

3rd workshop on

Molecular Quantum Technology

December 16-20 | Puerto Varas, Chile

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WELCOME NOTE

The Millennium Institute for Research in Optics (MIRO) is pleased to host the third edition of the Workshop for Molecular Quantum Technologies in Chile, in collaboration with sponsoring partners at the Institute for Theoretical Atomic and Molecular Physics (ITAMP), Philosophical Magazine Part C, The Journal of Chemical Physics, and APL Quantum.

The vision for MQT Workshops is to bring together engaging groups of renowned scientists and early career researchers across physics, chemistry and materials science, to discuss fundamental science and applications in which molecules and molecular materials can contribute to the field of quantum technology.

As in previous editions, the MQT 2024 organizing committee has made best efforts to have a fair representation of a diverse community of scientists. In numbers, 33% of invited speakers are women, 26% of all speakers are early-career scholars, and 35% of all participants are graduate students or postdoctoral researchers.

We are proud to continue our commitment to the development of molecular quantum science and look forward to an exciting week in Puerto Varas.

PROGRAM

	Mon 16	Tue 17	Wed 18	Thu 19	Fri 20	
08:00 - 08:20						
08:20 - 08:40						
08:40 - 09:00	Opening					
09:00 - 09:20	E. Bittner	F. Luis		A. Carroll	D. Leitner	
09:20 - 09:40	J. Baldoví	K. Aruachan		F. Recabal	M. Soley	
10:00 - 10:20	Y. Colón	I. Franco		G. Valtolina	M. Raschke	
10:20 - 10:40						
10:40 - 11:00	Coffee break			Coffee break		
11:00 - 11:20						
11:20 - 11:40	R. González-Férez	J. Owrutsky		C. Toninelli	N. Mueller	
11:40 - 12:00						
12:00 - 12:20	J. Schachenmayer	L. Marcassa		C. Kock	J. Sanz-Vicario	
12:20 - 12:40		T. Antosiewicz			M. Weichman	
12:40 - 13:00	Lunch		8:30 - 18:00 hrs. Group Excursion [Volcano Osorno, Lunch, Petrohue Falls, Boat Tour Lake Todos Los Santos]	Lunch		
13:00 - 13:20						
13:20 - 13:40						
13:40 - 14:00						
14:00 - 14:20	T. Shegai	A. M. Rey		M. Sheldon	Departure	
14:20 - 14:40						
14:40 - 15:00	I. Chi-Duran	S. Will		M. Shatruk		
15:00 - 15:20	A. Belyanin			H. Díaz		
15:20 - 15:40	MQT Poster Session	Coffee break		Coffee break		
15:40 - 16:00						
16:00 - 16:20		J. Anders		T. Li		
16:20 - 16:40						
16:40 - 17:00			W. Xiong		D. Clougherty	
17:00 - 17:20					R. Tichauer	
17:20 - 17:40			B. Simpkins			
17:40 - 18:00						
18:00 - 18:20						
18:20 - 20:00				Conference Dinner at Hotel Radisson (Address: Del Salvador 024, Puerto Varas)		

ORAL

Eric Bittner, University of Houston

A quantum analog of Huygens's clock: noise-induced spontaneous synchronization

Eric R Bittner and Bhavay Tyagi

University of Houston, Dept of Physics, Houston TX

We investigate the quantum dynamics of coupled quantum oscillators interacting with a shared, correlated dissipative environment. The equations of motion for operator moments and covariances are analytically solvable using the Lyapunov equations. Our results reveal that the oscillators relax into a phase-synchronized state under fully correlated or fully anti-correlated environmental noise. This synchronization persists for extended durations when the oscillators are nearly resonant and can last indefinitely when they are in exact resonance. The underlying mechanism is tied to the symmetry of the Lindblad dissipator, which induces firm damping in some areas of the state space and underdamped in others. In the extreme cases of fully correlated or fully anti-correlated environments, specific regions of the state space become entirely decoupled from environmental influence. Additionally, we demonstrate that environmental noise correlations lead to quantum entanglement, with all correlations between the oscillators arising purely from quantum mechanical effects. This work establishes a robust mathematical framework for understanding the connection between long-lived exciton coherences and vibronic correlation effects.



Fig. 1: Correlated noise can act to shield a quantum subsystem from dissipation, much like noise-cancellation headphones.

- [1] E. R. Bittner, H. Li, S. Shah, C. Silva-Acuna, and A. Piryatinski. Correlated noise enhancement of coherence and fidelity in coupled qubits. *Philosophical Magazine*, pages 1–17, 2024.
- [2] E. R. Bittner and B. Tyagi. Noise-induced synchronization in coupled quantum oscillators. arXiv preprint arXiv:2410.22495, 2024.
- [3] B. Tyagi, H. Li, E. R. Bittner, A. Piryatinski, and C. Silva-Acuna. Noise-induced quantum synchronization and entanglement in a quantum analogue of Huygens' clock. *The Journal of Physical Chemistry Letters*, 15:10896–10902, 2024.

José J. Baldoví, Universitat de València

Towards molecular controlled magnonics

José J. Baldoví^a, Alberto M. Ruiz^a, Gonzalo Rivero-Carracedo^a, Andrey Rybakov^a,
Dorye L. Esteras^a, Sourav Dey^a

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The recent isolation of two-dimensional (2D) magnets offers tantalizing opportunities for spintronics, magnonics and quantum technologies at the limit of miniaturization.[1] Among the key advantages of atomically-thin materials are their flexibility, which provides an exciting avenue to control their properties by strain engineering, and the more efficient tuning of their properties with respect to their bulk counterparts. In this presentation we will provide an overview of our recent results on this fascinating topic. First, we will focus on the magnetic properties, magnon dispersion and spin dynamics of the air-stable 2D magnetic semiconductor CrSBr (TC = 146 K)[2] and will investigate their evolution under mechanical strain and Coulomb screening using first-principles.[3] Then, we will introduce the modulation of the magnetic properties, magnon dispersion and spin dynamics of this 2D magnet after the deposition of sublimable organic molecules[4]. Our results predict a modulation of magnetic exchange, a shift in the magnon frequencies and an enhancement of their group velocities up to ~7%. Interestingly, we find a linear correlation between these effects and the donor character of the molecules. This will pave the way for the design of a new class of magnonic materials that can be selectively tailored by a chemical approach.

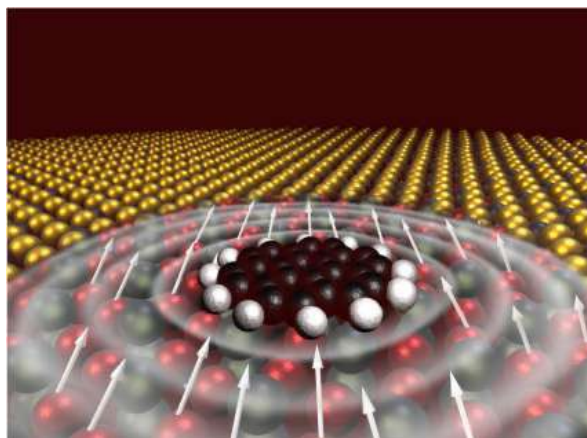


Figure 1. Artistic representation of a spin wave generated below a coronene molecule on the surface of a 2D magnetic material

[1] B. Huang et al., *Nature*, 546 (2017), 270–273.

[2] K. Lee, A. H. Dismukes, E. J. Telford, R. A. Wiscons, J. Wang, X. Xu, C. Nuckolls, C. R. Dean, X. Roy, X. Zhu, *Nano Lett.*, 21 (2021), 3511–3517.

[3] D. L. Esteras, A. Rybakov, A. M. Ruiz, J. J. Baldoví, *Nano Lett.*, 22 (2022), 8771–8778.

[4] A. M. Ruiz, G. Rivero-Carracedo, A. Rybakov, S. Dey, J. J. Baldoví, *Nanoscale Adv.* (2024), DOI: 10.1039/D4NA00230J.

Yamil Colón, University of Notre Dame

Entangled Photon Generation with Metal-Organic Frameworks

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New materials capable of efficiently generating entangled photons can lead to improved efficiency, increased scalability, enhanced signal stability, broadened operational conditions, and lower costs. Metal-organic frameworks (MOFs) are crystalline, nanoporous materials self-assembled from inorganic nodes and organic linkers. The building blocks of these materials offer an ideal materials platform to design and explore fundamental aspects of materials that govern entangled photon generation. In this presentation, I will discuss our efforts to establish a multiscale methodology that leverages quantum chemical calculations for predicting entangled photon generation in MOFs.¹ Then, I will discuss our efforts to determine structure-property relationships linking chemical and material properties to those of the entangled photons.^{2,3} Lastly, I will discuss chemical functionalization strategies and their effects on the optical properties of MOFs.

References

- [1] Fritz, R. A.; Colón, Y. J.; Herrera, F. Engineering entangled photon pairs with metal–organic frameworks. *Chemical Science* **2021**, *12*, 3475-3482.
- [2] Raj, S.; Fritz, R. A.; Herrera, F.; Colón, Y. J. Understanding the Correlation Between Structure and Entangled Photon Pair Properties with Metal–Organic Frameworks. *The Journal of Physical Chemistry C* **2023**, *127*, 10987-10996.
- [3] Raj, S.; Paiva, S.; Fritz, R. A.; Herrera, F.; Colón, Y. J. First-principles screening of metal–organic frameworks for entangled photon pair generation. *Materials for Quantum Technology* **2024**, *4*, 015404.

Rosario González-Férez, Universidad de Granada

Polyatomic ultralong-range Rydberg molecules: Electronic structure and Rydberg blockade

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Rydberg atoms have played a crucial role in the birth of quantum mechanics [1,2], and scattering theory as well as in the development of high precision spectroscopic techniques [3]. Due to their exotic properties, Rydberg atoms are unique probes of their environment, which can be easily controlled and manipulated using electromagnetic fields. In addition, Rydberg atoms also form exotic ultralong-range molecules when combined with ground-state atoms [4,5], ions [6], or polar molecules [7,8], which inherit these exciting properties.

We will explore the interaction of a polar molecule with a Rydberg atom creating a polyatomic Rydberg molecule [8,9]. Our focus is the regime where the charge-dipole interaction of the Rydberg electron with the molecular electric dipole moment induces a coupling between the quantum defect states and the nearest degenerate hydrogenic manifold [8-10]. Based on these non-adiabatic couplings, we propose a protocol to create the Rydberg molecules experimentally in a mixture of ultracold atoms and ultracold molecules [10]. We will also present the first experimental demonstration of the Rydberg blockade due to this charge-dipole interaction between a Rb atom and a RbCs molecule [11]. The atom and molecule are confined in optical tweezers, and for a separation of 310 nm, the charge-dipole interaction between the Rydberg electron and atomic core with the dipole moment of RbCs provokes the blockade of the transition to the Rb Rydberg state. The observed excitation dynamics are in excellent agreement with the theoretical results obtained using the electronic structure of the Rydberg molecule Rb-RbCs [11]

[1] J.J. Balmer, Ann. Phys. Chem. **25**, 80 (1885).

[2] J.R. Rydberg, Philosophical Magazine 5th Ser. **29**, 331 (1890).

[3] T. F. Gallagher, Rydberg Atoms, Cambridge University Press, U.K., (1994).

[4] C.H. Greene, et al, Phys. Rev. Lett. **85**, 2458 (2000).

[5] V. Bendkowsky, et al, Phys. Rev. Lett. **105**, 163201 (2010).

[6] N. Zuber, et al, Nature **605**, 453–456 (2022).

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[9] R. González-Férez, et al, Phys. Rev. Lett. **126**, 043401 (2021).

[10] R. González-Férez, et al, J. Phys. B: At. Mol. Opt. Phys. **53**, 074002 (2020).

[11] A. Guttridge, et al, Phys. Rev. Lett. **131**, 013401 (2023).

Johannes Schachenmayer, CESQ/ISIS, CNRS & Université de Strasbourg

Advancing phase space and matrix product state methods for cavity-coupled molecules

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Theoretically explaining collective cavity-modified physics and chemistry remains to be a challenge for a large number N of coupled molecules. Key mechanism responsible for experimental observations are still lacking a theoretical understanding up to date. In particular, the question about how local chemistry can be modified in a macroscopic limit (with vanishing single-emitter coupling strengths) is important and still open.

This talk will first review how we suggested that disorder and semi-localized [1] dark states can lead to a robust modification of nuclear dynamics, also in the large- N limit in a toy-model molecule setup [2]. Answering the question also for realistic molecules will require more advanced numerical methodologies for large-scale quantum many-body dynamics with long-range connectivity and couplings to an environment. This talk focuses on our recent advances with semi-classical phase space methods [3,4] and with new open-system matrix product state approaches [5], and explains how they could become important for the young research field of polaritonic chemistry.

