CUI/AIM & MPSD Workshop “Polaritonics from first principles”

Report of Contributions

https://indico.desy.de/e/abinitiopolaritons

Contribution ID: **2** Type: **not specified**

Exploring Electrical Conductance of Amorphous Non-conducting Polymers under Vibrational Strong Coupling

Monday 2 October 2023 09:00 (40 minutes)

Achieving electrical conductance in amorphous non-doped polymers is a challenging task. In my talk, I present how a serendipitous experiment led us to the finding that the vibrational strong coupling (VSC) of amorphous polymers such as polystyrene, deuterated polystyrene, poly (benzyl methacrylate), and a few other polystyrene derivatives enhance the electrical conductance by orders of magnitude. Remarkably, the electrical conductance enhancement in these polymers is selective to the VSC of its aromatic C-H(D) out-of-plane bending modes. The conductance characteristics vary from diffusive to ballistic, depending on the strong coupling strengths. The electrical characterizations are performed without external light excitation, demonstrating the role of quantum light in enhancing the long-range coherent transport even in amorphous non-conducting polymers.

Reference:

1. Kumar, S.; Biswas, S.; Rashid, U.; Mony, K. S.; Vergauwe, R.; Kaliginedi, V.; Thomas, A. Extraordinary Electrical Conductance of Non-Conducting Polymers Under Vibrational Strong Coupling. arXiv 2023.https://doi.org/10.48550/arXiv.2303.03777.

Presenter: THOMAS, Anoop (Indian Institute of Science)

Session Classification: Morning session

Contribution ID: **3** Type: **not specified**

Analysis of cavity-suppressed reactivity via vibrational depopulation

Monday 2 October 2023 09:40 (40 minutes)

Molecular vibrations can couple to optical cavities to create new hybrid states called polaritons. Under such conditions, one may realize modified materials processes such as photon emission, molecular energy transfer, and chemical reaction rates. In this talk, I will first discuss modeling and active control of cavity coupling to molecular vibrations. Next, I will discuss measurements of reaction-rate constants extracted from evolving cavity transmission spectra, which reveal resonant suppression of the intracavity reaction rate for alcoholysis of phenyl isocyanate with cyclohexanol. We observe up to an 80% suppression of the rate by tuning cavity modes to be resonant with the reactant isocyanate (NCO) stretch, the product carbonyl (CO) stretch, and cooperative reactant-solvent modes (CH). Cavity tuning was used to selectively couple to reactant, product, and cooperative solvent-reactant vibrational modes resulting in a chemical response that is cavity tuning dependent. These results are explained using an open quantum system model that predicts resonant modifications of chemical reactivity via light-matter quantum coherences that depopulate vibrational excited states, suggesting fundamental links between chemistry and quantum science to be explored. I will show numerical experiments we carried out in search of artifacts that could impact the measurements described above. Finally, I will be briefly present the design of an experimental apparatus aimed at making liquid phase cavity-modified chemistry experiments more reliable, repeatable, and accessible for new researchers in the field.

Presenter: SIMPKINS, Blake (Naval Research Laboratory)

Session Classification: Morning session

Contribution ID: **4** Type: **not specified**

Welcome

Monday 2 October 2023 08:45 (15 minutes)

Presenters: RUBIO, Angel (Max-Planck Institute for the Structure and Dynamics of Matter); PUPILLO, Guido (University of Strasbourg); RUGGENTHALER, Michael (MPSD (Max-Planck-Forschungsgruppe)); SCHWARTZ, Tal (Tel Aviv University)

Session Classification: Morning session

Contribution ID: **5** Type: **not specified**

Local vs. collective interplay of (thermal) fluctuations in polaritonic chemistry Submitting author

Monday 2 October 2023 11:05 (40 minutes)

We present recent ab-intio simulation results for matter strongly dressed by optical cavities. Our focus will be on the relevance of classical and quantum features emergent from the vibrational strong coupling to the vacuum field fluctuations at finite temperature. The role and description of cavity induced non-canoncial equilibrium condition for chemical systems, as well as the relevance of thermal baths for local and collective features will be explored and consequences for the future development of ab-initio methods will be derived.

Presenter: SIDLER, Dominik (Max-Planck Institute for the Structure and Dynamics of Matter)

Session Classification: Morning session

Contribution ID: **6** Type: **not specified**

How do chemical bonds break in ultrastrong coupling?

