List of Abstracts – Talks

Towards precision spectroscopy of parity violation in chiral molecules

Yuval Shagam

Technion, Haifa

T

Long-time Memory Effects in a Localizable Central Spin Problem

Eran Rabani

University of California at Berkeley

T

Weak- and Strong-Field Coherent Control of a Thermal Collision Pair via Ultrafast Bond Making Control

Zohar Amity

Technion, Haifa

T

Two dimensional infrared spectroscopy of molecules on surfaces of metal nanostructures

Lev Chuntonov

Technion, Haifa

T

Path integral molecular dynamics for indistinguishable particles

Barak Hirshberg

Tel Aviv University, Tel Aviv

Whether particles are bosons or fermions is a most fundamental property of quantum- mechanical systems. It is particularly important for accurately describing, for example,

ultracold trapped atoms, electrons in quantum dots and nuclear spin isomers of hydrogen. Path integral molecular dynamics (PIMD) simulations are widely used to study quantum effects in chemistry and physics. However, they completely neglect this property assuming the particles are distinguishable. We present a new method for simulating indistinguishable particles using PIMD. For

bosons, the main difficulty is enumerating all particle permutations, which scales expo- nentially with system size. We show that the potential and forces can be evaluated using

a recurrence relation that avoids enumerating all permutations while providing the correct thermal expectation values. The resulting algorithm reduces the scaling from exponential to cubic, allowing the first applications of PIMD to large bosonic systems $[1]$. For fermions, the infamous sign problem presents an additional formidable challenge,

limiting applications to moderate temperatures and strongly-repelling systems. By har- nessing the power of free-energy methods, we are able to alleviate the sign problem and

study small systems at temperatures three times lower than using standard PIMD $[2-3]$.

Applications ranging from models of ultracold trapped atoms $[1]$ and electrons in two- dimensional quantum dots $[2-3]$ to a supersolid phase of deuterium under high-pressure

and low temperature $[4]$ will be discussed. I will also present an analysis of the condensate fraction and the role of exchange effects at different temperatures, through the relative probability of different ring-polymer configurations.

References:
1. B. Hirshberg, V. Rizzi and M. Parrinello, Proc. Natl. Acad. Sci. USA (2019) 116 21445-21449.
2. B. Hirshberg, M. Invernizzi and M. Parrinello, J. Chem. Phys. (2020) 152, 171102.
3. T. Dornheim, M. Invernizzi, J. Vorberger and B. Hirshberg, J. Chem. Phys. (2020) 153, 234104.
4. C.W. Myung, B. Hirshberg, and M. Parrinello Phys. Rev. Lett. (2022) 128, 045301.

Molecular Rotors, Batteries, Plasmas and Strong Coupling in the terahertz frequency region

Sharly Fleischer

Tel Aviv University, Tel Aviv

T

HEIBT - a new experimental setup for ion-ion ion-neutral & ion-laser interaction studies

Daniel Strasser

Hebrew University, Jerusalem

I will describe the progress of our new Hybrid Electrostatic Ion Beam Trap (HEIBT) setup, in which by using a new type of "dichroic" electrostatic mirrors we are able to simultaneously trap cation and anion beams. By scaling the beam energy with the ion mass, we can explore low energy cation-anion collisions and image the velocities of coincident neutral products emerging from individual mutual neutralization reactions. Furthermore, the application of the HEIBT for ion-neutral and ion-laser interactions will be discussed. For example by imaging not only neutral products, but also cationic products of laser interaction with trapped anions.

High-Harmonic-Generation: necessarily a representative of extreme-nonliner aptics?