- [1] T. Botzung, D. Hagenmüller, S. Schütz, J. Dubail, G. Pupillo, and J. Schachenmayer, Phys. Rev. B 102, 144202 (2020)
- [2] D. Wellnitz, G. Pupillo, and J. Schachenmayer, Commun Phys 5, 120 (2022)
- [3] B. Zhu, A. M. Rey, and J. Schachenmayer, New J. Phys. 21, 082001 (2019)
- [4] L. J. Bond, B. Gerritsen, J. Minář, J. T. Young, J. Schachenmayer, A. Safavi-Naini, arXiv:2407.02617 (2024)
- [5] R. Daraban, F. Salas-Ramírez and J. Schachenmayer, Non-Unitarity Optimising Unraveling of Open Quantum Dynamics, [upcoming] (2024)

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

Timur O. Shegai^a

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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]. 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.

- [1] *Nat. Commun.*, 11, 4604, (2020)
- [2] *Laser & Photonics Rev.*, 17, 2200057, (2023)
- [3] *Nat. Photon.*, 18, 751-757, (2024)
- [4] *J. Chem. Phys.*, 154, 024701, (2021)
- [5] *Phys. Rev. Lett.*, 132, 193804, (2024)
- [6] *Nature*, 597, 214-219, (2021)
- [7] *Nat. Phys.*, 19, 271-278, (2023)
- [8] *Sci. Adv.*, 10, eadn1825, (2024)

Ignacio Chi Durán, University of Chicago

Multiplexing Quantum Biosensing with DNA-Functionalized Diamond Surfaces

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^c Department of Physics, The University of Chicago, Chicago, IL 60637

Nitrogen-vacancy (NV) centers in diamonds offer unprecedented sensitivity in detecting magnetic fields and other physical quantities, making them highly promising for diagnostic assays [1]. Over the past decades, sensor technology has trended towards miniaturization, parallelization in arrays, and reduced limits of detection. As a proof of concept, we fabricated an array of 49 distinct DNA spots on a 2x2 mm² diamond surface to detect binding between complementary DNA strands (Figure 1a). Our optimized surface functionalization method immobilized DNA strands at a density of 0.01 DNA/nm², with less than 10 nm separation between the DNA and the quantum sensor layer [2]. Using T1 relaxometry, we detected DNA binding by displacing a partially complementary Gd³⁺-labeled strand. The reduction in magnetic noise near the NV center, indicated by the recovery of NV T1 time, confirmed the specific binding of DNA strands (Figure 1b). Our results demonstrate that the hybridization efficiency of our DNA microarray on the diamond surface is comparable to those reported in the literature. Combining this multiplexed sensing platform with T1-based imaging microscopy techniques presents a convenient route to developing a high-throughput diagnostic quantum sensing device for complex biological samples.

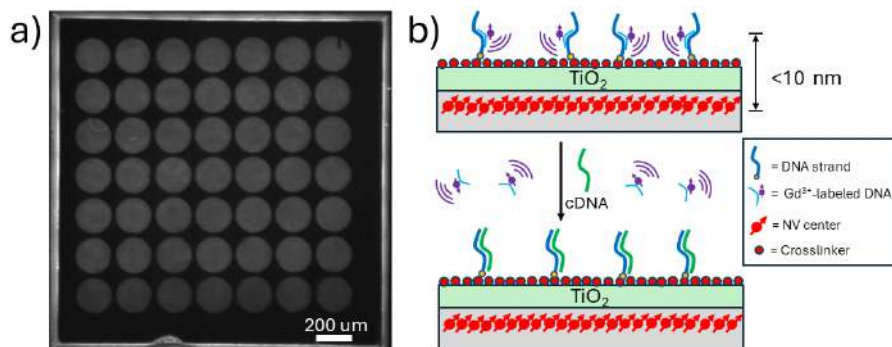


Figure 1. a) Dye-labeled single-stranded DNA on a diamond chip. b) Detection mechanism based T1 relaxometry.

[1] Aslam, Nabeel, et al. "Quantum sensors for biomedical applications." *Nature Reviews Physics* 5.3 (2023): 157-169.

[2] Xie, Mouzhe, et al. "Biocompatible surface functionalization architecture for a diamond quantum sensor." *Proceedings of the National Academy of Sciences* 119.8 (2022): e2114186119.

Alexey Belyanin, Texas A&M University

Quantum gates utilizing dark and bright states of molecules in dissipative cavities

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We present a general formalism and specific implementations of quantum gates based on interaction of light with open dissipative nanocavities containing ensembles of molecules. It is well known that many-body eigenstates in an ensemble of two or more quantum emitters are split into bright and dark entangled states with respect to the coupling to a cavity field [1]. While controlling individual molecules buried in a nanocavity is virtually impossible, one can use a whole cavity loaded with quantum emitters as a logical control qubit which interacts with an external incident photon serving as a flying qubit. The degrees of freedom of a photon (e.g., its polarization) are affected as it scatters or reflects off of a cavity, or interacts with a cavity while propagating in an evanescently coupled waveguide. This interaction can be controlled by changing the state of the molecules in a cavity with classical electromagnetic fields. In one of many possible implementations sketched in Fig. 1, an initial preparation of the state of molecules by a classical optical pulse controls the polarization state of the reflected photon [2].

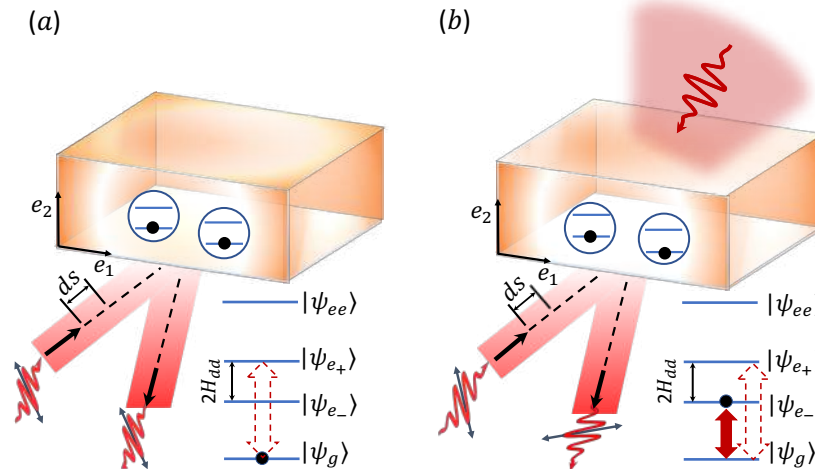


Figure 1. To illustrate one example of the CNOT gate, the polarization of a reflected photon resonant to a cavity mode depends on the state of the molecules which can be prepared by a classical optical field (filled arrow). Dashed arrow shows the cavity mode frequency

[1] M. Tokman et al., Dissipation-driven formation of entangled dark states in strongly-coupled inhomogeneous many-qubit systems in solid-state nanocavities, Phys. Rev. A 107, 013721 (2023).

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Fernando Luis, University of Zaragoza

Circuit QED with Molecular Spin Qudits

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Scaling up quantum processors remains very challenging, even for today's most successful platforms. Molecular complexes, able to encode d -dimensional qudits in their electronic and nuclear spin states, can act as universal quantum processors or even correct errors [1]. I'll discuss recent experiments aimed at exploiting these systems by coupling them to superconducting resonators [2,3]. A high cooperativity coupling to electronic and even nuclear spins has been achieved [3,4]. We also find that it is possible to couple excitations of remote, and distinct, spin ensembles by means of interactions mediated by the circuit. The results provide the basis for the control, readout and communication of spin qubits and qudits and for implementations of quantum algorithms.

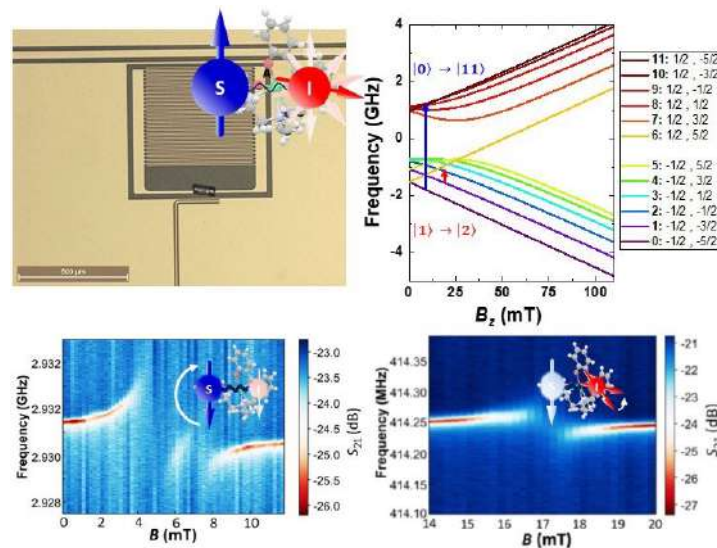


Figure 1 A single crystal of 173Ybtrensals, which hosts an electronic spin qubit and a $d = 6$ nuclear spin qudit, is coupled to a superconducting resonator. Microwave transmission experiments show that the spin-photon coupling reaches high cooperativity, which allows reading out the qudit states.

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Katy Aruachan, Universidad de Santiago de Chile

Atomistic Decoherence of Molecular Spin Qubits

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Magnetic molecules exhibit properties that make them promising fundamental units for quantum processing [1] given their long spin coherence timescales that are comparable with conventional solid-state color centers, and the availability of the chemical synthesis methods to produce crystalline molecular spin qubit arrays. The decoherence and relaxation processes that limit molecular spin coherence have so far been studied with electronic structure methods that are computationally demanding [2]. We developed two models to construct the Redfield tensor that determines the open system dynamics of molecular spin qubits, computing relaxation (T1) and decoherence (T2) timescales over a broad range of temperatures and magnetic fields.

The first model is semi-empirical [3], using a stochastic Haken-Strobl framework with fluctuating molecular gyromagnetic tensors and local magnetic fields, and parametrizing the bath spectral densities using a limited set of T1 relaxation measurements [4]. Taking a vanadium-based spin qubit as a case study, the theoretical predictions agree quantitatively with experiments [4] and represent a solid foundation for the theoretical characterization of other spin qubits. The second model considers spin-lattice and hyperfine spin-spin interaction. It incorporates ab-initio phonon-induced fluctuations of the gyromagnetic tensor from an atomistic model of the copper porphyrin qubit, with hyperfine interaction parameters calibrated from experiments [5]. Low magnetic field relaxation is treated with a phenomenological spin spectral density. The model was successfully tested by predicting T1 and T2 in qualitative agreement with experiments.

We discuss possible refinements of the spin bath spectral density to reach quantitative agreement with available T1 measurements in porphyrin qubits. Our modeling approach can be used for the characterization of quantum magnetometers based on molecular spin qubits.

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Ignacio Franco, University of Rochester

Mapping Decoherence Pathways in Molecules

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To unlock the sophistication of chemistry in building complex molecular architectures to develop next-generation quantum technologies, there is a critical need to identify robust molecular design principles that can be used to generate quantum subspaces with coherences that are protected or that are controllable by chemical means [1]. Systematic progress in this direction requires not only developing experimental and theoretical methods to quantify and manipulate quantum coherences, but also an understanding of *how* the decoherence (or quantum noise) introduced by the environment influences the dynamics of the system and how this influence can be modulated via chemical design. In this talk, I will summarize progress in our group developing strategies to address this problem and map decoherence pathways in molecules [2-4]. These maps quantify the contributions of specific vibrations or solvent modes to the overall dephasing and dissipation of molecular-based quantum subsystems, providing means to establish the basic chemical principles of quantum decoherence phenomena.

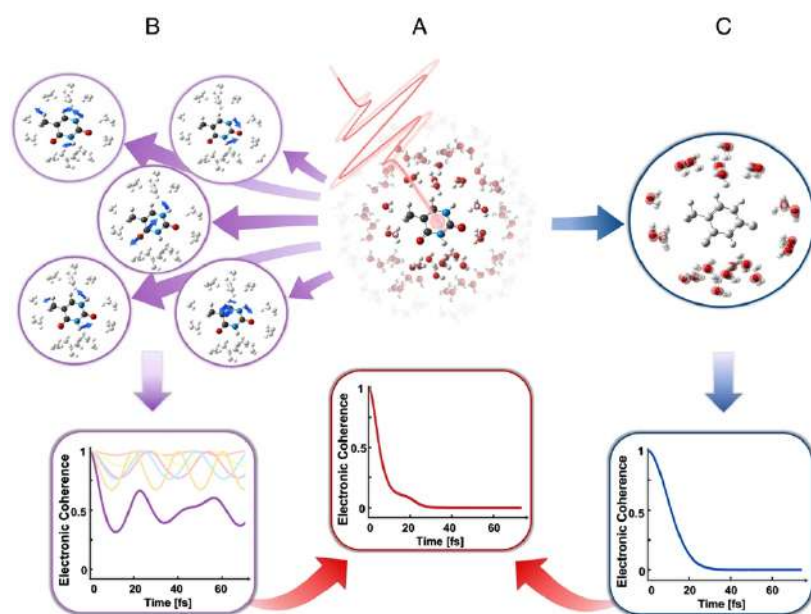


Figure 1. Mapping electronic decoherence pathways. (A) Photoexcitation of molecules creates electronic superposition states with coherences that decay on ultrafast timescales due to interactions of the electronic chromophore with the surrounding nuclei (solvent and intramolecular vibrations). Here we advance a method to quantify the overall decoherence and decompose it into contributions due to specific vibrational modes (B) and solvent (C), thus establishing decoherence pathways that link chemical structure with quantum

decoherence.

