List of Posters

Bosonic time correlation functions from ring polymer molecular dynamics

Amit Kedar

Tel Aviv University, Tel Aviv

In this work, we propose a method for calculating approximate time-correlation functions of bosonic systems, using ring-polymer molecular dynamics (RPMD). This will allow for a cheap estimation of the time-correlation functions of large bosonic systems, that can be used to evaluate many dynamical quantum mechanical properties, such as diffusion coefficients, chemical reaction rates, dipole absorption spectra, and more.

RPMD uses path integral molecular dynamics (PIMD) to approximate the Kubo- transformed quantum time-correlation functions of distinguishable particles by sampling the classical dynamics of a ring-polymer system whose classical partition function is isomorphic to the quantum system's partition function.

In an earlier work 1 , the first method to perform PIMD simulations of bosons was developed. Using a recursive algorithm, the computational scaling with system size was reduced from exponential to cubic, allowing the first PIMD simulations of large bosonic systems. We intend to use the same method for RPMD calculations of bosons.

So far, we have built the infrastructure needed for the performance of these calculations. We will present the relevant theory, and some benchmark calculations. Next, we will evaluate the time-correlation functions of different bosonic system, and analyze the quality and limits of this method.

The Impact of Fluorine on Structure and Noncovalent Interactions in a Hydrated Cluster

Rami Rahimi

Ben Gurion University, Beer Shava

Exposing the dynamics, structures, and conformational landscapes of isolated biomolecules and the role played by their inter- and intra-molecular interactions with each other and with solvents attracts considerable interest. Here we use the novel laser-based spectroscopic technique of ionization-loss stimulated Raman spectroscopy (ILSRS) and quantum calculations to measure for the first time the vibrational signatures and to reveal the structure of a model hydrated cluster of a fluorinated neurotransmitter. The predicted electronic structure together with noncovalent interaction analysis, provide new insight into electron densities and reduced density gradients, assessing the attractive/repulsive interactions and in-depth understanding of their role in affecting the structure.

The Effect of Cluster Size on the Intra-Cluster Ionic Polymerization Process

Estefanía Molina

Hebrew University, Jerusalem

Infrared Open Cavity-QED of Vibrational Polaritons

Bar Cohn

Technion, Haifa

Cavity Quantum Electrodynamics (Cavity-QED) has recently gained substantial attention due to its capability in altering chemical reactivity; specifically, with the discovery of molecular vibrational strong-coupling modified reactions. Traditional realization of optical Cavity-QED involves discrete photonic modes of a Fabry-Perot cavity coupled to a quantum emitter. However, due to the closed nature of the cavity, only the frequencies of the polaritonic transitions are transmitted, making spectroscopy of the quantum-state-dynamics within the cavity very challenging. We develop a novel open Cavity-QED system based on plasmonic Antenna Lattice Resonances (ALR) of high quality factor operating at mid-IR, providing an excellent platform to study strong light-matter interaction, including Cavity-QED chemistry, Bose-Einstein condensation, ultralow threshold lasing and many more.

Biomolecular Structure of L-Glutathione on “Naked” Silver Nanoparticle Surface

Arghyadeep Basu, Alexander Vaskevich , and Lev Chuntonov

Technion, Haifa

Understanding molecular structure of peptide-functionalized nanoparticles is very important for their efficient use in modern scientific and technological applications. Using $2D$ IR spectroscopy and isotope labeling on cysteine unit of Glutathione, we studied the secondary structure of glutathione (γ-Glu-Cys-Gly,GSH) on silver nanoparticle surfaces.

Molecular structure of peptides on metal nanoparticle surfaces have been the subject of extensive studies. We focused on GSH-capped nanoparticles, which are known for several important applications, such as NP-based drug delivery and amyloid fibril inhibiting agents. Recently, Ghosh et al. reported that GSH, which is made of only three amino acids, self-assembles on the surface of naked silver nanoparticles (AgNP) mostly into stable intermolecular beta-sheet-like structures[1]. Our recent localized surface plasmon resonance spectroscopy (LSPR) experiments revealed that glutathione form $6-7nm$ thick shell on the silver nanoparticle surface[2]. The X-ray photoelectron spectroscopy (XPS) data of the GSH-AgNP suggested that only a small fraction of GSH molecules is bound to silver surface via the thiol bond, forming the first layer whereas the majority of the thiol groups are protonated and remain unbound. We used ^{13}C isotope labeling on the amide carbonyl of the cysteine unit to learn more about the ordered GSH structure on the AgNP surface. Amide I transitions were assigned, $=1643cm^{-1}$ to glutamyl unit and $=1616cm^{-1}$ to the cysteine unit. Moreover, by isotope dilution experiment, local mode frequency of isotope labeled cysteine amide I was determined. This value was used in primitive mode coupling model to calculate cysteine inter-strand coupling $(\ 7cm^{\breve{}1})$. Further experiments with isotope-labeled γ-glutamyl amide will be performed to determine the corresponding inter-strand coupling. Depending on these coupling strengths values, we will be able to better interpret the secondary structure.

References:

1. Ghosh A, Prasad AK, Chuntonov L. Two-Dimensional Infrared Spectroscopy Reveals Molecular Self-Assembly on the Surface of Silver Nanoparticles. J Phys Chem Lett. 2019;10(10):2481-2486.
2. Basu A, Vaskevich A, Chuntonov L. Glutathione Self-Assembles into a Shell of Hydrogen-Bonded Intermolecular Aggregates on "Naked" Silver Nanoparticles. J Phys Chem B. 2021;125(3):895-906.

