oxford University Press, 1995).

[2] B. Dayan et al., A photon turnstile dynamically regulated by one atom, Science 319, 1062 (2008).

[3] J. Yuen-Zhou and A. Koner, *Linear response of molecular polaritons*, J. Chem. Phys. 160, 15 (2024).







Cathodoluminescence by backscattered and secondary electrons

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Cathodoluminescence (CL) microscopy in a scanning electron microscope (SEM) is a powerful technique for studying the optical properties of materials at the nanoscale. It has proven particularly useful in nanophotonics, enabling the investigation of nanoscale plasmonic structures and localized luminescent centers [1]. CL arises from a omplex excitation cascade triggered by an incident electron beam, typically confined to a generation volume on the order of $1 \mu m$ or smaller. However, electron beam interactions with a material produce not only CL but also a broad range of emissions, including X-rays, secondary electrons, and backscattered electrons [2]. Secondary electrons, with energies between $1 - 3 \,\text{eV}$, have extremely short mean free paths. In contrast, backscattered electrons, which are elastically scattered incident electrons, can travel significant distances before exiting the material's surface. Recent studies suggest that secondary electrons can also generate CL [3], with strong photon bunching effects observed in this excitation regime [4].

In this work, we systematically investigate the contribution of backscattered and secondary electrons to the cathodoluminescence response of a localized emitter system. Specifically, we study silicon vacancy centers in microdiamonds placed in different substrate environments to explore how these electron interactions influence CL excitation over multiple microns (Fig. 1(a-c)). By combining CL microscopy with Hanbury Brown and Twiss (HBT) interferometry we are able to identify three distinct excitation regimes based on secondary and back scattered electrons. The first excitation regime (Fig. 1(d)) is driven by backscattered electrons exhibiting complex diffusion characteristics. By placing the emitters on Si3N4-membranes we can systematically remove the contribution by backscattered electrons and reach the second excitation regime (Fig. 1(e)). This is characterized by pure secondary electron driven CL. The third regime is achieved by modifying the Si₃N₄ membrane surface with silicon spheres creating a geometry with enhanced secondary electron emission (Fig. 1(f)).









FIG. 1. Cathodoluminescence (CL) by backscattered electrons (BSE) and secondary electrons (SE). (a-c) Identified excitation regimes by BSE and SE. (d) Investigated zero phonon line at 738 nm of silicon vacancy centers. (e) Secondary electron image. (f) CL intensity map at 738 nm visualizing the excitation by SE and BSE electrons.

Overall, this work highlights the role of both elastically and inelastically scattered electrons in CL excitation of localized emitters, raising fundamental questions about the spatial resolution of cathodoluminescence spectroscopy. Additionally, our findings on CL excitation by secondary and backscattered electrons offer potential applications in low-current CL studies of beam-sensitive materials. Furthermore, they pave the way for developing ultra-low-energy electron sources in SEMs via secondary electron emission from nanostructures.

[1] T. Coenen and N. M. Haegel, *Cathodoluminescence for the 21st century: Learning more from light*, Appl. Phys. Rev. 4, 031103 (2017).

[2] L. Reimer, Scanning Electron Microscopy, Physics of Image Formation and Microanalysis (Springer Series in Optical Sciences, 1998).

[3] S. Meuret et al., Nanoscale relative emission efficiency mapping using cathodoluminescence g(2) imaging, Nano Lett. 18, 2288 (2018).

[4] V. Iyer et al., Photon bunching in cathodoluminescence induced by indirect electron excitation, Nanoscale 15, 9738 (2023).







A quantum description of the dissipative properties of cavity polaritons in the ultrastrong coupling regime

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Presentation type: Poster

The intriguing properties of polaritons resulting from strong light-matter coupling have been extensively investigated. Substantial research has focused on the ultrastrong coupling (USC) regime [1], where the light and matter coupling strength becomes comparable to the bare frequencies of the constituent modes. This effort has enabled the demonstration of polaritons in the USC regime across a variety of experimental platforms.

Quantum descriptions of polaritons have revealed many exotic phenomena, including a squeezed ground state with finite bound virtual photon excitations. However, despite significant research efforts, the connection between experimental observations and theoretical predictions of quantum effects in USC polaritonic systems remains tenuous, and the accurate description of these systems continues to be debated.

This work provides a toy model for cavity polaritons emerging through the photonic interaction with bosonic material excitations. Employing a simple two-mode Hopfield model, we derive analytical expressions for the polaritonic decay rates that are valid in the USC regime. Our work is motivated by the recent publication [2], which demonstrated that USC in a plasmon-microcavity system can significantly alter the polariton's dissipative properties. Specifically, [2] proposes a classical electromagnetic mechanism in which USC between two *lossy* modes results in a *narrow* lower polariton, contrary to common assumptions. However, this classical approach leaves open questions about the meaning of zero detuning and the connections to the light-matter interaction terms arising in a quantum mechanical description.