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Jeff Owrutsky, Precise Systems & U.S. Naval Research Laboratory

The short life and times of vibrational polaritons

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There is a lot of interest in vibrational polaritons because they appear to modify chemistry.¹ Following the initial studies of infrared spectra of strong coupling to polymer carbonyls and metal carbonyls dissolved in solution,^{2,3,4} the first transient infrared absorption studies were reported for $W(CO)_6$ in hexane which were aimed at identifying modified vibrational dynamics,⁵ especially for vibrationally excited polaritons, that might account for cavity modified reactions. However, clear evidence of modified vibrational dynamics for polaritons, or even convincing demonstrations of clear spectral signatures of vibrational polaritons, remains elusive. Although aspects of the signal were initially attributed to excited state polaritons, the observed transient response is dominated by polariton contraction and enhanced excited state absorption, all phenomena described by linear optical response of the bare molecules. Two dimensional infrared (2D IR) of vibration polaritons from strong coupling to $W(CO)_6$ showed that exciting reservoir, non-polariton states can create a response that includes polariton contraction and enhanced excited state absorption.⁶ Analysis of 2DIR measurements of nitroprusside anion in methanol involved subtracting the reservoir, non-polariton contributions to the signal and indicted that some of the remaining response is from excited polaritons.⁷ This analysis was challenged by the Kubarych group who provided insight that some early time response could be due to inhomogeneities in the molecular absorption band.^{8,9} In order to explain this early time response, a microscopic theory that expanded on a transfer matrix model by including excitation of the reservoir band was developed for the spectroscopy of cavity-coupled molecules that includes band inhomogeneity and can predict 2DIR spectra from molecular polaritons. The theory provides a unified picture for a global understanding of recent spectroscopic experiments on molecular polaritons. There is still the question of whether vibrationally excited polaritons exist and if so, how to reveal them.

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Luis Gustavo Marcassa, University of São Paulo

Multiphoton dressed Rydberg excitations in a room temperature microwave cavity with ultracold Rb atoms

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We investigate magneto-optical trap loss spectroscopy of Rydberg excited ^{85}Rb ($66 \leq n \leq 68 S_{1/2}$) atoms, placed inside a room temperature microwave cavity. The cavity frequency at 13.053 GHz is in resonance with the $67S_{1/2} \rightarrow 66P_{3/2}$ transition, inducing a ladder multiphoton microwave Rydberg absorption and emission. The observed spectra are modeled with an extended Jaynes-Cummings formalism that accounts for multiphoton absorption from and emission into the cavity, the loss from the trap due to Rydberg excitation, and cavity imperfection. We calculate the average photons in each spectral feature and find evidence for fractional photon emission into the cavity modes [1]. The microwave cavity Rydberg spectroscopy in this work should inform the application and technology development of Rydberg based sensors and hybrid Rydberg atom-superconducting resonator quantum gates.

This work is supported by grants 2019/10971-0 and 2021/06371-7, São Paulo Research Foundation (FAPESP), and CNPq (305257/2022-6). It was also supported by the Army Research Office - Grant Number W911NF-21-1-0211. V.R., S.I.M, H.R.S. and S.T.R. acknowledge support from the NSF through a grant for ITAMP at Harvard University.

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Tomasz Antosiewicz, University of Warsaw

Modification of hot carrier spectra in plasmon-molecule systems by optoelectronic strong coupling

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Metallic nanoparticles support plasmon resonances with ultra-small mode volumes and strongly enhanced electric fields which amplify light-matter interactions [1]. We employ real-time TD-DFT simulations to study optoelectronic strong coupling (SC) of magnesium nanoparticles to organic molecules to investigate and focus on two aspects of this interaction.

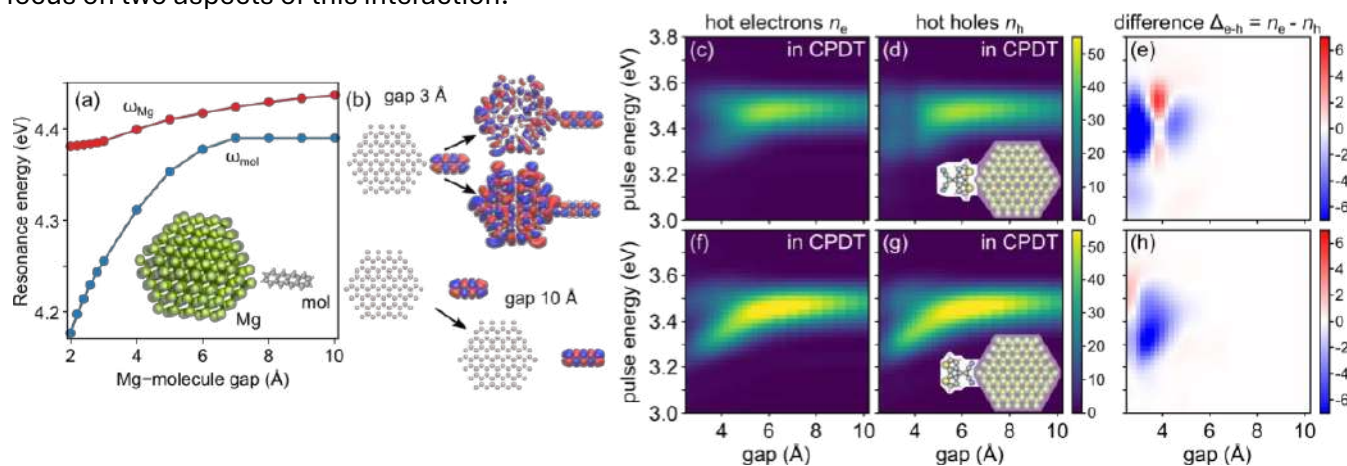


Figure 1. (a) Dependence of macroscopic properties of a SC system stems from (b) appearance of mixed transitions. (c-h) Hot carrier spectra in a Mg-plasmon – CPDT system show qualitative differences with orientation and gap (interaction strength).

We quantify, how macroscopic observables, i.e. as given by the coupled oscillators model, originate from microscopic changes visible on the level of single Kohn-Sham transitions and their energy shifts [2]. These changes are the result of modifications of the molecular oscillator in SC systems, such as the resonance energy redshift, Fig. 1a. They correspond to molecular absorption changes, which are attributed to purely molecular transitions coupling to the many nanoparticle energy states, forming mixed transitions, Fig. 1b. These effects can tailor the polaritonic states and were crucial for novel devices based on SC. Importantly, these changes modify hot carrier dynamics in the upper and lower polariton branches, which can be tuned via the interacting entities, their coupling strength or number. Figure 1c-h shows hot carriers in a SC Mg-CPDT-molecule system [3], where we demonstrate the existence of a SC-mediated charge transfer plasmon whose direction, magnitude, and spectral position can be tuned. We find that the orientation of CPDT changes the nanoparticle-molecule gap for which maximum charge separation occurs, while larger gaps result in trapping hot carriers within the moieties due to weaker interactions. This research highlights the potential for tuning hot carrier generation in strongly coupled plasmon-molecule systems for enhanced energy generation or excited state chemistry.

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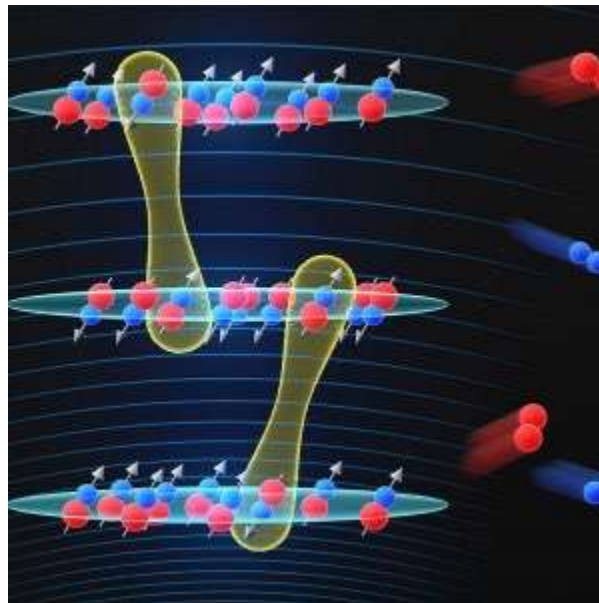
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Ana Maria Rey, University of Colorado Boulder, JILA

New frontiers in quantum simulation with dipolar gases

Ana María Rey
University of Colorado Boulder

Recent experimental developments on cooling, trapping, and manipulating ultra-cold dipolar gases are opening a door for the controllable study of their complex many-body quantum dynamics. In particular, by encoding a spin degree of freedom in rotational levels in polar molecules it is now possible to use these systems to emulate a variety of rich spin models exhibiting long range and anisotropic interactions. In this talk, I will discuss theoretical and experimental progress towards engineering quantum spin models in large molecule arrays trapped in 3D optical lattices relevant for quantum simulations and sensing.



Sebastian Will, Columbia University

Creating and exploring Bose-Einstein condensates of dipolar molecules

Sebastian Will^a

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The creation of Bose-Einstein condensates (BECs) of dipolar molecules has been a long-standing goal in ultracold quantum science. Already soon after the first atomic BECs, it was understood that molecular quantum systems with dipole-dipole interactions would open up novel opportunities for many-body quantum physics. But cooling of molecules to sufficiently low temperatures proved to be exceedingly hard due to strong collisional losses in molecular gases.

Recently, we have created the first BECs of dipolar molecules [1-3]. We evaporatively cool sodium-cesium molecules to below 10 nanokelvin, deep in the quantum degenerate regime. The BECs live for several seconds, reaching a level of stability similar to ultracold atomic gases. This dramatic improvement over previous attempts to cool molecular gases is enabled by collisional shielding via microwave fields, suppressing inelastic losses by four orders of magnitude. The creation of a BEC constitutes the first observation of a phase transition in an ultracold molecular gas.

In this talk, I will discuss our experimental approach, share latest insights, and give an outlook on novel opportunities for many-body quantum physics, quantum simulation, and quantum computing.

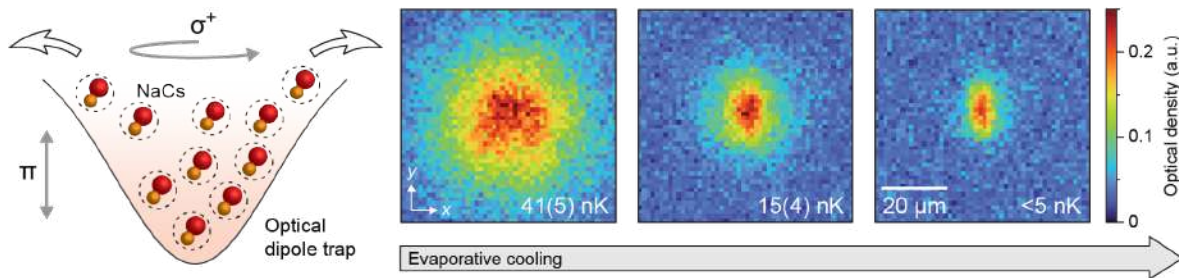


Figure 1. Evaporative cooling of NaCs molecules into a BEC

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Janet Anders, University of Exeter

Impact of system-bath coupling on a quantum system's steady state

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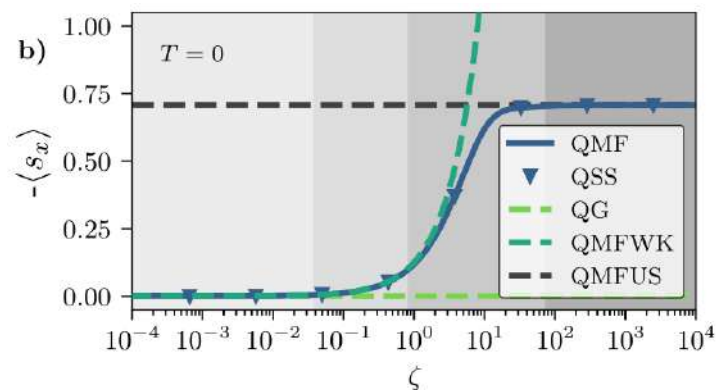
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The dynamical convergence to the Gibbs state is a standard assumption across much of classical and quantum thermodynamics. However, for nanoscale and quantum systems the interaction with their environment becomes non-negligible, and so-called mean force (Gibbs) states are the equilibrium states [1]. Their properties can be important whenever energy is exchanged on the nanoscale, from quantum chemistry to magnetism.