Avner Fleischer

Tel Aviv University, Tel Aviv

T

200 Years after Hamilton: A New Derivation of Classical Mechanics

David Tannor

Weizmann Institute Institute, Rehovot

In the years 1834-1835, Hamilton published two monumental papers that introduced three new formulations of classical mechanics that became the forerunners of quantum mechanics: the Hamilton-Jacobi equation, Hamilton's equations of motion and the principle of least action [1]. Ironically, none of these is what Hamilton was looking for! He was looking for a "magical" function, the principal function , from which the entire trajectory history can be obtained just by differentiation [2]. I will argue that Hamilton's principal function is almost certainly more magical than even Hamilton realized. Astonishingly, all of the above formulations of classical mechanics can be derived just from assuming that is additive, with no input of physics [3]. It appears that analytical mechanics is simply a footnote to the most basic problem in the calculus of variations: that the shortest distance between two points is a straight line.

References:

1. W. R. Hamilton, On a General Method in Dynamics, Philosophical Transactions, Part 2, p. 247 (1834); ibid., Second Essay on a General Method in Dynamics, Part 1, p. 95 (1835).
2. C. Lanczos, The Variational Principles of Mechanics (Oxford, 1949); M. Nakane and C. G. Fraser, The Early History of Hamilton-Jacobi Dynamics 1834-1837, Centaurus 44, 161 (2002).
3. D. J. Tannor, New derivation of Hamilton's three formulations of classical mechanics (preprint); ibid, Duality of the Principle of Least Action: A New Formulation of Classical Mechanics, arXiv:2109.09094 (2021).

Discovery of novel physical phenomena by NHQM (non-Hermitian quantum mechanics).

Nimrod Moiseyev

Technion, Haifa

In quantum mechanics the potentials are real and the atomic $\&$ molecular spectrum as obtained by solving the time-independent Schrödinger equation (TISE) consists of real bound states associated with Hermitian Hamiltonians. Dynamics of electronic and nuclear wave packets are studied by solving the time dependent Schrödinger equation (TDSE).
The NHQM enables us to evaluate closed form expressions for electronic and nuclear dynamical processes by solving the TISE with outgoing boundary conditions. The physical outgoing boundary conditions introduce the non-Hermiticity into the physics of electronic and light scattering from atoms$/$molecules$/$mesoscopic systems.
In my talk I will explain how novel phenomena in electron scattering experiments and light interaction with matter experiments can be discovered solely by NHQM.

What chiral molecules can teach us about quantum control

Christiane Koch

Free University, Berlin

.

Retrospect on I_3^- photochemistry: "Vintage" pump-probe to fs X-ray scattering

Sandy Ruhman

Hebrew University, Jerusalem

.

Methanol di-cation: Coulomb Explosion Story

Esti Livshits - Roi Baer group

Hebrew University, Jerusalem

A striking agreement between ab-initio theoretical predictions and experimental measurements, including branching ratios and channel-specific kinetic energy release, leads to the identification of new mechanisms in Coulomb-explosion (CE) induced two-and three-body breakup processes in methanol.
These identified mechanisms include direct nonadiabatic Coulomb explosion responsible for CO bond-breaking, a complex dynamics forming products such as and and a long-range " Inverse” harpoon mechanism dominating the production of.
These advances are enabled by a combination of recently-developed experimental and computational techniques, using weak ultrafast EUV pulses to initiate the CE and a high-level quantum chemistry approach to follow the resulting field-free non-adiabatic molecular dynamics.
Refs:
∗Luzon, Jagtap,Livshits, Lioubashevski, Baer, Strasser. Phys. Chem. Chem. Phys. 2017, 19, 13488–13495
∗Luzon, Livshits, Gope, Baer,Strasser. J. Phys. Chem. Lett. 2019, 10, 1361–1367
∗Livshits, Luzon, Gope, Baer, Strasser, Communications Chemistry 2020, 3 49
∗Gope, Livshits, Baer,Strasser. Nat Sci. 2021; 1:e10022
∗Barbatti, M. et al. WIREs Comput. Mol. Sci.(2014)
∗Shiozaki, T. WIREs Comput. Mol. Sci.(2018)

Molecular Dynamics using stochastic DFT forces

Roi Baer

Hebrew University, Jerusalem

.