Monday 2 October 2023 11:45 (40 minutes)

Current efforts for implementing cavity QED in the ultrastrong coupling (USC) regime using molecular vibrations in mid-infrared nanoresonators opens exciting opportunities for exploring novel chemical reactivities that exploit quantum correlations with the electromagnetic vacuum at room temperature. While most theoretical studies have primarily focused on rationalizing previous experimental reports of modified chemical reactions in collective strong coupling [1,2], the possibilities for single-molecule chemistry in the fully-quantum regime of light and matter still remains largely unexplored. We developed a coordinate representation approach for the light-matter Hamiltonian (DVR) that together with exact quantum dynamics techniques (MCTDH) provides key physical insights on the evolution of molecular and photonic observables over sub-picosecond timescales [3-5]. We use these methods to understand the quantum mechanical nature of molecular dissociation in infrared cavities under USC. For polar anharmonic bonds, we show that the combined influence of Bloch-Siegert shifts, wavepacket spreading, nuclear anharmonicity and vacuum field polarization via permanent dipoles, can lead to significant enhancement of the molecular dissociation rate relative to free space, for experimentally relevant non-equilibrium scenarios [6]. Our work thus provides conceptual and numerical tools for understanding the new possibilities that cavity QED physics can bring to nanoscale chemistry.

References:

[1] K. Nagarajan, A. Thomas, T. W. Ebbesen, JACS 143, 16877-16889, 2021.
[2] W. Ahn, J. F. Triana, F. Recabal, F. Herrera, B. Simpkins, Science 380, 1165, 2023.
[3] F. Hernández and F. Herrera, J. Chem. Phys. 151, 144116, 2019.
[4] J. F. Triana, F. J. Hernández, F. Herrera, J. Chem. Phys. 152, 234111, 2020.
[5] F. Triana and F. Herrera, J. Chem. Phys. 157, 194104, 2022.
[6] J. F. Triana, F. Herrera, DOI:10.26434/chemrxiv.12702419.v1, 2020.

Presenter: HERRERA, Felipe (Universidad de Santiago de Chile)

Session Classification: Morning session

Contribution ID: **7** Type: **not specified**

Exploring the zeroth-order mode of a Fabry-Perot cavity with Terahertz fields

Monday 2 October 2023 14:35 (40 minutes)

A planar Fabry-Perot cavity is studied at the deep-subwavelength inter-mirror distances, termed “zeroth-cavity mode”. Enhanced transmission is observed as d⟶0 using time-domain terahertz spectroscopy.

Presenter: FLEISCHER, Sharly (Tel Aviv University)

Session Classification: Afternoon session

Contribution ID: **8** Type: **not specified**

Kinetics in a cavity: the role of dynamical effects

Monday 2 October 2023 15:15 (40 minutes)

The observed modification of thermal chemical rates in Fabry-Perot cavities remains poorly understood. Mounting evidence supported by theory indicates that the cavity has a small effect on the potential energy barrier, suggesting that dynamical corrections may play a prominent role. This is in line with the relatively small modifications of the rate constant usually reported, often not exceeding a factor of 10, at most.

Here we will describe classical atomistic simulations on an ab initio potential of the cis-trans isomerization reaction of nitrous oxide (HONO) in a cavity [1]. Using the flux method to compute the recrossing coefficient, we will examine the conditions under which the cavity induces modifications of the reactive flux leading to modifications of the thermal rate. We will also consider small ensembles of HONO molecules in the gas phase, and thus shed light on the role of collective effects through specific numerical experiments [2].

Switching gears, and if time permits, I will introduce a few hints on the role of molecular symmetry and Jahn-Teller distortions in optical cavities, and how vibronic effects can result in exotic “inverse polarization” states.

References:

[1] Sun, Vendrell; JPCL 13, 4441 (2022)
[2] Sun, Vendrell; JPCL 14, 8397 (2023)
[3] Nandipati, Vendrell; PRA 107, L061101 (2023)

Presenter: VENDRELL, Oriol (University of Heidelberg)

Session Classification: Afternoon session

Contribution ID: **9** Type: **not specified**

”Disorder-protection” in many-body cavity-QED

Monday 2 October 2023 16:40 (40 minutes)

Theoretically explaining experimental observations of cavity-modified physics and chemistry remains to be a major challenge, in particular for a large number of coupled emitters (N). Here I discuss a bottom-up approach with minimal quantum many-body models, which include electronic, photonic, and motional degrees of freedom in their simplest form. Solving these (already complex) many-body models for large N with advanced numerical methods allows us to gain insight on fundamental physical processes.