In this talk I will first provide analytical expressions for the mean force state [2] and then discuss the impacts of system-bath coupling for a few examples. These include the presence of steady state coherences [2] in the V-level system interacting with noisy electromagnetic fields [3], and the significant system-bath entanglement emerging when a spin couples to a more realistic 3D environment instead of the commonly studied 1D environment [4]. I will close with a full characterisation of the transition from ultraweak to ultrastrong coupling regime for the widely applicable spin-boson model [5], see figure.

Figure. Characterisation of system-bath coupling regimes from ultraweak (lightest shade) to ultrastrong (darkest shade) for the spin-boson model in steady state at zero temperature.



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Wei Xiong, University of California San Diego

Role of Delocalization to Polariton Chemistry

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Polaritons, quasiparticles resulting from the strong collective coupling of light and matter, have demonstrated remarkable capabilities to alter chemical reactions, energy, and charge transport processes. These features have the potential to revolutionize the control of molecular properties. A key characteristic of polaritons is their delocalized wavefunctions, a hallmark of their behavior.

In many chemical systems, energy disorder is prevalent, and delocalization in polariton systems has been assumed to be robust against this disorder. In our investigation, we examined the criteria necessary to maintain delocalization in molecular polaritons. Contrary to previous assumptions, our study reveals that energy disorder disrupts delocalization in polaritons. To restore delocalization and mitigate the effects of disorder, the collective coupling strength must exceed three times the inhomogeneous linewidth. We further present experimental evidence to show the importance to reach delocalization, in order to achieve polariton-enabled energy transfer and modification to reaction dynamics.

This finding suggests a more stringent criterion for preserving the unique characteristics of polaritons than the conventional standard, which considers collective coupling strengths larger than photonic and molecular spectral linewidths. Moreover, we present a simple linear spectroscopic method to quantify the relative levels of delocalization. This work provides insights into why the onset of modified dynamics exceeds the strong coupling criteria, offering an important measure of polariton delocalization for chemical and materials research under strong coupling.¹

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Blake Simpkins, U.S, Naval Research Laboratory

Materials Design through Quantum Mechanical Coupling

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Sensing processes are simply a material's response to some stimulus. There are many conventional ways to tailor a material's transduction response including nanostructuring to enhance surface effects, chemical functionalization for specificity, optical mode engineering to enhance optical cross-section and/or tune frequency response, or system design to incorporate multi-functionality. In this program, we are interested in basic research aimed at drastic or fundamental alterations of a material's response to stimuli. For instance, introducing quantum mechanical coupling (interaction) in material design is expected to improve material transduction and related sensing functionality through increased sensitivity and improved power efficiency. Utilization of quantum-mechanical interactions as a mode of materials design can take various forms. Coupling of material excitations (e.g., excitons, phonons, vibrations) to optical cavity modes has yielded exciton control, polariton formation and condensation, and opto-mechanical sensors operating in the quantum squeezed regime. Tailored design of molecular excitonic and spin transitions has advanced the interrogation of protein structure and function, and the examination of molecular-cavity optomechanical systems allows one to drive nonlinear population of vibrational excitations, manipulate molecular dephasing, and influence chemical behavior.

Annette Carroll, University of Colorado Boulder, JILA

Tunable Spin Dynamics with Ultracold Polar Molecules

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Ultracold molecules enable exploration of many-body physics due to their highly tunable dipolar interactions. Here, I will review our recent observations of out-of-equilibrium spin dynamics with polar molecules. With spin encoded in the lowest rotational states of the molecules, we realized a generalized t - J model with dipolar interactions [1]. We explored the role of Ising and spin-exchange couplings tuned with dc electric fields and the effect of motion regulated by optical lattices on Ramsey contrast decay. Theoretical understanding of the experimental measurements will also be discussed. Further, we realized XXZ spin models with Floquet engineering [2] and verified that they produced similar dynamics as those controlled by dc electric fields. We additionally used Floquet engineering to realize a two-axis twisting Hamiltonian, inaccessible with static fields, and studied its mean-field dynamics. This work sets the stage for future explorations of exotic spin Hamiltonians with the tunability of molecular platforms.

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Felipe Recabal Rivas, Universidad de Santiago de Chile

Non-canonical steady state of two coupled oscillators in the strong coupling regime

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In a previous work, we describe the suppression of reaction rate inside a cavity where the electromagnetic vacuum is in strong coupling with an ensemble of molecule [1]. We model the system through a master equation where the local-Lindblad terms describe the relaxation processes of the cavity mode and the molecular vibrational modes. We show that local-Lindblad theory is necessary to obtain a non-canonical steady state for the system that describes the reaction rate suppression. Meanwhile, Redfield master equation in the system eigenbasis leads to canonical steady state that does not capture the resonant behavior.

Based on the previous discussion, in the work we microscopically derive a master equation for two coupled harmonic oscillators in the strong coupling regime (see Fig.1(a)). The derivation considers weak coupling and Born-Markov approximation for the system-bath interaction. The master equation obtained contains local terms, that describes relaxation, and non-local terms, due to include the oscillator coupling in the derivation [2,3]. Results shows that the non-local terms are associate to production of coherences induced by the baths, leading the system to a canonical steady state. When the non-local terms are neglected, the system have a non-canonical steady state, with results that agree with the resonant behavior observed using local Lindblad master equation (see Fig.1(b)). Possible ways to neglect the non-local terms are discussed, including tuning temperature and energy detuning, and the inclusion of non-linear system bath interactions.

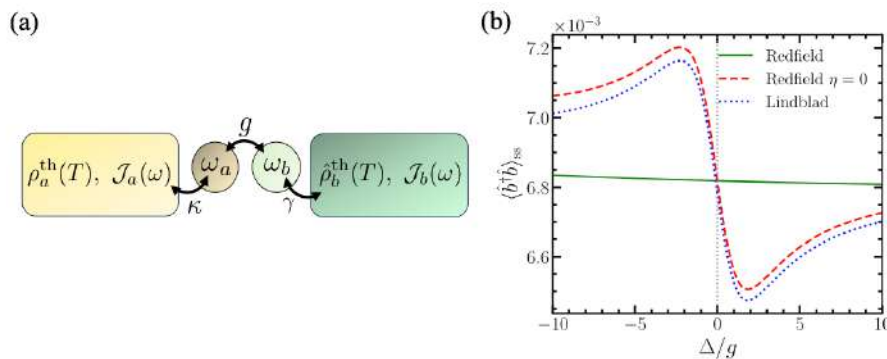


Figure 1. (a) System of two coupled oscillators with system-bath interactions. (b) Steady state occupation as a function energy detuning for Redfield (green line), Redfield with non- local terms (red line) and Lindblad (blue line) master equations.

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Giacomo Valtolina, Fritz-Haber Institute

Towards quantum control of complex cold molecules inside a cavity

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The ability to control chemistry with optical cavities is enticing, but theory and experiment have not yet fully agreed. Cold and ultracold molecules offer near ideal state preparation which can help in identifying the parameter space where cavities can affect chemical reactions. Here, we want to present two different experimental strategies to study polariton chemistry in the gas phase with quantum-state resolution. The first experiment entails the creation of an ultracold molecular gas of dysprosium dimers strongly coupled to an optical cavity. The second focuses on the creation of internally cold molecular ions of dysprosium monoxide. With these platforms, we plan to develop techniques for cavity-control of bimolecular reactions and for photo-ionization and -dissociation of molecular species.

Constanza Toninelli, University of Florence

Organic Molecules in Solids for Photonic Quantum Technologies

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In this contribution, the generation of quantum states of light from single molecules is discussed, tailored to multiple and diverse applications. We will focus on the use of polycyclic aromatic hydrocarbons (PAH), embedded in host matrices [1]. These molecules, due to their small size and well-defined properties, serve as nanoscopic sensors for pressure, strain, temperature, and various fields. The talk discusses recent advancements in coupling single PAH molecules to photonic structures to enhance and control their interaction with light [2]. Notably, two-photon interference experiments between photons emitted by different molecules on the same chip are presented, addressing a fundamental challenge in solid-state platforms for photonic quantum technologies [3]. The experiment relies on multiple milestones, including addressing several molecules simultaneously as on-demand single-photon sources [4], independently tuning their frequencies optically [5,6], and conducting real-time measurements of two-photon interference [3,7]. Additionally, the presentation explores the use of organic molecules as nanoscopic thermal sensors, enabling semi-invasive local temperature measurements in a temperature range (3 K to 30 K) unattainable by most commercial technologies [8]. These results offer insights into the local phononic environment in complex structures and an unexplored temperature regime. Finally, we will comment on the new prospects of using single molecules as interfaces between spin, optical and mechanical degrees of freedom.

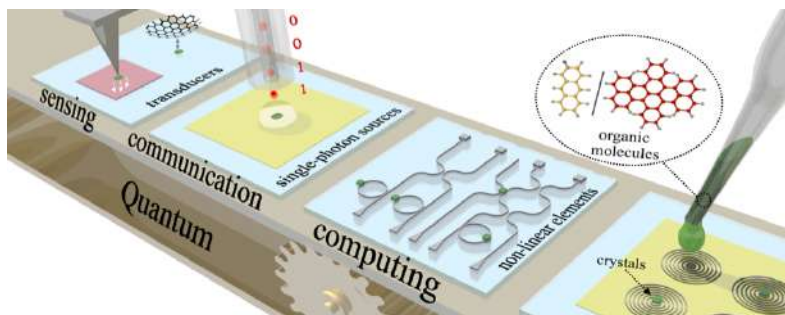


Figure 1. Artistic picture of the different experiments performed with molecules in our Lab

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- [4] P. Lombardi et al., Ad.Q.Tec. 3, (2020)
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Quantum effects in controlled molecular dynamics

Christiane P. Koch

Freie Universität Berlin, Germany

Molecular quantum science and technology hinge on the exploitation of quantum effects. In my talk I will present two such examples. (i) Quantum resonances in low-energy collisions are a sensitive probe of the intermolecular forces. Even for strong and highly anisotropic interactions, they may dominate the final quantum state distribution, as recently observed after Penning ionization of H₂ molecules [1]. Theoretical predictions for the cross sections from full quantum scattering calculations involve only the approximations made when constructing the potential energy surface (PES). Changes in the shape of the PES thus translate directly into modifications of the cross sections. This can be used to improve calculated PES, starting from the experimental data [2]. Conversely, one can also ask by how much the experimental resolution of measured cross sections must improve in order to unambiguously discriminate predictions derived from different levels of *ab initio* electronic structure theory [3]. (ii) Moving from low energy to short time scales, I will discuss the quantum control of photoelectron circular dichroism (PECD) in the photoionization of chiral molecules. Here, the control arises from the interference of various two-photon photoionization pathways that can be manipulated by suitably shaped ionization pulses [4,5]. PECD, remarkably, requires light-matter interaction only in the electric dipole approximation even for randomly oriented molecules. This results not only in a very large dichroic effect, but provides also a recipe for how to induce and subsequently probe chiral dynamics in initially achiral molecules [6]. The preparation of chiral superposition states with a preferred handedness may be useful in future experiments, e.g. on measuring parity violation with chiral molecules.

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Matthew Sheldon, University of California Irvine

Plasmonic Platforms for Polaritonic Chemistry

Matthew Sheldon^a

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We are developing experimental platforms to probe vibrational strong coupling (VSC) between molecules and resonant infrared (IR) nanophotonic architectures, in order to understand how this coupling can fundamentally control chemical reactivity, as well as enable new classes of light-matter interaction. This method of altering the potential energy surface of a chemical process via coherent, electromagnetic perturbation of vibrating bonds has also been termed “polaritonic chemistry”. We employ a combined experimental strategy leveraging expertise in (1) the design of IR “metasurfaces” composed of plasmonic metal substrates that provide tailorable VSC to molecules within their optical near-field; and (2) multiple continuous wave (CW) spectroscopic techniques that enable analysis of several non-equilibrium, dynamic electronic effects in the metal substrate. Taken together, these tools allow studies into new regimes of spectral bandwidth (e.g. simultaneous multi-mode coupling), coupling strength, and time domains (e.g. studies of long lived and steady-state phenomena) that have been inaccessible using conventional optical cavities and time-resolved spectroscopies performed to date.

Vibrational strong coupling is fundamentally interesting because it is a coherent interaction between radiation and molecular motion. The direct manipulation of a molecular process using externally controlled forcefields to obtain a desired outcome, i.e. “coherent control” or “quantum control”, has been a long-standing goal connected to the central aims of chemical science. Thus, this presentation will discuss the limits of chemical analysis and chemical control at interfaces leveraging a framework based on coherent interactions between controllable features of the engineered surface geometry and the molecular systems under study.