We present a quantum model to explore these effects, addressing the influence of the rotatingwave approximation and the A^2 term on the polariton's dissipative properties. Additionally, we relate the polaritonic linewidths to the properties of the uncoupled modes. Hence, our framework could guide the engineering of polaritonic resonances at will, e.g., for spectroscopy and sensing







applications. Finally, this poster addresses the technical subtleties of modeling cavity polaritons in the USC regime, including the concepts of "measurable" fields and gauge invariance [3]. Notably, we highlight the role of the bath-coupling form in open-quantum-system descriptions [4,5], which significantly influences the predicted polaritonic behavior.

[1] A. F. Kockum et al., Ultrastrong coupling between light and matter, Nat. Rev. Phys. 1, 19 (2019).

[2] A. Canales et al, Polaritonic linewidth asymmetry in the strong and ultrastrong coupling regime, Nanophotonics 12, 4073 (2023).

[3] L. Garziano et al., *Gauge invariance of the Dicke and Hopfield models*, Phys. Rev. A 102, 023718 (2020).

[4] S. Hughes et al., Reconciling quantum and classical spectral theories of ultrastrong coupling: role of cavity bath coupling and gauge corrections, Optica Quant. 2, 133 (2024).

[5] L. Garziano et al., Gauge-independent emission spectra and quantum correlations in the ultrastrong coupling regime of open system cavity-QED, Nanophotonics 11, 1573 (2022).







Mirror-induced effects in cavity polaritonics: influence on edge states

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Presentation type: Poster

Optical cavities are widely used to induce strong light–matter coupling and thereby enable the presence of polaritons. While polaritons are at the source of most of the observed physics, identifying



 \Rightarrow emergence of Tamm states

 \Rightarrow protection of polaritonic edge states

Effects of the longitudinal cavity mirrors



 \Rightarrow image-induced topological phase transition

FIG. 1. Sketch summarizing the system under study in this work as well as the mirror-induced effects we unveil. A chain of dipolar emitters is embedded in a closed cuboidal cavity with perfect metallic mirrors in all directions. The first and last emitters of the chain are placed at a distance d_{cav} from the cavity walls transverse to the direction of the chain. By investigating the effect of these transverse cavity mirrors we unveil the emergence of Tamm states and the protection of edge states in a topological chain as the distance d_{cav} is reduced. We also explore the specific impact of the longitudinal cavity mirrors, which generate image dipoles (represented in gray) that renormalize the dipolar degrees of freedom and that may induce topological phase transitions.







effects resulting strictly from the polaritonic nature of the excitation may be difficult. Indeed, the mirrors forming the cavity may themselves be responsible for a number of phenomena, leading to recent discussions on cavity-induced nonpolaritonic effects [1,2]. Motivated by these debates, we use here a toy model of a chain of dipolar emitters embedded in a cuboidal multimode cavity to unveil direct effects originating solely from the boundary conditions that such a cavity imposes on the electromagnetic field, independently of the presence of polaritons (see Fig. 1) [3]. We demonstrate that mirrors in the direction transverse to the chain may act as effective defects, leading to the emergence of Tamm edge states. In a topological chain, such transverse mirrors may also protect edge states against the effects of the strong light-matter coupling. Moreover, mirrors parallel to the chain, by the image charges they involve, may induce topological phase transitions even in the case of highly off-resonant photons.

 P. A. Thomas et al., Non-polaritonic effects in cavity-modified photochemistry, Adv. Mater. 36, 2309393 (2024).

[2] M. A. Michon and B. S. Simpkins, Impact of cavity length non-uniformity on reaction rate extraction in strong coupling experiments, J. Am. Chem. Soc. 146, 30596 (2024).

[3] T. F. Allard and G. Weick, Mirror-induced effects in cavity polaritonics: influence on edge states, Phys. Rev. B 110, 125423 (2024).







Organic microcavity polariton dynamics with non-Markovian quantum state diffusion

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The collective light-matter coupling between the electronic states of a large number of organic molecules and the electromagnetic field in a cavity results in hybrid light-matter states called polaritons. We build a model to explore the processes in these organic microcavity polariton systems. The electronic states of the molecules and the cavity modes form an open quantum system that interacts with the molecular vibrations and loses photons through the imperfect cavity mirrors. We model the environment interactions with non-Markovian quantum state diffusion [1]. This can be used to efficiently calculate the linear electric susceptibility for the cavity-molecule system. From this we get the refractive index, which can be used in the transfer matrix method to calculate the angle-dependent absorption, reflectivity and transmission spectra. We fit the absorption spectrum of the bare molecules calculated from our model to experimental data to get the properties of the molecules as fitting parameters. These parameters can then be used for simulating the whole cavitymolecule system to calculate the reflectivity spectrum which we fit to an experimentally measured one. We find that the calculated reflectivity spectrum fits the experimental data well for small angles, and for large angles the location of the polariton peaks remain within estimated error [2]. Next we will investigate the effects of a microcavity on an organic light emitting diode by simulating electrical excitation of the system and calculating the angle-dependent emission spectrum.