In this talk I discuss how disorder in cavity-QED models can lead to exotic localization features. In particular, I introduce how dark states acquire a “semilocalized” nature with unusual properties in terms of level statistics and other localization quantifiers. I then discuss how such states can play a crucial role for energy transport and cavity-modified vibrational dynamics. As a main conclusion, our work shows that disorder and semilocalized states can make cavity-effects more robust in a large-N limit, where individual emitter cavity-coupling strengths are small.

Presenter: SCHACHENMAYER, Johannes (University of Strasbourg)

Session Classification: Afternoon session

Contribution ID: **11** Type: **not specified**

Insight from first principles: Embedding Techniques, Rate and Thermodynamic Changes, Solvation Effects, and Chiral Polaritonics

Tuesday 3 October 2023 09:00 (40 minutes)

Various experiments have demonstrated that strong collective interaction influences chemical reactivity – it remains to build a conclusive understanding.
Certain aspects such as a resonant behaviour, specific trends in the thermodynamic observables, and a strong sensitivity to the solvent have been identified as critical.
Capturing those effects in a consistent theoretical model is challenging. Here, we will discuss the beneficial use of ab initio quantum electrodynamics, embedding techniques [3,4], and machine learning [2]. Specific observations include cavity induced resonant changes in rate, enthalpy, and entropy that are qualitatively consistent with experiment [1,2] and changes in the local solute-solvent behaviour induced by collective strong coupling [5]. Lastly, we discuss the consequences of breaking chiral symmetry with specifically designed electromagnetic environments, thus suggesting a new direction for polaritonics and chiral recognition [6,7].

References:

[1] C. Schäfer, J. Flick, E. Ronca, P. Narang, and A. Rubio, Nature Communications, (2022) 13:7817.
[2] C. Schäfer, J. Fojt, E. Lindgren, P. Erhart, to be submitted (2023).
[3] C. Schäfer and G. Johansson, PRL 128, 156402, (2022).
[4] C. Schäfer, Phys. Chem. Lett. 2022, 13, 30, 6905-6911.
[5] M. Castagnola, T. Haugland, E. Ronca, H. Koch, C. Schäfer, to be submitted (2023).
[6] C. Schäfer, D. Baranov, J. Phys. Chem. Lett. 2023, 14, 15, 3777-3784.
[7] D. Baranov, C. Schäfer, M. Gorkunov, ACS Photonics 2023, 10, 8, 2440-2455.

Presenter: SCHAEFER, Christian (Chalmers University)

Session Classification: Morning session

Contribution ID: **12** Type: **not specified**

Toward simulations of quantum dynamics at complex plasmonic interfaces

Tuesday 3 October 2023 14:35 (40 minutes)

Noble-metal nanostructures support collective electron resonances, known as plasmons, that can “squeeze”optical fields down to nanometer-scale volumes, well below the conventional diffraction limit. The strong localization of electromagnetic fields greatly enhances nonlinear optical processes, both in the metal nanostructure itself and in adjacent optical materials. Because the processes occur entirely within a sub-wavelength volume, the usual phase-matching conditions for these processes are lifted. Ensembles of quantum emitters optically coupled to plasmonic nanocavities, have recently emerged as new platforms for strong light-matter interactions. When the strength of coupling between the plasmons in the metal nanostructures and resonant materials (such as molecules, molecular aggregates, quantum dots) exceeds decay rates in the coupled system, new phenomena appear. In particular, mixed light-matter systems known as polaritons are formed in this “strong-coupling”regime. These quasiparticles synergistically combine the nonlinear properties of the plasmonic and excitonic constituents and are thus ideal for the development of efficient nonlinear photonic devices. As we advance our understanding of the physics of such systems the need for multiscale simulations arises. The major challenge is to combine Maxwell’ s equations with quantum dynamics pertaining to molecules driven by electromagnetic radiation, which results in a highly unbalanced load of multiple processors when done conventionally. In this talk I will discuss the efficient parallel methodology recently developed for simulating nonlinear dynamics of a large number of molecules with ro-vibrational degrees of freedom explicitly considered in time domain and directly coupled to Maxwell’s equations in three dimensions in real time.