[1] Z. Brawley*, S. Pannir-Sivajothi*, JE. Yim*, YR. Poh, J. Yuen-Zhou, M. Sheldon. "Vibrational weak and strong coupling modify a chemical reaction via cavity-mediated radiative energy transfer." *Nature Chemistry* 2024, (accepted, in press)

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[3] Z. T. Brawley, S. D. Storm, D. A. C. Mora, M. Pelton, M. Sheldon. "Angle-Independent Plasmonic Substrates for Multi-Mode Vibrational Strong Coupling with Molecular Thin Films." *Journal of Chemical Physics* 2021, 154, 104305

Michael Shatruk, Florida State University

High-Symmetry Lanthanide Complexes as Clock-Transition Qubits

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Qubit is an elementary unit of quantum computing, allowing coherent superposition of states that can be initialized, manipulated, and read-out for quantum information processing. Molecular spin qubits are appealing due to their synthetic tunability and a broad range of spin states that can be incorporated by using various transition and lanthanide metal ions. The generally short coherence time, however, remains the major obstacle for implementation of molecular qubits in quantum computing technology. In this contribution, we demonstrate how this challenge can be addressed by using high-symmetry lanthanide complexes to achieve clock transitions,^{1,2} characterized by a dramatically enhanced coherence time at specific values of resonant magnetic field. We use variable-frequency EPR spectroscopy to determine the clock-transition energy gaps for the ground-state doublets of crystal-field generated manifolds of $\pm m_J$ states and demonstrate the correlations of the gap magnitude to the geometry of the local coordination environment around the lanthanide ion.

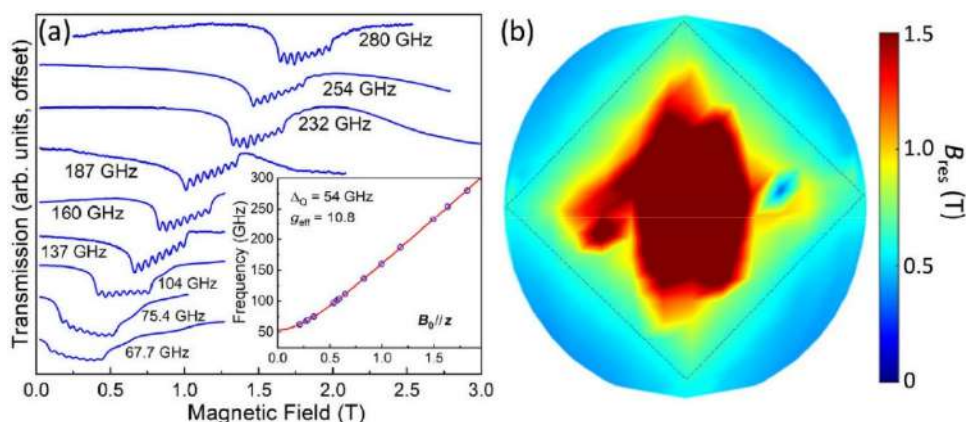


Figure 1. (a) Variable-frequency EPR spectra of a Ho complex that exhibits a quantum clock transition. (b) Spherical plot of the applied magnetic field, B_{res} , at the center of the spectrum at 80.6 GHz, viewed along the 4_1 crystallographic axis.

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Herbert Díaz Moraga, Pontificia Universidad Católica de Chile

Variational Quantum Eigensolver for Estimating the Equilibrium Configuration of Molecules

Herbert Díaz^a, Dardo Goyeneche^a

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With the emergence of new technologies, quantum computing opens up a new world of possibilities, capable of tackling computationally complex problems such as optimization, combinatorial challenges [1], and molecular simulations [2]. One of the current challenges is the noise and intermediate scale reached by quantum computers (NISQ era), where errors associated with quantum phenomena prevent several algorithms from demonstrating their full utility. A way to leverage current quantum computers is through the Variational Quantum Eigensolver (VQE) [3], a quantum-classical algorithm utilized to compute the expectation value of the minimum energy of the electronic structure for molecules. By mapping fermionic systems to qubits, we use this algorithm to obtain the equilibrium energy and geometric configuration of diatomic molecules such as H₂, LiH, and HF, and for the triatomic molecules H₂O and O₃.

Tao Li, University of Delaware

A Tale of Two Methods for Simulating Molecular Polaritons

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Polariton formation between molecular transitions and cavity photon modes provides a novel strategy for modifying local molecular processes. However, our theoretical understanding of many polariton experiments remains elusive. For a realistic simulation of molecular polaritons in the collective regime, here, we introduce two numerical schemes developed in our group: the reduced semiclassical electrodynamics approach [1], and the mesoscale cavity molecular dynamics (CavMD) approach [2]. In the former scheme, only a few molecules, referred to as quantum impurities, are treated quantum mechanically, while the remaining macroscopic molecular layer and the cavity structure are modeled using dielectric functions with Maxwell's equations. In the latter method, a grid of realistic molecular ensembles coupled to many cavity modes are propagated within the framework of molecular dynamics.

Using the reduced semiclassical electrodynamics approach, we study the polariton-induced Purcell effect under electronic strong coupling: the radiative decay rate of the quantum impurity is significantly enhanced by the cavity when the impurity frequency matches the polariton frequency, while the rate can sometimes be greatly suppressed when the impurity is near resonance with the bulk molecules forming strong coupling [1]. Equipped with mesoscale CavMD, we simulate elementary polariton-polariton scattering events under vibrational strong coupling. This approach also facilitates the understanding of vibrational polaritons with broken in-plane translational symmetry [3].

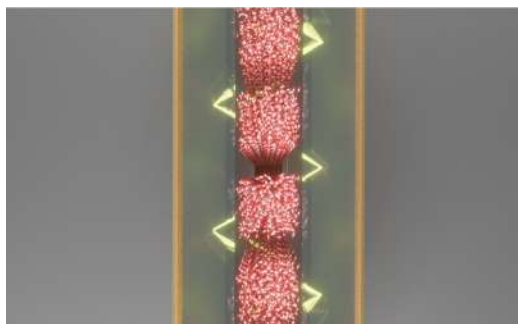


Figure 1. Illustration of the simulation setup: A layer of molecules confined between a pair of parallel mirrors.

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Dennis P. Clougherty, University of Vermont

Variational approach to atom-membrane dynamics

Dennis P. Clougherty

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The adsorption of cold atoms to a surface differs in many ways from the adsorption of atoms at higher energies. It has been established both theoretically [1] and experimentally [2] that the adsorption rate of cold atoms can be dramatically suppressed in comparison to rates at higher energies by two quantum mechanical effects: (1) quantum reflection of the cold atoms away from the surface, and (2) a phonon orthogonality catastrophe [3] resulting from the surface displacement that accompanies adsorption. The first effect is a single particle phenomenon that depends on the wave mechanics of the cold atoms; the second effect is a many-body phenomenon that results from the behavior of the phonon matrix element between the initial and final states of the surface. In the most extreme case of adsorption on a 2D material, it has been proposed that this phonon reduction factor can completely suppress cold atom adsorption [4].

A time-dependent, nonperturbative description of phonon-assisted cold atom adsorption on a membrane will be presented. Using the Dirac-Frenkel variational principle, closed-form expressions for adsorption rates can be obtained. One strength of this method is that the case of intermediate atom-phonon coupling can be treated where the adsorption rate is found to change discontinuously with atom-phonon coupling strength at low membrane temperatures. The framework presented can be customized in a straightforward way to describe a variety of reactions in the quantum regime. Possible applications of these results to emerging quantum technologies will also be discussed.

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Ruth Helena Tichauer, Universidad Autónoma de Madrid

Modelling Light-Matter Interactions with Atomic Resolution

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In addition to their advantageous size, cost-effective and reproducible synthesis [1], the large binding energy of Frenkel excitons makes organic molecules promising candidates for future applications in energy harvesting [2], transport [3-5], and the design of novel quantum devices [6] relying on room temperature polariton formation [7,8]. However, the complex photo-physics taking place in such emitters, challenges conventional quantum optics approaches that consider idealised two-level systems characterised by a well-defined (broadened) resonance.

Employing an atomistic QM/MM description, we develop an approach [9,10] that captures the photo-chemistry within such molecules in the presence of intense and inhomogeneous electromagnetic fields emerging in plasmonic or hybrid metallo-dielectric nano-resonators of arbitrary shape [11,12]. I will introduce the model and share insights concerning the excitation dynamics of systems composed of organic molecules embedded in photonic resonators of various shapes.

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Photonics. 9, 1096 (2022)

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David Leitner, University of Nevada, Reno

Quantum ergodicity and energy flow in molecules

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Under what conditions does a molecule thermalize under its own internal dynamics, and if it does how long does it take? I will discuss a theoretical framework, local random matrix theory, that establishes criteria for quantum ergodicity and energy flow in the vibrational state space of large molecules. I will also discuss some of the ways in which both limitations to and the rate of energy flow in the vibrational state space impact the kinetics of conformational isomerization in gas and condensed phase [1], reactions involving molecules attached to plasmonic nanoparticles [2], as well as thermal conductance of molecular junctions [3], which can now be measured for single molecules [4]. Comparison with results of a variety of experiments will be discussed.

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Micheline B. Soley, University of Wisconsin-Madison

From Ultracold Molecules to Quantum Computing: Collisions Under Quantum Control

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Molecules below one milliKelvin offer an unprecedented opportunity to examine chemical reactions at the level of individual quantum states and to prepare ultracold molecular qubits. A central problem in ultracold physics, which could provide new insight into the development of qubits, is how to simulate the dynamics of ultracold molecules. The long wavelengths associated with low temperatures, coupled with the exponential growth of computational cost as dimensionality increases, currently makes simulation of exact quantum dynamics of ultracold molecules cost-prohibitive for systems of more than three atoms. In addition, anomalous reflection of low-energy components of wavepackets from complex absorbing potentials (optical potentials or perfectly matching layers) has been a problem for decades in quantum scattering and time-dependent calculations, and is especially problematic in ultracold simulations given the systems' low temperature. A solution to this problem could provide a new view into both the dynamics and control of ultracold molecular qubits. In this talk, I will present a novel method that addresses this problem based on the finding that, whereas low temperatures are typically associated with quantum mechanics, classical and semiclassical Wentzel-Kramers-Brillouin corrections can be added to complex absorbing potentials to reduce anomalous reflection by orders of magnitude. This technique offers a possible way to simulate the dynamics of ultracold molecular qubits and ultracold chemical reactions with higher accuracy and computational efficiency.

Quantum control also offers an exciting possibility to create ultracold molecular qubits by directly controlling the outcome of ultracold molecular collisions. In this talk, I will introduce a method that applies reflectionless scattering mode (RSM) theory, recently developed in optics, to direct product formation in chemical reactions with zero reformation of reactants. Surprisingly, the method only requires two variable parameters, which offers an efficient and unique approach to the creation of ultracold molecules in desired quantum states. The talk will discuss how, by combining Wentzel-Kramers-Brillouin force analysis and reflectionless scattering mode theory, one can reveal theoretically the existence of a long-sought-after fundamental physical property - PT-symmetry-breaking phenomena in fundamental quantum scattering - via standard cold-atom experiments in programmable traps.¹ The talk will conclude with a discussion of quantum computing algorithms, both on quantum computers and classical computers via tensor networks/matrix product states.^{2,3}

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Markus Raschke, University of Colorado Boulder

Quantum vibrational nano-imaging: a molecular ruler to image structure, coupling, and disorder in molecular materials

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Properties and functions of molecular materials often emerge from intermolecular interactions and associated nanoscale structure and morphology. However, defects and disorder disturb from energy conversion to carrier transport. Conventional microscopy techniques lack spatial resolution and sensitivity to the low-energy scales of intermolecular interactions and intra-molecular energy relaxation. We address these problems in novel combinations of spatio-spectral and spatio-temporal infrared nano-imaging. Here, coupling between molecular vibrations leads to collective modes, with distinct spectral features sensitive to intermolecular distance and relative molecular orientation. Resolving this vibrational exciton formation as a molecular ruler in IR nano-spectroscopy, we image competing phases and local disorder in molecular solids – information inaccessible by conventional X-ray or electron-based crystallography. In the application to the growth of porphyrin model organic electronic nanocrystals we observe the evolution of defects in competing amorphous and crystalline phases with nanometer spatial resolution [1,5]. Similarly, imaging vibrational coupling in polymers [2] and molecular monolayer [3], we resolve domain formation from the molecular to nano-scale. Further, in another modality through mode selective coupling of vibrational resonances to IR nano-antennas and associated Purcell-enhanced modification of vibrational lifetimes, we resolve intramolecular vibrational interaction and vibrational energy redistribution (IVR) [4]. I will summarize with a perspective for nm-fs resolved precision vibrational nano-spectroscopy for functional imaging in the low-energy landscape of molecular matter.

