

MO 22: Theoretical and Computational Molecular Physics

Time: Friday 11:00–13:15

Location: F142

Invited Talk

MO 22.1 Fri 11:00 F142

A QED Theory of Mediated RET Between a Pair of Chiral Molecules — ●AKBAR SALAM — Department of Chemistry, Wake Forest University, Winston-Salem, NC 27109, USA

A QED [1] theory of relayed resonance energy transfer (RET) [2] between a pair of identical chiral molecules by one or two neutral, passive, electric dipole polarizable bodies, is developed. Migration of excitation energy occurs via the exchange of a single virtual photon between any two interacting particles. Isotropic Fermi golden rule transfer rates are evaluated for direct and bridge-mediated pathways. Rates from both routes depend on the chirality of the donor and acceptor moieties [3-5]. For conveyance of energy to take place indirectly, a multi-level model must be adopted for each mediator polarizability. Furthermore, the rate is found to be a maximum when the three or four particles are collinear. Useful understanding is achieved of propagation of energy between optically active entities in a medium with uniform dielectric constant [6]. [1]*A. Salam, *Molecular Quantum Electrodynamics*, John Wiley & Sons, Inc., 2010. [2]*A. Salam, *Atoms* 6, 56 (2018). [3]*D. P. Craig and T. Thirunamachandran, *J. Chem. Phys.* 109, 1258 (1998). [4]*A. Salam, *J. Phys. Chem. A* 125, 3549 (2021). [5]*A. Salam, *J. Chem. Phys.* 157, 104110 (2022). [6]*J. Franz, S. Y. Buhmann and A. Salam, arXiv:2209.15400 [quant-ph].

MO 22.2 Fri 11:30 F142

Excitation transport in molecular aggregates with thermal motion — ●RITESH PANT and SEBASTIAN WÜSTER — Indian institute of science education and research, Bhopal, India

Molecular aggregates can under certain conditions transport electronic excitation energy over large distances due to the long range dipole-dipole interactions. These interactions are also the characteristics of Rydberg aggregates which have been proved as the quantum simulators for molecular aggregates. An idea that naturally arises in Rydberg aggregates, is adiabatic excitation transport through atomic motion, where slow motion of the atoms combined with excitation transport can result in efficient and guided transport of the excitation from one end of an atomic chain to the other. Based on the analogy between Rydberg- and Molecular aggregates, in ref. [1] we explore whether the adiabatic excitation transport can play a functional role in molecular aggregates in the absence of intra-molecular vibrations. But because the transport is partially adiabatic and because it involves transitions between non-eigenstates, it is challenging to estimate the adiabaticity of transport in molecular aggregates. Hence, in ref [2] we established a measure to quantify the adiabatic character of quantum transitions in general. Next, the effect of intramolecular vibrations is included by extending our calculation for excitation transport to an open-quantum-system technique [3].

[1] R. Pant and S. Wüster, *Physical Chemistry Chemical Physics* 22, 21169 (2020). [2] R. Pant, et al., <https://arxiv.org/abs/2007.10707>. [3] R. Pant, et al., (Manuscript in preparation)

MO 22.3 Fri 11:45 F142

Rotational and vibrational decay of diatomic molecules near a surface — ●NICOLAS SCHÜLER, OMAR JESÚS FRANCA SANTIAGO, and STEFAN YOSHI BUHMANN — Institute of Physics, University of Kassel, Germany

We study the rotational and vibrational motion of chiral diatomic molecules near a dielectric surface [1]. We use macroscopic quantum electrodynamics [2] to obtain the decay rate of the molecule. We determine the critical distances at which retarded (nonretarded) interactions with the surface become dominant, which happens when the distances are much larger (smaller) than the atomic transition wavelengths, respectively. We explore potential links of the rotational and vibrational dynamics with quantum friction.

References:

[1] Stefan Yoshi Buhmann, M. R. Tarbutt, Stefan Scheel, and E. A. Hinds, *Phys. Rev. A* 78, 052901 (2008).

[2] D.T. Butcher, S.Y. Buhmann, and S. Scheel, *New J. Phys.* 14, 113013 (2012).

MO 22.4 Fri 12:00 F142

Medium-assisted chiral discrimination via resonance energy transfer — ●JANINE CHIRSTINE FRANZ¹, STEFAN YOSHI

BUHMANN², and AKBAR SALAM³ — ¹University of Freiburg, Germany — ²University of Kassel, Germany — ³Wake Forest University, Winston-Salem, USA

Resonance energy transfer between two chiral molecules can be used in principle to discriminate enantiomers [1]. Using macroscopic quantum electrodynamics, we study how to enhance and control this effect by means of an intervening medium and propose a distinct, but related way to discriminate between enantiomers by using a medium with known chirality [2].

When embedding molecules in a macroscopic medium, the microscopic structure of the medium close to the embedded molecule needs to be taken into account; we achieve this by using the so-called local-field correction. We present the influence of a magneto-(di)electric medium surrounding the energy-exchanging molecules, where exotic media have a non-trivial impact on the degree of discrimination [3]. When considering a chiral medium that takes an active part in the chiral discrimination, we find that considering local-field effects results in a surprising prediction for the discrimination.