Presenter: SUKHAREV, Maxim (Arizon State University)

Session Classification: Afternoon session

Contribution ID: **13** Type: **not specified**

Cavity-induced cooperativity of a handful of molecules

Tuesday 3 October 2023 16:40 (40 minutes)

Over the past two decades, we have achieved a high degree of mastery in studying fundamental nano-optical interactions between single molecules and single photons. These include controlled coherent experiments in the near field, far-field demonstration of efficient extinction, various nonlinear measurements and single-molecule strong coupling in a microcavity. More recently, we have pursued projects, in which we aim to examine the cooperative coherent interaction of several well-controlled individual molecules via a common photonic mode. In this talk, I present results on two different platforms, involving an open Fabry-Perot microcavity and a chip-based integrated circuit.

Presenter: SANDOGHDAR, Vahid (Max-Planck Institute for the Physics of Light, Erlangen)

Session Classification: Afternoon session

Contribution ID: **14** Type: **not specified**

Molecular cavity dynamics: Numerical experiments and theoretical interpretations

Tuesday 3 October 2023 11:45 (40 minutes)

Recent numerical experiments on (a) consequences of vibrational strong coupling (b) polariton propagation and (c) manifestation of collective molecular response in optical cavities will be presented and discussed.

Presenter: NITZAN, Abraham (University of Pennsylvania)

Session Classification: Morning session

Contribution ID: **15** Type: **not specified**

Beyond the cavity

Tuesday 3 October 2023 09:40 (40 minutes)

In many of the envisaged applications of strong coupling, especially polaritonic chemistry, open access to the molecules involved is vital, as is independent control over polariton dispersion, and spatial uniformity of the coupling. Existing cavity designs are not able to offer all of these advantages simultaneously. Here we demonstrate an alternative yet simple cavity design that exhibits all the desired features. We hope that our new approach will provide a new platform to both study and exploit molecular strong coupling. Although our experimental demonstration is based on excitonic strong coupling, we also indicate how the approach might be achieved for vibrational strong coupling.

Presenter: BARNES, Bill (University of Exeter)

Session Classification: Morning session

Contribution ID: **16** Type: **not specified**

Modification of charge transport properties of water in optical cavity

Tuesday 3 October 2023 15:15 (40 minutes)

The effect of vibrational strong coupling of water on the ionic conductivity is discussed. Modification of the ionic transport in aqueous electrolyte solutions depends on the hydration structures, showing improved conductivities than those based on the classical interpretation.

Presenter: MURAKOSHI, Kei (Hokkaido University)

Session Classification: Afternoon session

Contribution ID: **17** Type: **not specified**

Ultrafast dynamics of molecular polaritons

Tuesday 3 October 2023 11:05 (40 minutes)

Through the strong coupling of molecular vibrational modes with photonic modes, intriguing molecular vibrational polariton states and dark reservoir modes emerge. These polaritons, exhibiting a dual nature of matter and light, have the potential to modify chemical reactions under thermally activated conditions, paving the way for the emerging field of polariton chemistry. Distinguishing polaritons from dark modes has been challenging, but ultrafast two-dimensional infrared (2D IR) spectroscopy proved instrumental in overcoming this hurdle. Our research demonstrated that polaritons facilitate intra- and intermolecular vibrational energy transfer, providing a means to control vibrational energy flow in liquid-phase molecular systems. Moreover, in studying a single-step isomerization event, we confirmed polaritons' role in modifying chemical dynamics under strong coupling conditions, while dark modes behaved like uncoupled molecules, leaving dynamics unchanged. This finding solidified the central concept of polariton chemistry and laid the groundwork for designing future polariton cavities.