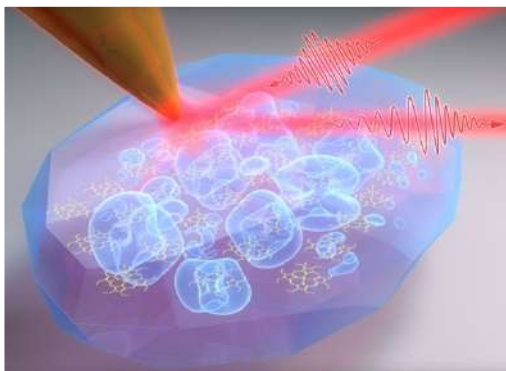


Fig. 1. Infrared nano-imaging of spatial delocalization of the vibrational wavefunction, serving as quantum sensor and molecular ruler of molecular disorder, crystallinity, and intermolecular coupling that control the properties of functional molecular materials at their elementary level.

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- [4] Wilcken et al. PNAS **120**, e2220852120 (2023);
[5] Puro, et al. **24**, 1909 (2024).

Niclas Mueller, Fritz-Haber Institute

Detecting Collective Molecular Vibrations with Surface-Enhanced Raman Spectroscopy

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In surface-enhanced Raman scattering (SERS), vibrations of molecules couple with optical modes of a plasmonic nanocavity via a molecular optomechanical interaction. This effect is typically assumed to occur at individual molecules neglecting intermolecular vibrational coupling. Here, we show instead how collective vibrations are observed in SERS through two different mechanisms: 1. Molecules can couple into collective vibrations through direct infrared (IR) dipole coupling, which is usually observed with IR spectroscopy. We show that cooperative frequency shifts from these collective IR vibrations can be also detected with SERS [1]. These collective states can be tuned with mixed self-assembled monolayers and allow us to monitor photochemical reactions. 2. Molecules can also form collective vibrations through the coupling of induced Raman dipoles when driven with intense laser pulses [2]. We show how such collective Raman vibrations enhance the optomechanical coupling with plasmonic nanocavities and reduce the onset of vibrational nonlinearities.

Finally, I will show recent experiments where we probe the lifetime of molecular vibrations in plasmonic nanocavities with ultrafast Raman spectroscopy [3]. Using time-resolved coherent and incoherent anti-Stokes Raman spectroscopy we separate the contributions of vibrational dephasing and population decay, and isolate molecular signals from four-wave mixing.

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José Luis Sanz-Vicario, Universidad de Antioquía

Ultrafast photodynamics of molecular polaritons analyzed with nonlinear multidimensional spectroscopies

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In recent experiments J-aggregates of organic dye molecules immersed in optical Fabry-Perot microcavities are subject to the strong coupling interaction between the matter and the quantized light field [1]. New entangled polariton states result from the hybridation between molecular states (electronic and vibrational) and cavity photon states. To model the electronic and nuclear molecular structure of an ensemble of molecules inside a cavity and its related photodynamics from first principles is by today's computational capability a formidable task. Parameterized models in quantum optics like the Jaynes-Cummings Hamiltonian for a single emitter, the Tavis-Cummings Hamiltonian for a collective of emitters and the Holstein-Tavis-Cummings (including vibrational states) have served well to understand the fundamental physics of atomic and molecular emitters in cavities. In addition, these dressed-by-light emitters are subject to dissipative processes due to cavity photon losses, intra- or inter-molecular vibrational relaxation, solvent or phonon effects if aggregates are in liquid solution or in solid matrices, respectively. It implies to deal with an open quantum system coupled to a complex thermal bath. In this work we

present a scrutinized study of the ultrafast photodynamics of an ensemble of molecular polaritons by using nonlinear coherent two-dimensional spectroscopy, for which we have recently introduced a very efficient computational method of solution [2] and we show that the role of uncoupled dark states cannot be underestimated.

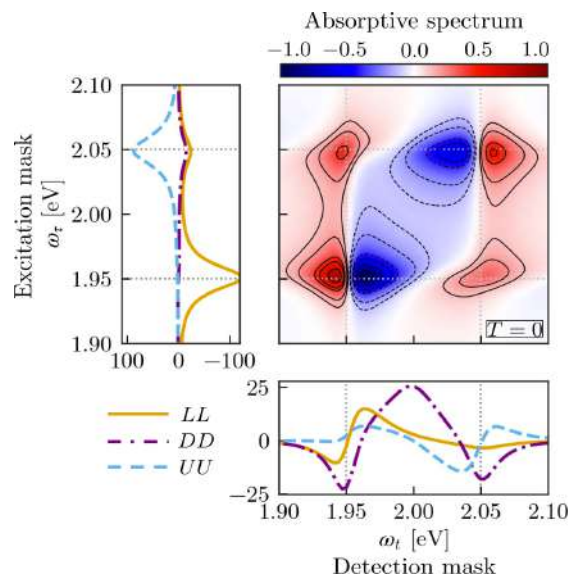


Figure 1. 2D absorptive spectrum for a Tavis-Cummings model built as a sum of products of excitation and mask functions, prior to dissipation due to photon loss and vibrational relaxation. This spectrum displays diagonal peaks for linear absorption-emission processes and cross peaks as a result of nonlinear absorption-emission coherences.

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Marissa Weichman, Princeton University

New Experimental Platforms for Molecular Polaritonics

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Polaritons are hybrid light-matter states that arise from strong interactions between a molecular ensemble and the confined electromagnetic field of an optical cavity. Cavity-coupled molecules appear to demonstrate chemical behavior distinct from their free-space counterparts, but the mechanisms and scope of these phenomena remain open questions [1]. I will discuss new experimental platforms that the Weichman Lab is developing to investigate molecular reaction dynamics under vibrational strong coupling.

While polaritons are now well-established in solution-phase and solid-state systems, they had not been previously reported in gas-phase molecules, where attaining sufficiently strong light-matter interactions is a challenge. We access the strong coupling regime in an intracavity cryogenic buffer gas cell optimized for the preparation of cold, dense ensembles and report a demonstration of strongly coupled rovibrational transitions in gas-phase methane [2,3]. In ongoing work, we will harness this infrastructure as a new testbed for fundamental studies of polariton physics and chemistry [4].

We are also searching for signatures of cavity-altered dynamics in benchmark solution-phase systems [5,6]. So far, we have focused on radical hydrogen-abstraction processes, which have well-characterized reactive surfaces and can be initiated with photolysis and tracked directly on ultrafast timescales. We use ultrafast transient absorption to examine intracavity reaction rates with the goal of better understanding exactly how and when reactive trajectories may be influenced by strong light-matter interactions.

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POSTER SESSION

Ruben Daraban, University of Strasbourg

Non-Unitarity Maximising Unraveling of Open Quantum Dynamics

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In many-body quantum physics, entanglement causes an exponential increase in the computational cost of studying these systems. When open systems are considered, noise can suppress entanglement growth, making the systems more amenable for classical simulations. Recent developments [1] show that optimizing the trajectory-averaged entanglement can enhance the efficiency of pure state trajectory unravelings of open quantum dynamics.

We introduce a novel adaptive unraveling strategy to minimize entanglement, called non-unitarity maximizing unraveling (NUMU) [2]. NUMU is inexpensive compared to other adaptive entanglement-lowering algorithms. We demonstrate its effectiveness through large-scale simulations with random quantum circuits. However, we note that trajectory entanglement remains high compared to the operator entanglement of full density matrix simulations. This disparity indicates that matrix product density operator simulations can be significantly more efficient than the quantum trajectory method in our regimes of interest.

[1] T. Vovk and H. Pichler, Entanglement-Optimal Trajectories of Many-Body Quantum Markov Processes, Phys. Rev. Lett. **128**, 243601 (2022).

[2] R. Daraban, F. Salas-Ramírez and J. Schachenmayer, Non-Unitarity Maximising Unraveling of Open Quantum Dynamics, [upcoming]

Pietro De Checchi, University of Padova

Effects of Different Noise Environments on the Coherence Time of Open Quantum Systems

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The use of Stochastic Schrödinger Equations (SSE) to describe open quantum systems is a well-known class of methods, that can be used as an unravelling scheme of associated Quantum Master Equations and as a starting point to derive new ones. From the perspective of simulating quantum systems in Quantum Computers (QC), one can exploit stochastic averages to implement intrinsically contractive mappings into unitary-gates bases QC since each trajectory is a unitary evolution of the system [1,2] and harnesses the repeated noisy measures in lieu of classical parallelization. Recent studies have shown the ability to engineer environmental effects, in particular concerning non-Markovian environments [3,4]. Such noises can be detrimental to or enhance the coherence time and the transport properties of the system. Here, we present a theoretical description of the differences between using a stochastic process and its derived noise as sources of fluctuations in the stochastic formulation of the dynamics of an open quantum system, and their associated QME.

The starting point is the model presented in [6], leading to non-Markovian quantum evolution in small model systems. Showing the different interpretations, we remark the differences with respect to the usual memoryless approximations and the effects on initial coherence time and stationary distributions.

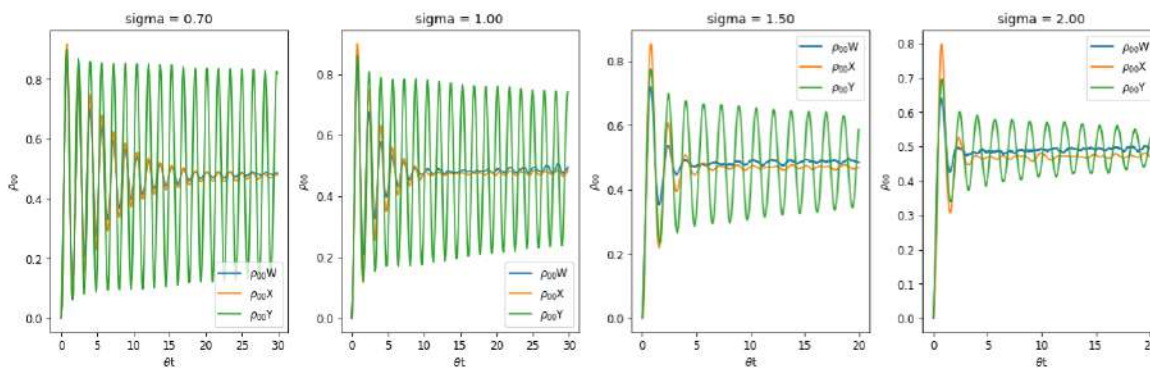


Figure 1. Comparison of the dynamics of population of one site of a two-site system under different noises type and intensities (σ).

- [1] Lloyd, S. (1996), *Science*, 273(5278)
- [2] Hu, Z., Xia, R., Kais, S. (2020), *Scientific Reports*, 10(1)
- [3] Cialdi, S., Benedetti, C., Tamascelli, D., Olivares, S., Paris, M. G. A., Vacchini, B. (2019),

- Physical Review A*, 100(5)
- [4] Carmele, A., Parkins, S., Knorr, A. (2020), *Physical Review A*, 102(3)
- [6] Barchielli, A., Pellegrini, C., Petruccione, F. (2010), *Europhysics Letters*, 91(2)

Rubén Fritz, Universidad de Santiago de Chile

Computational discovery and characterization of MOFs for chemical sensors coupled with fiber optics

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Metal-organic frameworks (MOFs) are not just crystalline materials; they hold immense potential for optical sensing applications [1]. Their unique characteristics, such as porosity, tuneability, and specificity, make them a perfect fit for this field. When coupled with optical fiber (OF), the sensor becomes portable, reusable, and non-destructive. While some single-crystal MOFs coupled with fiber optics systems have shown promising results, the materials used are still limited to a few well-studied examples [2, 3]. Sensitivity to different molecules and the performance of distinct system OF-MOFs upon loading are necessary to develop new sensor devices. The chemical space formed by MOFs is complex and large, but computational methods can efficiently explore and characterize the optical properties, providing an avenue for further design. In this work, we search for suitable MOF candidates for chemical sensors using chemoinformatics methods based on text pattern search of chemical names, compute adsorption isotherms with Monte-Carlo simulation to predict sensitivity limits for adsorbate concentration and the corresponding refractive index change is obtained using periodic density functional theory. We aim to establish the basis for a computational filter and characterization of MOFs for optical sensing of gases in interferometric setups that can be further used in quantum sensing protocols with nonclassical light.

[1]Anik, U., S. Timur, and Z. Dursun, Metal organic frameworks in electrochemical and optical sensing platforms: a review. *Mikrochim Acta*, 2019. 186(3): p. 196.

[2].Zhou, K., et al., Perspective Single-crystal metal-organic frameworks for electronic and optoelectronic devices. *Cell Reports Physical Science*, 2023. 4(11).

[3].Zhu, C., et al., Chemical Detection Using a Metal-Organic Framework Single Crystal Coupled to an Optical Fiber. *ACS Appl Mater Interfaces*, 2019. 11(4): p. 4393-4398.