Quantum tomography of Feshbach resonance states

Baruch Margulis – Ed Narevicius group

Weizmann Institute, Rehovot

.

Spectral Tuning in Rhodopsins: Insights from Molecular Dynamics and QM/MM Simulations

Jonathan Church - Igor Shapiro group

Hebrew University, Jerusalem

Rhodopsins are a class of photoreceptor proteins that are ideal candidates for applications such as optogenetics. These proteins obtain their light sensitivity from a covalently bound retinal chromophore, which initiates the biological function of the protein through photoisomerization. Rhodopsins are naturally found in a variety of lifeforms with both microbial and animal origins. These proteins often absorb in the same region as other biological molecules hindering their use as photoswitches in biological fields. Ongoing research in the field has focused on how to rationally design rhodopsins to absorb in the optical window of the body which ranges from red light (650 nm) to the infrared (1350 nm). Here, we focus on proteorhodosin (PR), a rhodopsin that has been found in a wide variety of marine life and is thought to play a key role in solar energy conversion. [1–3] PR has developed a spectral tuning mechanism in order to adapt to the changing wavelength of light at different depths in the ocean. A single amino acid mutation from glutamine (Q) to leucine (L) at position 105 (Q105L) is responsible for a 20 nm spectral shift. [4–7] Typically, color tuning in rhodopsins can be explained using the external point charge model by Honig and coworkers. [8,9] However, the mutation at position 105 is located near the center of the polyene chain of the chromophore and is not well described by the Honig model. We have used state of the art QM/MM simulations with sampling from molecular dynamics for excitation energy calculations to elucidate the spectral tuning mechanism in this protein. We find that the major effect stems from the direct interaction between position 105 with the chromophore. In the case of Q105, the positive partial charge is oriented towards the retinal polyene chain and effectively disrupts the excited state delocalization. Upon mutation to L105 this interaction vanishes, extending the conjugation and leading to a red-shift in the absorption maximum. We derived this mechanism by projecting the electrostatic potential of the protein onto the chromophore. The strategy in this work can be further applied to other systems in order to explain spectral shifts or to guide rational design.

References:

[1] T. Ran, G. Ozorowski, Y. Gao, O. A. Sineshchekov, W. Wang, J. L. Spudich, H.Luecke, Acta Crystallogr. Sect. D Biol. Crystallogr. 2013, 69, 1965–1980.

[2] O. Béja, E. N. Spudich, J. L. Spudich, M. Leclerc, E. F. DeLong, Nature 2001, 411, 786–789.

[3] W. Rudiger, F. Thummler, E. Cmiel, S. Schneider, Proc. Natl. Acad. Sci. 1983, 80,6244–6248.

[4] J. Mao, N.-N. N. Do, F. Scholz, L. Reggie, M. Mehler, A. Lakatos, Y.-S. S. Ong, S. J.Ullrich, L. J. Brown, R. C. D. D. Brown, J. Becker-Baldus, J. Wachtveitl, C. Glaubitz, J.Am. Chem. Soc. 2014, 136, 17578–17590

[5] W. W. Wang, O. A. Sineshchekov, E. N. Spudich, J. L. Spudich, J. Biol. Chem. 2003, 278, 33985–33991.

[6] J. J. Amsden, J. M. Kralj, V. B. Bergo, E. N. Spudich, J. L. Spudich, K. J. Rothschild,Biochemistry 2008, 47, 11490–11498.

[7] Y. Ozaki, T. Kawashima, R. Abe-Yoshizumi, H. Kandori, Biochemistry 2014, 53,6032–6040.

[8] H. Barry, D. Uri, N. Koji, B. N. Valeria, M. A. Gawinowicz, A. Maria, M. G. Motto, J.Am. Chem. Soc. 1979, 101, 7084–7086.

[9] K. Nakanishi, V. Balogh-Nair, M. Amaboldi, K. Tsujimoto, B. Honig, J. Am. Chem.Soc. 1980, 102, 7945–7947.