L. Diosi et al., Non-Markovian quantum state diffusion, Phys. Rev. A 58, 1699 (1998).
 T. Leppälä et al., Linear optical properties of organic microcavity polaritons with non-Markovian quantum state diffusion, Nanophotonics 13, 2479 (2024).







Selectively guiding excitation energy by collaborative strong light-matter coupling

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Strong light-matter coupling induces formation of polaritons, coherent superpositions of the excited states of the material and the confined light mode. In a typical Fabry-Pérot cavity strong coupling is achieved by introducing the coupled molecule inside the cavity in sufficient concentration. However, with the increased concentration the number of dark states, located at the molecular absorption energy, increases in proportion. Within all these hybrid states, i.e., the bright polaritons and the dark modes, the excitation is delocalized among all the molecules taking part in the coupling, and the excitation energy can easily be transferred from the bright polaritons to the dark states depending on their spectral overlap [1]. Simulations and experiments suggest that in their relaxation the polaritonic states, including the dark states, follow Kasha's rule and relax to the lowest energy state available for the system before decaying [1–4]. This hints that if a photoreactive molecule with the energy of the photoproduct well below the lower polariton is part of the superposition state, then the initially delocalized excitation will localize into one single reactive molecule which



FIG. 1. Artistic representation of polariton enhanced light-harvesting.





then undergoes the reaction bringing the system to its relaxed state [3]. We have shown that the excitation spectrum of 10-hydroxybenzo[h]-quinoline (HBQ) under strong coupling is a product of the excitation spectrum of the bare molecules and the absorption spectrum of the molecule–cavity system, suggesting that polaritons act as gateways for channeling an excitation into a molecule, which then proceeds normally and undergo excited state intermolecular proton transfer reaction like bare HBQ [5]. We also experimentally demonstrate that the extent of this excitation leak from polaritons to the bare molecules is determined by the spectral overlap between the bright polaritonic states and the molecular absorption.

Here we have combined HBQ with an inert molecule with a strong absorption at the same energy, forming collaborative strong coupling of both molecules with the same cavity mode. In this case the excitation energy initially shared among all the molecules, is expected to rapidly localize into one of the reactive HBQ molecules. By varying the amounts and ratio between the molecules we investigate the efficiency of the excitation energy guiding. The phenomenon could be utilized for example in the enhanced light-harvesting [3], as depicted in Fig. 1. Moreover, the strong absorption of the inert molecule enhances the coupling strength of HBQ, allowing us to further investigate our earlier hypothesis on the influence of bright/dark state overlap and thermal disorder in energy channeling — an idea that has also gained recent support from others [5, 6].

 H. L. Luk et al., Multiscale molecular dynamics simulations of polaritonic chemistry, J. Chem. Theory Comput. 13, 4324 (2017).

[2] S. Baieva et al., Dynamics of strongly coupled modes between surface plasmon polaritons and photoactive molecules: the effect of the Stokes shift, ACS Photonics 4, 28 (2016).

[3] G. Groenhof and J. J. Toppari, *Coherent light harvesting through strong coupling to confined light*, J. Phys. Chem. Lett. 9, 4848 (2018).

 [4] E. Hulkko et al., Effect of molecular Stokes shift on polariton dynamics, J. Chem. Phys. 154, 154303 (2021).

[5] A. Dutta et al., Thermal disorder prevents the suppression of ultra-fast photochemistry in the strong light-matter coupling regime, Nat. Commun. 15, 6600 (2024).

[6] T. Liu et al., Unlocking delocalization: how much coupling strength is required to overcome energy disorder in molecular polaritons?, Chem. Sci. 16, 4676 (2025).







Sympathetic mechanism for vibrational condensation enabled by polariton optomechanical interaction

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In this work, we develop an optomechanical formalism for macroscopic quantum states in excitonpolariton systems with strong exciton-phonon interactions (Fig. 1). Strong exciton–vibration coupling induces an effective optomechanical interaction between cavity polaritons and vibrational degrees of freedom of molecules.