[1] D. P. Craig et al., *J. Chem. Phys.* 109, 1258 (1998)

[2] S. Y. Buhmann et al., *New J. Phys.* 14 083034 (2012)

[3] J. Franz et al., arXiv:2209.15400 [quant-ph]

MO 22.5 Fri 12:15 F142

Analysis of the dopamine D2 receptors interaction with three radiopharmaceuticals using molecular dynamics techniques — NASTASIA-SANDA MOLDOVEAN-CIOROIANU, ●DIANA-GABRIELA TIMARU, and VASILE CHIS — Babes-Bolyai University, Faculty of Physics, Cluj-Napoca, Romania

It is widely known that the dopamine neurotransmitter and its receptors play a pivotal role in neuronal signal transduction and other metabolic processes. Using molecular dynamics (MD) techniques, this study seeks to find and describe the optimal binding positions between the D2 dopamine receptor (D2DR) and three [11C]-labeled synthetic compounds: [11C]-FLB 457, raclopride ([11C]-RACL), and halobenzazepine ([11C]-SCH).

Previous research has demonstrated that the FLB 457 molecule has a high affinity for the D2 dopamine subunit. In addition, [11C]-tagged RACL is already employed as a PET scan radiotracer.

Our analysis showed, in good agreement with experimental results, that RACL ligand bound at the upper portion of the D2 dopamine receptor had the highest interaction energy patterns. In addition, these discoveries pave the path for future research. Perspectives include more MD studies as well as thorough production time and free energy estimates. The same set of radiopharmaceuticals will be docked in the receptor's top pocket, with the D2DR structure encased in a DOPC/phospholipid bilayer membrane.

MO 22.6 Fri 12:30 F142

Molecules for photodynamic therapy. Photophysical properties of 1O2 and 1,3-diphenylisobenzofuran — ●STEFAN STAN and VASILE CHIS — Babes-Bolyai University, Faculty of Physics, Cluj-Napoca, Romania

The first part of the study focuses on the calculation of the excitation energies of the O2 molecule in the first and second excited states, using the [8,6]-CASSCF method and the complete basis set extrapolation technique. The calculated energies are 0.983 and 1.671 eV, in excellent agreement with the available experimental data (0.98 and 1.63 eV). As a result of the transition to the first excited state, the DPBF molecule is significantly planarized, and the bond lengths undergo significant changes only in the benzofuran group.

Several density functionals were tested for the reproduction of the photophysical parameters of the molecule, and the results clearly show that the cam-B3LYP functional provides the best agreement with the experimental data, for all photophysical parameters.

The first excited state of the molecule is due to the $\pi-\pi^*$ transition between the HOMO-LUMO orbitals. The transition was described based on the natural transition orbitals and the difference between the electronic densities corresponding to the excited state and the fundamental one. An excellent agreement has been obtained between the experimental radiative lifetime (5.52 ns) and the calculated calculated counterpart (5.34 ns) at PCM(DMSO-cam-B3LYP/6-311G(2d,p)//APFD/6-311+G(2d,p) level of theory.

MO 22.7 Fri 12:45 F142

Gas phase sugar sythesis: the formation of glycolaldehyde —
•WEIQI WANG¹, XIANGYUE LIU¹, and JÉSUS PÉREZ-RÍOS² — ¹Fritz-Haber Institute, Max-Planck Society, Berlin — ²Stony Brook University, Stony Brook, New York

Gas-phase sugar synthesis is one of the most important reactions for the understanding of prebiotic chemistry in space. In this work, ab initio molecular dynamics has been employed to study the formation of glycolaldehyde from protonated formaldehyde, which is the first step of the carbon chain growth reaction. In particular, the isomerization of neutral formaldehyde molecule has been discussed. The reaction networks between neutral formaldehyde and protonated formaldehyde has been constructed to understand the formation of glycolaldehyde.

MO 22.8 Fri 13:00 F142

Kinetic and thermodynamic theory study of sequential reacti-

on of Koop type hydroamination and [3 and 2] Meisenheimer rearrangement — •MORTEZA FARAHANI — Department of Physical Chemistry, Islamic Azad university, Malayer Branch, Malayer, Iran Farahanijokar@yahoo.com

Koop Norborne type hydroamination using N-allyl-N-methylhydroxylamine at 120 °C and tetrahydrofuran solvent leads to the production of unstable dipolar N-oxide intermediate which turns into a neutral product with higher stability after [3 and 2] Meisenheimer rearrangement. The kinetics and thermodynamics of this sequential reaction have been studied at the computational level of MN15L/aug-cc-pVTZ. The first stage of the reaction is exothermic and non-spontaneous, while the second stage is exothermic and spontaneous. The adverse effects of entropy and high temperature make the rate constant of the first stage, despite the lower activation energy, smaller than the second stage, and the balance is not towards the production of the dipolar N-oxide intermediate.