Control of Uracil anion dissociation via the EP mechanism

Gal Bouskila – Nimrod Moiseyev group

Technion, Haifa

External radiation results in the formation of free electrons that are temporarily trapped by Uracil. The high resonance Uracil anions have enough energy to dissociate Uracil anion (damage of RNA). However, the ground and first excited uracil anions do not dissociate but in time decay to neutral Uracil in its ground electronic state. We will show how by applying a weak chirp laser pulse an asymmetric transition from the high energy uracil anions to the low energy resonances (and not vice versa) occur, minimizing the damage of RNA due to dissociation of Uracil. The asymmetric switch is obtained when the laser parameters encircle a specific value(s) where two of the resonance Uracil anions coalesce to form a special non-Hermitian degeneracy.

Excited state transannular interactions in [2,2]-Paracyclophane - a direct observation using ultrafast femtosecond spectroscopy

Omer Haggag - Sandy Ruhman group

Hebrew University, Jerusalem

.

Comparison of the photoisomerization of three retinal ground states of Bacteriorhodopsin

Partha Malakar - Sandy Ruhman group

Hebrew University, Jerusalem

Photoisomerization of prototypical proton pumping bacteriorhodopsin (BR) is investigated with femtosecond transient absorption spectroscopy. Three retinal ground states, $all-trans-15-anti(AT), 13-cis-15-syn(13C), 13-cis-15-anti(K)$, are studied in the same opsin without altering the protein environment. Continuous illumination of BR with visible light generates a pure AT resting state, whereas dark-adaptation provides the 13C resting-state access. Excitation of AT initiates a photocycle consisting of several distinct intermediates where K is the first groundstate intermediate. The fluorescent state of AT decays with a $0.5ps$ lifetime, but the $13C$ excited state decays within $200fs$. Surprisingly, excited states of K lives more than ten ps. The excited state of K is expected to be shorter-lived than $13C$ because of an increased twist in the retinal backbone. Our mapping in the BR sets a starting point for the theoretical simulation. Initial results obtained with an ongoing collaboration suggested a barrier in the isomerization pathway of K that extends excited state lifespan despite backbone twist. In addition, AT and $13C$ ratio in the dark-adapted BR is $65:35$, which revised the earlier estimates of $40:60$.
Reference:

Wand, A.; Friedman, N.; Sheves, M.; Ruhman, S. Ultrafast Photochemistry of Light-Adapted and Dark-Adapted Bacteriorhodopsin: Effects of the Initial Retinal Configuration. J. Phys.Chem. B 2012, 116 (35), 10444–10452.

TBA

Bar Ezra - Ronnie Kosloff group

Hebrew University, Jerusalem

TBA

Dynamics of driven quantum systems beyond the adiabatic limit

Roie Dann - Ronnie Kosloff group

Hebrew University, Jerusalem

TBA

Inverse Autoionization of Water

Alon Bogot - Daniel Strasser group

Hebrew University, Jerusalem

Water is the most prevalent molecule on Earth and its considered essential for life. One of the interesting processes in pure water is its autoionization according to:

$$eq.1:\ 2H_2O \rightleftharpoons H_3O^+ + OH$$

This equilibrium between water autoionization and the inverse mutual neutralization (MN) reaction determines the pH of pure water. Nevertheless, due to the short timescales and microscopic length scales involved, there are very few direct experimental studies of these isolated reactions[1] that are mainly studied by theoretical simulations[2-5]. The MN reaction can accure via two different processes, electron trasfor or proton transfer. The result of MN through a proton transfer machanism according to eq.1 to the ground state of $2H_2O$ molecules can release up to $9.77eV$ even with a vanishing collision energy[6]. Where this excess energy can be released as kinetic energy, internal product excitation that may result in sequential fragmentation of the water molecules. In contrast, MN by electron transfer, will result in OH and an unstable intermediate H_3O^* that will probably dissociate, for example into H_2O+H.[7-10] Early MN studies assumed that as opposed to the text book eq. 1, isolated molecules would exhibit an electron transfer mechanism.[3]