Felipe Isaule, Pontificia Universidad Católica de Chile

Rotational properties of two interacting ultracold polar molecules

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The last decade has seen rapid progress in the experimental realisation of cold and controlled molecules [1]. Experimentalists have been able to produce degenerate gases of ultracold dimers [2], and have made important progress in the direct cooling of polyatomic molecules [3]. Due to the rich internal molecular structure, these developments have motivated the study of many applications of cold-controlled molecules, such as the realisation of cold dipolar gases [4] and molecular quantum computing platforms [5]. These applications often rely on the polar nature of heteronuclear molecules, which enables their control by electromagnetic fields. In particular, it enables the control of the rotational degrees of freedom and the dipole-dipole interaction between molecules.

Motivated by these recent developments, we have recently studied the problem of two static polar and rotating molecules under the influence of an external dc electric field. In this talk, I will present the main results of this study, including a comprehensive examination of the low-energy spectrum and the polarisation of the dipole moments. In particular, I will show that these properties depend strongly on the direction of the external field and the separation between molecules. These findings should be important to take into account in future studies of polar molecules in many-body and quantum computing applications.

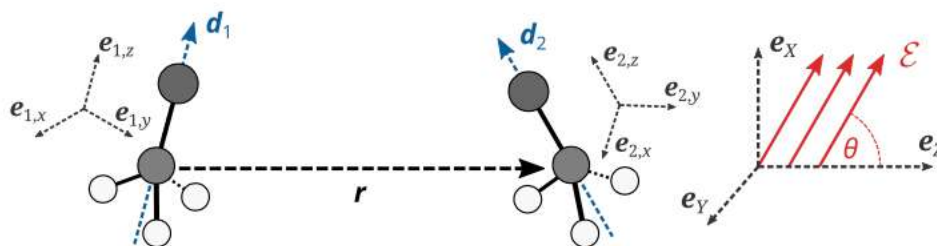


Figure 1. Illustration of the system in consideration. Two rigid rotors with permanent dipole moments \mathbf{d}_1 and \mathbf{d}_2 interacting with an external dc field.

[1] J. L. Bohn, A. M. Rey, and J. Ye, *Science* **357**, 1002 (2017).

[2] L. De Marco, G. Valtolina, K. Matsuda, W. G. Tobias, J. P. Covey, and J. Ye, *Science* **363**, 853 (2019).

[3] T. A. Isaev, *Physics-Uspokhi* **63**, 289 (2020).

[4] S. Moses, J. Covey, M. Miecniowski, D. Jin, and J. Ye, *Nature Physics* **13**, 13 (2017). [5] P. D. Gregory, J. A. Blackmore, S. L. Bromley, J. M. Hutson, and S. L. Cornish, *Nature Physics* **17**, 1149 (2021).

Mateo Londono Castellanos, Stony Brook University

Ultracold long-range Van der Waals Rydberg trimers

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Ultracold Rydberg atoms and dipolar molecules are groundbreaking tools for advancing quantum information protocols [1] and exploring novel quantum phases [2] due to their large dipole moments and long-range interactions [3]. This work demonstrates the potential to create ultracold, long-range Van der Waals Rydberg trimers within a cold Rydberg- molecule hybrid system. Our approach uses a two-photon association scheme that drives atom-diatom collisions into bound Rydberg-diatom states, with estimated photoassociation rates between 10^{-13} and 10^{-11} $\text{cm}^3 \text{s}^{-1}$ and binding energies ranging from 10^{-2} to 10^2 μK . The resulting Van de Waals trimer has lengths of thousands of Bohr radius. This opens the possibility of investigating new quantum complexes in ultracold regimes within current experimental achievements [4].

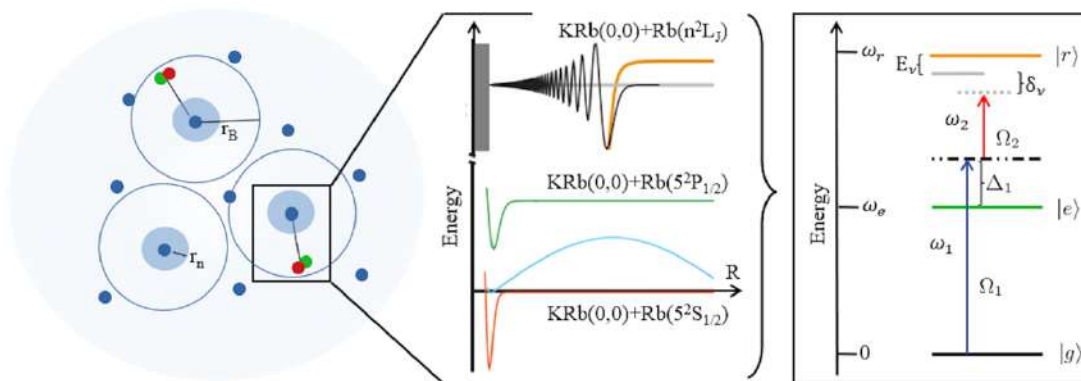


Figure 1. Photoassociation of a Rydberg atom and a diatomic molecule into an ultra-long range Rydberg trimer.

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[2] Karman, T., Tomza, M. & Pérez-Ríos, J. Ultracold chemistry as a testbed for few-body physics. *Nat. Phys.* 20, 722–729 (2024).

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[4] Bigagli, N. et al. Collisionally stable gas of bosonic dipolar ground-state molecules. *Nat. Phys.* 19, 1579–1584, (2023).

Michael A. Michon, U.S. Naval Research Laboratory/ Poster contest awardee

Impact of Cavity Length Non-Uniformity on Reaction Rate Extraction in Strong Coupling Experiments

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^a National Academies of Science NRC Post-Doctoral Researcher

^b Naval Research Laboratory, Chemistry Division

Reports of altered chemical phenomena under vibrational strong coupling (VSC), including reaction rates, product distributions, intermolecular forces, and cavity-mediated vibrational energy transfer, have been met with a great deal of skepticism due to several irreproducible results and the lack of an accepted theoretical framework. In this work, we add some insight by identifying a UV-vis measurement artifact that distorts observed absorption peak positions, amplitudes, and consequently, chemical reaction rates extracted in optical microcavities. We predict and characterize the behavior of this artifact using the Transfer Matrix (TM) method and confirm its presence experimentally. We then present a correction technique whereby an effective molar absorption coefficient is assigned to an absorbing species within the cavity. These revelations have important implications for many existing examples of cavity-modified chemistry and establishing best practices for carrying out robust future investigations.

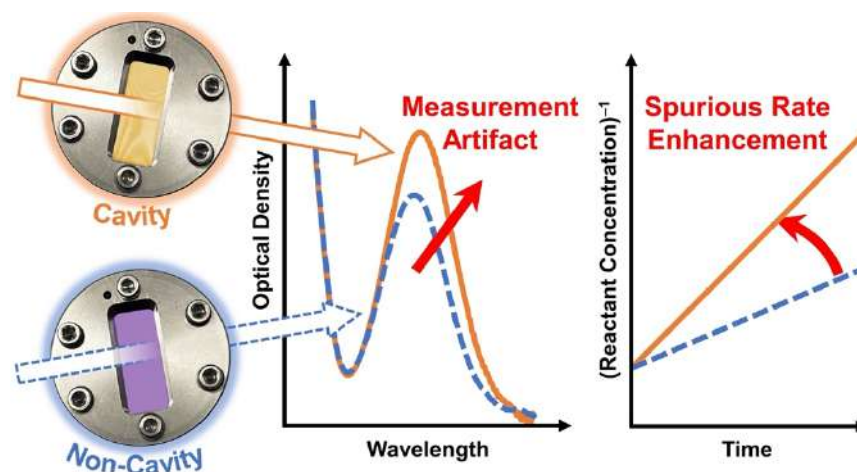


Figure 1. A UV-vis measurement artifact arising from spatial non-uniformity of a Fabry Péro cavity distorts spectral lineshapes and leads to erroneous reaction rate modification.

[1] Michon, M.; Simpkins, B.; Impact of Cavity Length Non-Uniformity on Reaction Rate Extraction in Strong Coupling Experiments. *J Am Chem Soc* **2024** (accepted manuscript)

Joaquín Molina Ulloa, Pontificia Universidad Católica de Chile

Isomerization Reactions in Quantum Computers

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^aPontificia Universidad Católica de Chile

Quantum computers can solve a wide range of computational problems, including molecular optimization and simulations. An effective approach to harnessing the capabilities of quantum computing is the Variational Quantum Eigensolver (VQE), an algorithm that combines quantum and classical resources to determine the minimum energy of molecules. In this work, we apply the VQE algorithm to explore molecular electronic structures, focusing on the simulation of molecular configurations and isomerization reactions by using currently available quantum computers.

Niclas Mueller, Fritz-Haber Institute/ **Poster contest awardee**

Sum-Frequency Spectro-Microscopy to Image Infrared Materials Excitations

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Ryan A. Kowalski^b, Ben John^a, Soeren Wasserroth^a, Guanyu Lu^{b,c}, Joshua D.
Caldwell^b, Piran R. Kidambi^b, Martin Wolf^a, Martin Thamer^a, Alexander Paarmann^a

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^b Vanderbilt University, Nashville, TN, 37235 USA

^c Northwestern University, Evanston, IL, 60208 USA

Nonlinear optical microscopy and spectroscopy are powerful tools to characterize interfaces and lower-dimensional materials. Here, I show two examples how we use infrared + visible sum-frequency generation to image mid-infrared materials excitations with wide-field optical microscopy. The techniques provide combined spatial and spectral information. 1. We visualize the propagation patterns of infrared phonon polaritons in a metasurface of silicon carbide (Fig. 1a-c) [1]. Through a combination of microscopy and spectroscopy, we observe the hybridization and strong coupling of propagating and localized polaritons. 2. We visualize monolayers of hexagonal boron nitride on an insulating substrate (Fig. 1d). This material is usually optically invisible because of its large band gap. Resonant infrared excitation of phonons and heterodyne sum-frequency imaging enable us to image, both, its topography and crystal orientation.

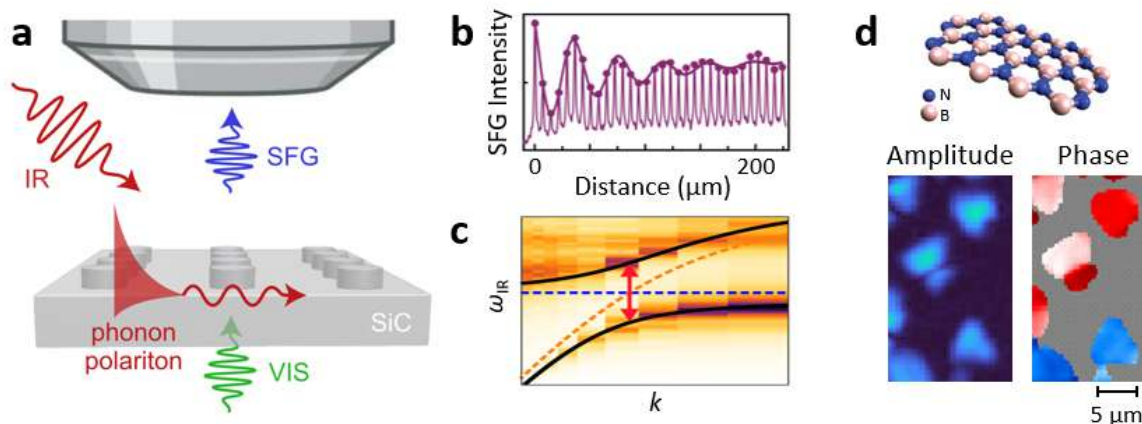


Figure 1. (a) Sum-frequency generation (SFG) spectro-microscopy of a silicon carbide (SiC) metasurface, combining infrared (IR) and visible (VIS) laser excitation. (b) Propagation pattern and (c) dispersion relation of phonon polaritons. (d) Amplitude and phase SFG images of hexagonal boron nitride (hBN) monolayers.

[1] Niemann, Mueller et al. *Advanced Materials* 36, 2312507 (2024)

Michael Reitz, University of California San Diego

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 particular in the limit of large numbers of molecules N coupled to the cavity mode. Our approach is based on a semiclassical, mean-field evolution of the molecular Hamiltonian and the cavity field [1], complemented by a perturbative expansion of both light and matter counterparts in the input pulses entering the cavity. This formalism can be regarded as an extension of traditional free-space nonlinear spectroscopy, now 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 the pump-probe polariton response and in multidimensional coherent polariton spectroscopy [2].