Presenter: XIONG, Wei (University of California)

Session Classification: Morning session

Contribution ID: **18**

Type: **not specified**

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Wednesday 4 October 2023 09:00 (40 minutes)

Presenter: KOCH, Henrik (Norwegian University of Science and Technology)

Session Classification: Morning session and closing remarks

Contribution ID: **19** Type: **not specified**

Cooperative quantum optics with molecules

Wednesday 4 October 2023 11:05 (40 minutes)

Cooperative effects in complex, coupled quantum systems, cannot be understood by sole consideration of the individual constituents, as they arise from the interplay among them. Light-matter platforms provide an optimal playground for the observation and exploitation of quantum cooperative effects [1]. For example, structured subwavelength arrays of quantum emitters trapped in optical lattices, are ideal showcases of such cooperative behavior, as their optical response can be efficiently enhanced by controlling the hopping of surface excitations via the quantum electromagnetic vacuum induced dipole-dipole interactions.
While subwavelength separations are not easily achieved in standard quantum optics setups, molecular dimers and molecular aggregates (i.e.~arrays of identical molecules, such as J- and H-Aggregates) can feature deeply subwavelength separations on the nanometer scale. The downside of such systems is the much more complex structure, which introduces coupling of electronic degrees of freedom with intra- and inter-molecular vibrations. We have introduced a quantum Langevin equations approach to electron-vibron interactions for single molecules subject to either classical or cavity quantum light fields [2]. The extension of this method to many particles allowed us to benchmark the scaling of cooperative effects such as super- and subrradiance to molecular rings or chains, to quantify the effect of vibrations onto the operation of such systems as nanoscale coherent light sources [3] and to quantitatively describe couplings among collective electronic states via vibrations, in a process known as Kasha's rule [4].

References:

[1] M. Reitz, C. Sommer, and C. Genes, Cooperative Quantum Phenomena in Light-Matter Platforms, PRX Quantum 3, 010201 (2022).
[2] M. Reitz, C. Sommer and C. Genes, Langevin approach to quantum optics with molecules, Phys. Rev. Lett. 122, 203602 (2019).
[3] R. Holzinger, S. Oh, M. Reitz, H. Ritsch and C. Genes, Cooperative subwavelength molecular quantum emitter arrays, Phys. Rev. Research 4, 033116 (2022).
[4] R. Holzinger, N. S. Bassler, H. Ritsch and C. Genes, Scaling law for Kasha's rule in photoexcited subwavelength molecular aggregates, arxiv: 2304.10236 (2023).

Presenter: GENES, Claudiu (Max-Planck Institute for the Physics of Light, Erlangen)

Session Classification: Morning session and closing remarks

Contribution ID: **20** Type: **not specified**

Chemistry and photochemistry in the strong coupling regime - from single molecules to ensemble of molecules

Wednesday 4 October 2023 09:40 (40 minutes)

The theoretical description of molecular polaritons and their properties is still subject to many open questions. In the past years we studied a variety of effects, such as dissipation, to determine its influence on the photochemistry. More recently we have implement Hartree-Fock with the cavity Born-Oppenheimer approximation to investigate the multi-molecule effects in the electronic ground state. Here, we have identified the dipole self energy contributions as a new mechanism to mediate inter-molecular interactions.

Presenter: KOWALEWSKI, Markus (University of Stockholm)

Session Classification: Morning session and closing remarks

Contribution ID: **21** Type: **not specified**

Some aspects of light-matter interactions in chiral Fabry-Pérot cavities

Wednesday 4 October 2023 11:45 (40 minutes)

In this presentation, I will review some recent experimental [1,2] and theoretical attempts [3,4] to design innovative electromagnetic cavities for chiral-sensing purposes or polaritonic chemistry. In particular, I will focus on the interplay between the degree of polarization of the optical cavitymode and the intra-cavity light-matter interactions, exhibiting some difficulties to enhance circular-dichroism in standard optical Fabry-Pérot cavities. I will then provide some general theoretical framework [4,5,6] to describe the rich physical mechanisms that are responsible for the polarization-dependent transmission and absorption signals, developing in linear and reciprocal Fabry-Pérot optical cavities. An example will be shown of helicity-preserving cavity [3,4] made of two photonic dielectric mirrors. A Pasteur medium hosted inside such a cavity is shown to couple efficiently, in a narrow frequency range, to both the output electromagnetic field and to the helicity-preserving cavity mode, thus inheriting an enhanced chiral character. For such cavities, I will show some interesting theoretical predictions, reporting clear signatures of the formation of chiral cavitypolaritons. Some open perspectives in the theoretical modeling [7,8], experimental design and use of such chiral Fabry-Perot cavities in the strong-coupling regime for ultrasensitive chiral sensing in optics or stereochemistry will be given.