We will present new experimental data recorded at the DESIREE facility,[1]1 which allows storing and merging velocity matached cation and anion beams. By directly imaging the coincident neutral fragments of individual hydronium and hydroxide reactions at low collision energies we determine the product channels and their kinetic energy release (KER) distributions. Our preliminary analysis indicates both electron transfer and proton transfer occur in isolated molecules and that their relative contribution depends on the collision energy as well as on the internal temperature of the reactents. Demonstrating the crucial importance of the noval (and for the time unique) capability of trapping and cooling the initially hot ions[12] for the study of isolated MN reactions.

References:

[1] J. V. Coe, J. Phys. Chem, 99, 1, 1995
[2] A. Hassanali et al, PNAS, 108 (51), 20410-20415, 2011
[3] P. L. Geissler et al, Science 291, 5511, 2121-2124, 2001
[4] E. Brodskaya et al, J. Phys. Chem. B 2002, 106, 25, 6479–6487, 2002
[5] Hassanali AA et al, Trans. R. Soc. A 372, 2012
[6] P. Maksyutenko et al, J. Chem. Phys. 125, 181101, 2006
[7] A. Neau et al, J. Chem. Phys. 113, 1762, 2000
[8] M. J. Jensen et al, ApJ 543 764, 2000
[9] G.I. Gellene et al, J. Chem. Phys. 81, 5570, 1984
[10] J.E. Mann et al, J. Chem. Phys. 130, 041102, 2009
[11] H.T. Schmidt et al, Rev. Sci. Inst. (2013)
[12] H.T. Schmidt et al, Phys. Rev. Lett. 119, 0730

Molecular Orientation-Induced Second Harmonic Generation: deciphering different contributions apart

Amit Beer - Sharly Fleischer group Tel Aviv University, Tel Aviv

Probing gas-surface and interaction via magnetically manipulated molecular beam

Yosi Alkoby - Yuval Shagam group

Technion, Haifa

The collision of molecules with surfaces and its outcomes is important in wide variety of research fields and applications.
Quantum state-resolved experiments could allow us to study these collisions. By probing the molecule interaction with the surface and using coherent control over the quantum states of the molecule before and after the interaction one can contribute to the understanding of the collision process. Here we demonstrate how by controlling the rotational projection state of a ground state o-H_2 molecule, which determine the alignment and orientation of the rotational motion of the molecule relative to the surface we can extract information about the dependence of the molecule alignment upon scattering.
In particular, we show that by performing molecular-beam scattering experiments from a LiF surface we can experimentally determine a scattering matrix, which governs the behavior of the molecule (its quantum states) after scattering.
Using this technique also allows us as to benchmark models of gas-surface interactions.