We show that polariton optomechanical interactions induce dynamical backaction, resulting in dispersive and dissipative shifts in the complex vibrational response functions [1], opening the avenue for the vibrational control of polariton condensates at room temperature. Unlike conventional



FIG. 1. Schematic of the setup to generate macroscopic vibrational states within exciton-polariton BEC. A coherent laser drive (Pump) in the blue-detuned optomechanical configuration resonantly excites bright excitonic states (Exc), causing vibrational amplification through optomechanical coupling between molecular vibrations (Vib) and the cavity polariton BEC satisfying the phase-matching condition: $\hbar \mathbf{k}_{Pump||} = \hbar \mathbf{k}_{BEC} +$







optomechanical systems, polariton optomechanics features high-dimensionality and phase-space confinement due to the dispersion relations of exciton polaritons. Consequently, vibrational modes exhibit effective positive or negative mass depending on the detuning parameter.

We demonstrate a macrocoherent regime in exciton-polariton systems, where nonequilibrium polariton Bose–Einstein condensation coexists with macroscopically occupied vibrational states [2]. An effective optomechanical interaction between cavity polaritons and vibrational degrees of freedom of molecules, leading to vibrational amplification in a resonant blue-detuned configuration. This interaction provides a sympathetic mechanism to achieve vibrational condensation with potential applications in cavity-controlled chemistry, nonlinear, and quantum optics.

 V. Yu. Shishkov et al., Room-temperature optomechanics with light-matter condensates, Phys. Rev. B 110, 134321 (2024).

[2] V. Yu. Shishkov et al., Sympathetic mechanism for vibrational condensation enabled by polariton optomechanical interaction, Phys. Rev. Lett. 133, 186903 (2024).







Feedback cooling of fermionic atoms in optical lattices

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We discuss the preparation of topological insulator states with fermionic ultracold atoms in optical lattices by means of measurement-based Markovian feedback control. The designed measurement and feedback operators induce an effective dissipative channel that stabilizes the desired insulator state, either in an exact way or approximately in the case where additional experimental constraints are assumed. Successful state preparation is demonstrated in one-dimensional insulators as well as for Haldane's Chern insulator, by calculating the fidelity between the target ground state and the steady state of the feedback-modified master equation. The fidelity is obtained via time evolution of the system with moderate sizes. For larger 2D systems, we compare the mean occupation of the single-particle eigenstates for the ground and steady state computed through mean-field kinetic equations.

[1] H. M. Wiseman, Quantum theory of continuous feedback, Phys. Rev. A 49, 2133 (1994).

[2] L.-N. Wu and A. Eckardt, *Cooling and state preparation in an optical lattice via Markovian feedback control*, Phys. Rev. Res. 4, L022045 (2022).

[3] W. P. Su et al., Solitons in polyacetylene, Phys. Rev. Lett. 42, 1698 (1979).

[4] W. Kohn, Analytic properties of Bloch waves and Wannier functions, Phys. Rev. 115, 809 (1959).

[5] M. Modugno and G. Pettini, Maximally localized Wannier functions for ultracold atoms in onedimensional double-well periodic potentials, J. Phys. 14, 055004 (2012).

[6] D. Vorberg et al., Nonequilibrium steady states of ideal bosonic and fermionic quantum gases, Phys. Rev. E 92, p. 062119 (2015).







Enhanced delayed fluorescence in non-local metasurfaces: the role of electronic strong coupling

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Strong light-matter coupling with molecular excitons has attracted increasing interest due to its potential applications in chemistry and optoelectronics. Strong coupling, which emerges when the interaction between molecular electronic transitions and photons in an optical cavity exceeds a critical threshold, introduces a new regime of light-matter interaction. To achieve this regime, Fabry-Perot cavities have been extensively used to confine photonic modes due to their ease of fabrication and flexible tuning capabilities. However, the closed structure of Fabry-Perot cavities limits their potential implementation in optoelectronic applications.

In this study [1], we designed open cavities featuring non-local metasurfaces composed of aluminum nanoparticle arrays. The surface lattice resonances in these metasurfaces exhibit electronic strong coupling with the boron diffuoride curcuminoid derivative, known for its highly efficient thermally activated delayed fluorescence in the near-infrared [2]. Our findings show that delayed



FIG. 1. Electronic strong coupling effects on the delayed fluorescence dynamics in metasurfaces.






fluorescence induced by triplet-triplet annihilation can be enhanced by a factor of 2.0 - 2.6 in metasurfaces, independent of their photonic mode alignment with molecular electronic transitions. We demonstrate that this enhancement primarily arises from increased absorption within the organic layer, facilitated by nanoparticle arrays, while strong coupling has a minimal effect on reverse intersystem crossing rates. Finite-difference time-domain simulations further support our results. This study provides new insights into light-matter interactions in delayed fluorescence, highlighting potential applications in optoelectronic devices.

[1] Y.-C. Wei et al., Enhanced delayed fluorescence in non-local mMetasurfaces: the role of electronic strong coupling, arXiv:2501.08731 (2025).

[2] D.-H. Kim et al., *High-efficiency electroluminescence and amplified spontaneous emission from* a thermally activated delayed fluorescent near-infrared emitter, Nat. Photon. 12, 98 (2018).