Open cavities for probing vacuum field chemical reactions

Shmuel Sufrin

Technion, Haifa

Cavity quantum dynamics (cQED) is a developing field which offers a wide range of applications and intriguing phenomena[1]. Among them, great attention is directed to the exotic hypothesis of vacuum field chemistry (VFC) [2]. According to this theory, the existence of a cavity, even if not populated by photons, can alter properties of molecules inside the cavity and even prompt chemical reactions. Although some experimental demonstrations were published, many researchers remain skeptical about the validity of those experiments[3], since in conventional cavities it is challenging to probe the cavity without populating it. Here, we propose the utilization of open cavities, based on plasmonic antenna lattice resonance (ALR) [4]. Using this concept, we fabricated an open cavity with resonant frequency for matching the vibrational C-O stretching mode in DMF (N-dimethylformide) solvent ($1715cm^{-1}$). The cavities were used with solutions of SCN^- in DMF and SCN^- and Li^+Br in DMF, and 2 dimensional infra-red spectroscopy (2DIR) was deployed for measuring the $C-N$ stretching dynamics of those and comparing to the case with no cavity[5]. Although for the first no conclusive result was obtained, we found that the open cavity significantly shorten the vibrational lifetimes of free SCN^- ion and LiSCN contact ion pair, and suppresses the dynamic process of ion pairing beyond the scope of our probing ability. Although more experimental verifications required, these experiments shows some validation of a VFC process, and demonstrates another advantage of the 'behind the curtain view' approach of open cavities.

Reference:

[1] B. S. Simpkins, A. D. Dunkelberger, and J. C. Owrutsky, “Mode-Specific Chemistry through Vibrational Strong Coupling (or A Wish Come True),” J. Phys. Chem. C, vol. 125, 2021, doi: 10.1021/acs.jpcc.1c05362.
[2] F. J. Garcia-Vidal, C. Ciuti, and T. W. Ebbesen, “REVIEW SUMMARY Manipulating matter by strong coupling to vacuum fields,” doi: 10.1126/science.abd0336.
[3] G. D. Wiesehan and W. Xiong, “Negligible rate enhancement from reported cooperative vibrational strong coupling catalysis ARTICLES YOU MAY BE INTERESTED IN,” J. Chem. Phys, vol. 155, p. 241103, 2021, doi: 10.1063/5.0077549.
[4] B. Cohn, K. Das, A. Basu, and L. Chuntonov, “Infrared Open Cavities for Strong Vibrational Coupling,” J. Phys. Chem. Lett. 2021, vol. 12, p. 12, 2022, doi: 10.1021/acs.jpclett.1c01438.
[5] K. K. Lee et al., “Ion-pairing dynamics of Li^+ and SCN^- in dimethylformamide solution: Chemical exchange two-dimensional infrared spectroscopy,” J. Chem. Phys., vol. 134, no. 6, p. 064506, Feb. 2011, doi: 10.1063/1.3552961.

Ramsey-like spectroscopy for chiral molecules in a 3-level system: enantiomer resolved simultaneous measurement

Itay Erez

Technion, Haifa

The mirror image symmetry between enantiomers is broken by the weak interaction. As a result, the spectrum of opposite enantiomers is expected to be slightly shifted, however this effect has never been observed in molecules.
We are developing precision spectroscopy methods to place a bound on the parity violation (PV) shift. To this end, we apply a Ramsey-like scheme using chiral resolution techniques, in order to simultaneously measure the transition frequencies of both enantiomers in a 3-level system. Using this method we can benefit from common mode noise rejection on the PV signal bringing us closer to achieve the standard quantum limit and rejection of certain categories of systematics.

Quantum Calculation of Energies and Life Time of Electron Attachment to Uracil (RNA Base)

Gal Buskila

Technion, Haifa

Absorption of slow moving electrons by neutral ground state nucleobases have been known to produce resonance metastable states. There are indications that such metastable states may play a key-role in DNA/RNA damage. Therefore, herein, we present an ab initio non-Hermitian investigation of the resonance positions and decay rates for the low lying shape-type states of the uracil anion. We employ the resonance via Pad e (RVP) method to calculate these complex properties from real stabilization curves by analytical dilation into the complex plane. This method has already been successfully applied to many small molecular systems and herein we present the first application of RVP to a medium-sized system. The presented resonance energies are converged with respect to the size of the basis set and compared with previous theoretical works and experimental findings. The ability to calculate ab initio energies and lifetimes of biologically relevant systems pave the way for studying reactions of such systems in which autoionization takes place.

Black box automated resonance calculation: automatic RVP

Idan Haritan

Technion, Haifa

Resonances are metastable states with a finite lifetime. They play an important role in a large variety of fields in physics and chemistry. Such as unimolecular and cold molecular reactions, electron scattering from molecules, molecular auto-ionization, photo-induced reactions, surface chemistry, nuclear physics and optics. Non-hermitian quantum mechanics is a formalism that excels in describing these metastable states. It does so by time independent computational algorithms originally developed for the calculations of bound states. Under this formalism, the resonances are described as complex eigenvalues of the Hamiltonian. The real part of the eigenvalue stands for the energy value, whereas the imaginary part is inversely proportional to the lifetime of the state. Nevertheless, calculating complex eigenvalues is not a trivial task.

In this poster, I will present a method that enables calculating these complex eigenvalues from real data of any system. This method, termed RVP – Resonances Via Padé, was implemented on various physical systems, and even helped give birth to two new research fields. The first, calculation of complex energy surfaces and transition dipoles of molecules, highly contributed to the understanding of cold chemistry processes. The second, threshold energies and poles identification for hadron physical problems, was a consequence of a collaboration with an elementary particle physicist.

As a final note, I will present how we were able to take this method further by writing an automated “black-box” code, linked to standard quantum chemistry packages, for the use of chemists in their studies of chemical reactions.