References:

[1] Semnani et al., Light: Science & Applications, 9:23 (2020).
[2] J. Gautier et al., ACS Photonics, 9, 778−78 (2022)
[3] K. Voronin et al., ACS Photonics, 9, 2652−2659 (2022).
[4] L. Mauro, J. Fregoni, J. Feist, and R. Avriller, Phys. Rev. A 107, L021501 (2023).
[5] R. Carminati et al., Phys. Rev. A, 62, 012712 (2000).
[6] I. Fernandez-Corbaton et al., Phys. Rev. X, 6, 031013 (2016).
[7] R. R. Riso et al., Phys. Rev. X 13, 031002 (2023).
[8] C. Schäfer et al., J. Phys. Chem. Lett. 14, 3777−3784 (2023).

Presenter: AVRILLER, Remi (Bordeaux University)

Session Classification: Morning session and closing remarks

Contribution ID: **22** Type: **not specified**

Closing remarks

Wednesday 4 October 2023 12:25 (15 minutes)

Presenters: RUBIO, Angel (Max-Planck Institute for the Structure and Dynamics of Matter); PUPILLO, Guido (University of Strasbourg); RUGGENTHALER, Michael (MPSD (Max-Planck-Forschungsgruppe)); SCHWARTZ, Tal (Tel Aviv University)

Session Classification: Morning session and closing remarks

Contribution ID: **23** Type: **not specified**

Poster: Theoretical investigation of electron transfer reaction rate accelerated by molecules-vacuum field strong coupling

We will discuss the possibility of modulation of interfacial electrochemical reaction rate via polaritonic effect under dark conditions. Based on the previously documented proposal, acceleration of the electron transfer reaction rates are estimated by assuming the coupling strength of molecules at electrified interfaces.

Presenter: HAYASHI, Takahiro

Contribution ID: **24** Type: **not specified**

Poster: Computationally efficient methods for studying strong coupling in cavities

Strong coupling between molecules and electromagnetic cavities is commonly studied using few-level quantum models, such as the Jaynes–Cummings or Dicke models, which describe matter in a highly simplified way. Alternatively, there are first-principles approaches such as time-dependent density-functional theory (TDDFT) and quantum-electrodynamical density-functional theory (QEDFT), which are computationally very costly. Here, we explore methods that have low or intermediate computational cost, yet still retain material specificity. Using only dipolarly coulpled polarizable units in a classic electrostatics framework, we are able to almost quantitatively reproduce optical spectra of strongly coupled nanoparticle-molecule assemblies [1]. Training neuroevolution potentials (NEPs) for predicting the energetic landscape, as well as permanent dipoles of molecules, we are able to study the modification of chemical reaction rates in cavities [2].

References:

[1] J. Fojt, T. P. Rossi, T. J. Antosiewicz, M. Kuisma, P. Erhart, J. Chem. Phys. (2021).
[2] C. Schäfer, J. Fojt, E. Lindgren, P. Erhart, to be submitted (2023).

Presenter: FOJT, Jakub (Chalmers University)

Contribution ID: **25** Type: **not specified**

Poster: Enabling multiple intercavity polariton coherences by adding quantum confinement to cavity molecular polaritons

A new microcavity infrastructure was designed and constructed to study molecular vibrational polaritons, which are hybrid particles blending aspects of light and matter. This implementation of photonic confinement within the Fabry-Perot cavity introduced additional 'quantized' cavity modes and allowed for the creation of polaritonic multi-qubit systems, also referred to as qudits. These qudits exhibited notable coherence stability even in the presence of environmental fluctuations. This work has implications for the development of molecular vibrational polaritons in the context of quantum information technology and may have broader applications in topological polariton systems and quantum light spectroscopy for molecular systems.