Phase and Polarization Effects in HHG from a Diatomic

Jonathan Berkheim - David Tannor group

Weizmann Institute, Rehovot

TBA

Complex Trajectory Approach to High Harmonic Generation. I

Oran Ayalon - David Tannor group

Weizmann Institute, Rehovot

TBA

Complex Trajectory Approach to High Harmonic Generation. II

Noam Ottolenghi - David Tannor group

Weizmann Institute, Rehovot

TBA

Roaming H_2 Dynamics in the Methanol Dication

Krishnendu Gope - Daniel Strasser group

Hebrew University, Jerusalem

Roaming of atomic or molecular moieties is one of the most intriguing examples of molecular dynamics that can give rise to surprising products. In particular, the formation of H_3^+ by ionization of organic molecules has attracted significant experimental and theoretical attention.[1⌣4] By performing time- resolved EUV pump – near IR probe study we can experimentally follow the roaming H_2 dynamics, initiated by single photon double ionization of an isolated methanol molecule.[5] By circumventing the widespread use of strong-field laser pulses for ionization[2,3] we are able to perform a detailed comparison of the experimental data with theoretical ab initio (CASPT2) molecular dynamics simulations that neglect the ionizing laser pulse.[4,5] The multi-faceted agreement between experiment and theory[4] allowed us to describe two competing pathways for H_3^+ formation by proton transfer from the dication to a roaming neutral H_2 molecule as shown in fig. 1.[6] Furthermore, we identified a new long range electron-transfer (LRET) mechanism to the dication from the neutral roaming H_2. Where the kinetic energy released in the resulting Coulomb explosion of the $HCOH^+ + H_2^+$ products provides unique insight into the donor-acceptor distance distribution. In the "traditional" harpoon mechanism, LRET creates two oppositely charged species that attract each other. Here the inverse process occurs: the LRET creates two positively charged moieties that explode due to Coulomb repulsion. We therefore term this mechanism: “inverse” harpooning, a first observation of LRET in dynamics of multiply ionized systems.

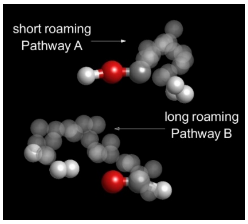

Figure 1: Two competing pathways of H_3^+ formation from methanol dication. 6

References:

1. Mebel, A. M. & Bandrauk, A. D. Theoretical study of unimolecular decomposition of allene cations. J. Chem. Phys. 129, 224311 (2008).
2. Ekanayake, N. et al. H2 roaming chemistry and the formation of H_3^+ from organic molecules in strong laser fields. Nat. Commun. 9, 5186 (2018).
3. Nakai, K., Kato, T., Kono, H. & Yamanouchi, K. Communication: Long-lived neutral H_2 in hydrogen migration within methanol dication. J. Chem. Phys. 139, 181103 (2013).
4. Luzon, I., Livshits, E., Gope, K., Baer, R. & Strasser, D. Making Sense of Coulomb Explosion Imaging. J. Phys. Chem. Lett. 10, 1361–1367 (2019).
5. Livshits, E., Luzon, I., Gope, K., Baer, R. & Strasser, D. Time-resolving the ultrafast H2 roaming chemistry and H3+ formation using extreme-ultraviolet pulses. Commun. Chem. 3, 49 (2020).
6. Gope, K., Livshits, E., Bittner, D. M., Baer, R. & Strasser, D. Two pathways and an isotope effect in H_3^+ formation following double ionization of methanol. Nat. Sci. 1, ntls.10022 (2021).

The pursuit of chiral-molecule candidates for trapped ion spectroscopy via ab-initio calculations

Arik Landau - Nimrod Moiseyev group

Technion, Haifa

Parity violation due to the weak interaction has been predicted to change structure of chiral molecules giving rise to different vibrational constants for each enantiomer among other effects. Precision spectroscopy experiments of molecules have substantially advanced with the development of quantum state preparation and detection techniques and the enhanced sensitivity they often exhibit to physics beyond the Standard Model. Particularly, trapped molecular ion experiments have grown in popularity taking advantage of the ease of trapping charged particles allowing the molecular cation to fine tuned to answer the question at hand.

Herein, we present ab-initio calculations that assist the selection of molecules for parity violation studies. For that, the ionization energies, dissociation energies (of the cations) and excitation energies (of the neutrals) have been calculated. We employ MP2 (second order Møller Plesset perturbation) for optimizing the geometrical structures and CCSD(T) [coupled cluster with singles, doubles and perturbative triples] for energy calculations.These calculations involve molecules with a singlet (closed-shell) and doublet (radicals) multiplicity. Therefore, we carefully monitored these calculations using the EOM-CCSD (equation-of-motion CCSD), which is an appropriate method for treating radical doublet electronic states. We have found that in some cases MP2 does not provide a reliable geometries for the doublet-radicals, although in most cases it does. Thus, our recommended values are based on the EOM-CCSD calculations. We have found a range of suitable candidates, which fulfill these conditions.