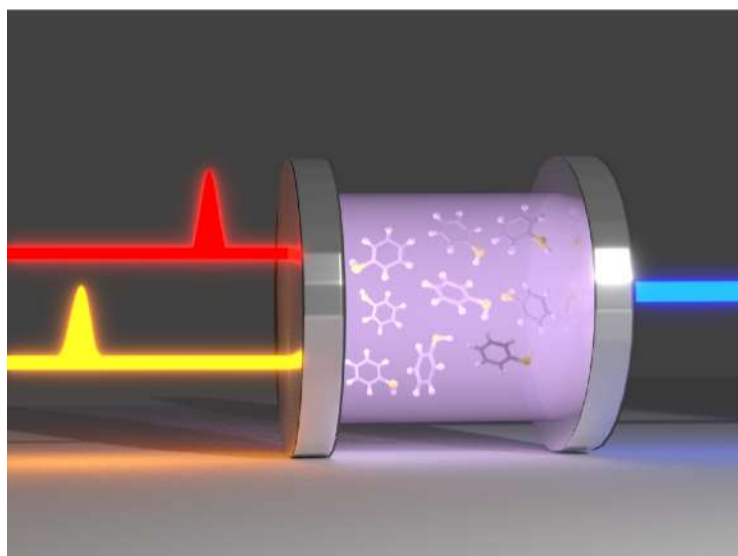


Figure 1. Schematics: An optical cavity containing an ensemble of organic molecules is driven by a set of input pulses.

[1] P. Fowler-Wright, B. W. Lovett, and Jonathan Keeling, *Phys. Rev. Lett.* 129 (2022) [2] M. Reitz, A. Koner, and J. Yuen-Zhou, *in preparation* (2024)

Athul Sambasivan Rema, Universidad de Santiago de Chile

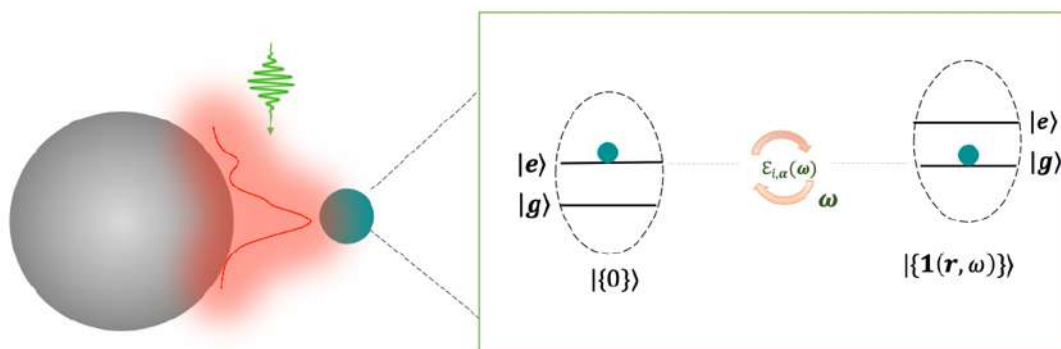
Manipulating the light-matter coupling of a plasmonic nanosphere and a dipole emitter with laser fields

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^aDepartment of Physics, Universidad de Santiago de Chile, Av. Victor Jara 3493, Santiago, Chile

^bMillennium Institute for Research in Optics, Chile

Near field plasmonic resonances that happen in metallic nanostructures provide a fascinating way to engineer the interaction between light and matter [1]. It has been demonstrated that the coupling between the cavity field and matter can lead to strong coupling with a single emitter in plasmonic cavities [2]. We study the strongly coupled dynamical processes of a dipole emitter coupled to near-field modes of a plasmonic nanosphere using macroscopic quantum electrodynamics (QED) and explore the possibility of manipulating the photonic local density of states by driving the system with narrowband laser sources. To achieve this, we develop a pseudo-mode representation of the electromagnetic dyadic Green's tensor of a nanosphere with dipolar and higher-order plasmonic resonant modes, to build semi-analytical solutions of the set of coupled non-Markovian integro-differential equations (IDE) that describe the laser-driven dynamics of material dipoles and photonic degrees of freedom. We solve for experimentally relevant photonic and dipolar observables for a single dipole emitter in a resonant optical nanocavity and compare the results with recent phenomenological Markovian models developed for molecular polaritons [3].



[1] T. W. Ebbesen, *Acc. Chem. Res.* 49, 11, 2403–2412, (2016)

[2] A. Delga, J. Feist, J. Bravo-Abad, and F. J. Garcia-Vidal, *Phys. Rev. Lett.* 112, 253601, (2014)

[3] J. Triana, M. Arias, J. Nishida, E. Muller, R. Wilcken, S. C. Johnson, A. Delgado, M. B. Raschke & F. Herrera, *J. Chem. Phys.* 156, 124110, (2022).

Claudia Reyes-San-Martin, University Medical Center Groningen/ **Poster contest awardee**

Diamond-based quantum sensing for free radical detection in cells and tissues

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^a University Medical Center Groningen, The Netherlands

Diamond-based quantum sensing, through the use of nitrogen vacancy (NV) centers embedded in diamond crystals, has revolutionized the study of cellular biology by providing unprecedented capabilities in free radical sensing [1,2]. This groundbreaking technique, known as relaxometry, leverages the unique properties of NV centers to translate magnetic noise into optical signals, achieving remarkable sensitivity and nanoscale spatial resolution.

The ability of relaxometry to detect free radical production within living cells is pivotal for understanding various biological processes. For example, this technology has illuminated the role of free radicals in sperm development [3] and provided real-time, subcellular monitoring of their involvement in cancer cell migration [4], offering critical insights into metastatic mechanisms.

Moreover, diamond-based quantum sensing paves the way for exploring complex biological samples, including tissues and entire organisms, with NV centers. Despite the potential, challenges such as particle control, reproducibility, and biocompatibility must be overcome to fully harness this technology.

In summary, diamond-based quantum sensing represents a transformative advancement in the detection and characterization of free radicals within living cells, offering profound implications for our comprehension of cellular dynamics, disease progression, and the essential roles of free radicals in biological systems.

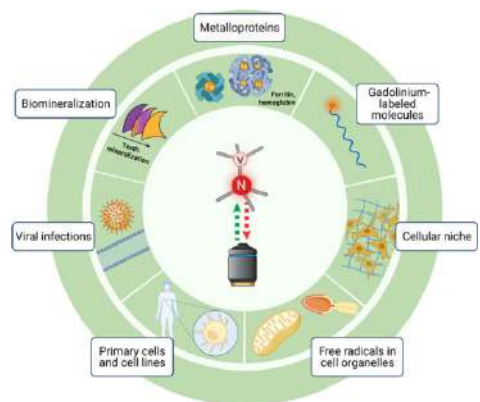


Figure 1. Biomedical Applications of Relaxometry. Relaxometry detects magnetic signals from natural magnetic crystals and metal ions in metalloproteins using NV-based sensing with diamond plates. Gadolinium can label molecules for relaxometry measurements, and nanodiamonds with NV centers can be used for precise detection of magnetic signals, like free radicals, within cells. Reproduced from [1].

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[2] Nie, L., Nusantara, A. C., Damle, V. G., Sharmin, R., Evans, E. P. P., Hemelaar, S. R., Van Der Laan, K. J., Li, R., Martinez, F. P. P., Vedelaar, T., Chipaux, M., & Schirhagl, R. (2021). Quantum monitoring of cellular metabolic activities in single mitochondria. *Science Advances*, 7(21).

[3] Reyes-San-Martin, C., Hamoh, T., Zhang, Y., Berendse, L., Klijn, C., Li, R., Llumbet, A. E., Sigaeva,

A., Kawałko, J., Mzyk, A., & Schirhagl, R. (2022).

Nanoscale MRI for selective labeling and localized free radical measurements in the acrosomes of single sperm cells. *ACS Nano*, 16(7), 10701–10710.

[4] Reyes-San-Martin, C., Elías-Llumbet, A., Escobar-Chaves, E., Manterola, M., Mzyk, A., & Schirhagl, R. (2024). Diamond-based quantum sensing of free radicals in migrating human breast cancer cells. *Carbon*, 228, 119405

Adrián Rubio López, Universidad de Santiago de Chile

Stochastic resonant behaviours and steady state control in harmonic systems

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^b Millenium Institute for Research in Optics (MIRO), Chile.

Brownian motion and parametric resonance are two paradigmatic phenomena particularly taking place on harmonic dynamical systems, covering a plethora of branches in science. While the former gives a pathway to include dissipation and noise (additive noise) in a system, the latter stands for a physical mechanism that supplies energy to a system by exploiting the resonant variation of the characteristic frequency. Both aspects find their syncretism in the so-called stochastic resonance, where the competition between dissipation and the strength of the fluctuations in the characteristic frequency of the system (multiplicative noise) defines whether the system undergoes exponential growth (as in parametric resonance) or stabilises in a steady state in the long-time limit[1]. Typically, the impact of this competition is neglected due to relatively high dissipation rates that overcome resonant effects. However, the development of harmonic systems with increasingly quality factors makes this competition to come into play, raising as a potential limiting factor but also as a possibility for a novel control mechanism. In this talk, I will introduce the basics of the mentioned dynamical phenomena to quantify its impact on experimental setups, such as optically levitated nanoparticle. Moreover, I will also show how these concepts enter interacting harmonic systems, giving place to enhanced resonant behaviours in the steady state. The latter can be exploited, for instance, for heat transport and thermalisation[2].

[1] B. J. West, K. Lindenberg and V. Seshadri, *Physica* 102A, pp. 470-488 (1980).

[2] A. E. Rubio Lopez and F. Herrera, *Stochastic resonant behaviours and steady state control in harmonic systems*, in preparation.

Johan Triana, Universidad Católica del Norte

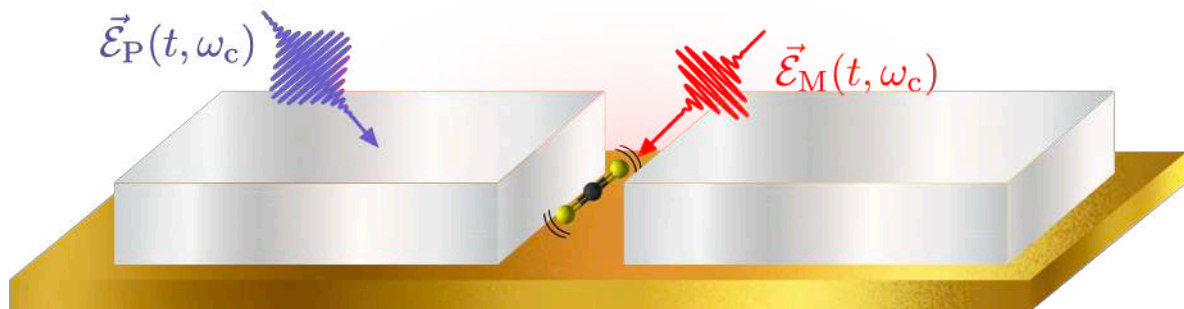
The electromagnetic vacuum enhances infrared photodissociation under strong light-matter coupling

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^a Department of Physics, Universidad Católica del Norte, Antofagasta, Chile

^b Department of Physics, Universidad de Santiago de Chile, Santiago, Chile

Vibrational light-matter coupling is currently one of the most studied research topics due to the unexpected modifications of chemical reaction rates and branching ratios at room temperature. Most theoretical efforts are mainly focused on describing previous experimental measurements in the many-body regime [1], which opens a great opportunity to explore the single-molecule scenario in the full quantum regime [2]. We study the photodissociation dynamics of a single molecular mode coupled to an infrared electromagnetic vacuum in the vibrational strong light-matter coupling for two driven scenarios, *i*) molecule-driving [$\vec{\mathcal{E}}_M(t, \omega_c)$] and *ii*) nanocavity-driving [$\vec{\mathcal{E}}_P(t, \omega_c)$] (see figure). We show a significant enhancement of intracavity dissociation probabilities relative to free space scenarios for the same laser intensities. Similar dissociation probabilities are obtained for the cavity-driving scenario with much lower laser intensities than the implemented in the molecule-driving case [3]. Results are based on the modified ladder climbing process in the polariton quasi-continuum below the dissociation threshold, in comparison with the energy levels scheme of a single anharmonic mode [4]. Our work extends the tools to control dissociation yields of small molecules in confined infrared electromagnetic environments.



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[4] B. Carmeli, A. Nitzan. *The Journal of Chemical Physics*, 72, 2070 (1980).

Houlong Zhuang, Arizona State University

Exploration of New High-Entropy Materials Enabled by Quantum Computing

Houlong Zhuang and Payden Brown

School for Engineering of Matter, Transport and Energy, Arizona State University, Tempe, AZ 85287, USA

High-entropy materials (HEMs) represent a promising category of materials with multi-principal elements and a wide range of molar ratios, offering novel solutions to critical challenges in energy and the environment ranging from climate change to semiconductor chip shortages. Within this material family, high-entropy catalysts, oxides, semiconductors, superconductors, ceramics, and more have gained prominence. The common challenge among these diverse frontiers lies in the selection of elements and their molar ratios across the extensive compositional space. In this talk, we will explore emerging quantum computing technologies, encompassing quantum simulators and hardware, to effectively address the complex task of elemental design and contribute to the discovery of novel HEMs. Furthermore, we will highlight the potential of quantum machine learning algorithms in expediting the training process. Lastly, we outline several prospective directions for HEM research that can benefit significantly from the transformative capabilities of quantum computing.

[1] P. Brown and H. L. Zhuang, *Materials Today*, **63**, 18 (2023).