Presenter: YANG, Zimo (University of California)

Contribution ID: **26** Type: **not specified**

Poster: Controlling Excimer Formation Through Strong Coupling

Rapid and efficient energy migration necessitates minimization of trap state formation during excited state processes, such as excimer in organic photovoltaic devices [1]. In this direction, we show that the strong coupling of matter with quantum light can provide the handle to regulate the excimer formation dynamics. Our static fluorescence investigation demonstrates that electronic strong coupling can provide an alternative pathway to channel the radiative emission through the polaritonic state and hinder the excimer formation by tuning the ladder of molecular energy levels without perturbing the excimer state energy level. Interestingly, their ultrafast decay kinetics reveal the enhancement in the excited state deactivation rate and reduction of the excimer lifetime under the strong coupling regime. The interaction between quantum light and matter selectively facilitates efficient energy migration by reducing excimer formation in a strong coupling regime and enhancing excimer emission under weak coupling scenarios.

References:

[1] Morishima, Y.; Tominaga, Y.; Nomura, S.; Kamachi, M. Fluorescence and Energy Migration in the Random Copolymers of (1-Naphthylmethy) methacrylamide and Sodium 2-(Acrylamido)-2-methylpropanesulfonate. Macromolecules. 1992, 25, 861-866.

Presenter: BISWAS, Subha (Indian Institute of Science)

Contribution ID: **27** Type: **not specified**

Poster: ENGINEERING ORGANIC MOLECULES WITH LONG-LIVED QUANTUM COHERENCE

Single organic molecules in the solid-state are one of the promising optical platforms for realizing quantum networks owing to their remarkable coherent properties and flexibility in their chemical synthesis. However, the molecular excited states associated with the strong Fourier-limited zero-phonon lines of these systems decay within nanoseconds, posing a challenge for practical applications in quantum technologies.

In this theoretical work, we propose a new molecular system with quantum coherences up to millisecond time scales. Here, we exploit the inherent optomechanical character of organic molecules in a solid organic crystal. The proposed scheme consists of a single organic molecule in a host matrix with a structured phononic environment. By suppressing phononic decay channels, we realize and exploit long optomechanical coherence times up to milliseconds for storing and retrieving information. We show that the resulting long-lived vibrational states facilitate reaching the strong optomechanical regime at the single photon level. The proposed system shows the promise of organic molecules for achieving unexplored optomechanical phenomena and long-lived quantum memories.

Presenter: GUERLEK, Burak (Max-Planck Institute for the Structure and Dynamics of Matter)

Contribution ID: **28** Type: **not specified**

Poster: Beyond electron-photon exchange potential for effective photon-free QEDFT: suppression factor and electron-phonon correlation potential

Here we show the effective photon-free QEDFT approach works well in the strong coupling regime and requires the 'suppression'factor and electron-photon correlation potential in the weak coupling regime. We apply the approach to 1D, 2D, and 3D one-electron finite systems to focus on the electron-photon interaction.

Presenter: LU, I-Te (Max-Planck Institute for the Structure and Dynamics of Matter)

Contribution ID: **29** Type: **not specified**

Poster: Linear Response in Relativistic Dirac–Kohn–Sham Quantum-Electrodynamical Density Functional Theory

We derived and implemented relativistic QEDFT in linear response regime. The electrons are treated at the four-component Dirac–Kohn–Sham level of theory and coupled to transverse photons treated as dynamical variables. We show that light–matter coupling can enhance the strength of usually weak singlet–triplet transitions via a new type of spin–orbit interaction mediated by cavity photons.

Co-author: RUGGENTHALER, Michael

Presenters: KONECNY, Lukas (Max-Planck Institute for the Structure and Dynamics of Matter); RUGGENTHALER, Michael; KOSHELEVA, Valeriia (Max-Planck Institute for the Structure and Dynamics of Matter)

Contribution ID: **30** Type: **not specified**

Poster: Individual and Collective Nonadiabaticity in Cavities

Full-dimensional quantum-dynamical simulations of homonuclear diatomic molecules inside Fabry-Pérot cavities are performed. Accompanied by analytical results, conical intersections (CIs) between polaritons are identified whose branching spaces are spanned by molecular rotations inside the cavity polarization plane. They are closely related to light-induced CIs (LICIs). However, unlike LICIs, they are of collective nature since they depend on the relative orientation of molecular electronic transition dipoles. We demonstrate numerically that, in the collective regime, ultrafast rotational dynamics around both types of CIs are coexistent, adding to the complex interplay among molecular structure and photonic modes in gas-phase cavity chemistry. Such rotational dynamics depends critically on light-molecule coherence which can be exploited to probe nonradiative decay towards the dark state manifold.

Presenter: KRUPP, Niclas (University of Heidelberg)