SYCC 1: Controlled Molecular Collisions

Time: Wednesday 11:00–13:00 Location: Paulussaal

Invited Talk SYCC 1.1 Wed 11:00 Paulussaal
Dynamics of CO_2 activation by transition metal ions - The importance of intersystem crossing — •Jennifer Meyer — Fachbereich Chemie und Forschungszentrum OPTIMAS, RPTU Kaiserslautern-Landau, Kaiserlautern, Germany

Understanding chemistry at the level of a reactive collision, that is how atoms rearrange during the reactive event, is a fundamental question for physics and chemistry. The energetics along the reaction coordinate are important and the influence of barriers is often used to predict the outcome of a reaction. However, in small systems, especially in gas phase, submerged barriers with respect to reactants can exert a profound influence.

Here, we present a joint experimental and theoretical study on the possible effects of intersystem crossing on the dynamics of transition metal ion-molecule reactions and determine the nature of the bottleneck for $Ta^+ + CO_2$. Recent crossed beam imaging experiments in our group on the dynamics on oxygen atom transfer (OAT) reaction $Ta^+ + CO_2 \rightarrow TaO+ + CO$ showed dominantly indirect dynamics despite the thermal rates being close to collision rate and the reaction being highly exothermic. The question to the nature of the bottleneck along the reaction coordinate arose: a submerged transition state or the intersystem crossing. A combination of differential cross sections, thermal rate constants and high level theory for the OAT for Ta^+ and its lighter homologue niobium Nb^+ could shed some more light on the question.

Invited Talk SYCC 1.2 Wed 11:30 Paulussaal
Angular momentum of small molecules: quasiparticles and topology — •Mikhail Lemeshko — Institute of Science and Technology Austria (ISTA), Am Campus 1, 3400 Klosterneuburg, Austria

I will present our recent findings on small molecules kicked by laser pulses. First, I will describe a technique that allows to probe highly excited molecular states in the presence of an environment, such as superfluid 4He, and a theory based on angulon quasiparticles that describes such states, in good agreement with experiment. Second, I will show how that even the simplest of existing molecules - closed-shell diatomics not interacting with one another - host topological charges when driven by periodic far-off-resonant laser pulses. A periodically kicked molecular rotor can be mapped onto a "crystalline" lattice in angular momentum space. This allows to define quasimomenta and the band structure in the Floquet representation, by analogy with the Bloch waves of solid-state physics. We predict the occurrence of Dirac cones with topological charges, protected by reflection and time-reversal symmetry. These Dirac cones – and the corresponding edge states – are broadly tunable by adjusting the laser strength and can be observed in present-day experiments by measuring molecular alignment and populations of rotational levels. This paves the way to study controllable topological physics in gas-phase experiments with small molecules as well as to classify dynamical molecular states by their topological invariants. [1] I. Cherepanov et al. Phys. Rev. A 104, L061303 (2021); New J. Phys. 24 075004 (2022) [2] V. Karle et al. Phys. Rev. Lett. 130, 103202 (2023); arXiv:2307.07256 (2023)

Invited Talk SYCC 1.3 Wed 12:00 Paulussaal
Manoeuvring chemical reactions one degree of freedom at a time — •Jutta Toscano — University of Basel, Switzerland

The combined use of electric fields, magnetic fields and laser light affords us an ever-increasing level of control over the properties of atoms and molecules, enabling reactivity to be probed as a function of their various degrees of freedom [1].

Here, we discuss how electrostatic deflection [2] can be employed to disentangle the reactivity of molecules in different rotational states, or with different spatial orientation of their constituent atoms [3]. Furthermore, we demonstrate for the first time the sympathetic cooling of different conformational isomers within a Coulomb crystal [4], setting the scene for fully conformationally selected ion–molecule reaction studies.

[1] J. Toscano et al., PCCP 22, 9180 (2020)
[2] Y.-P. Chang et al., Int. Rev. Phys. Chem. 34, 557 (2015)
[3] A. Kilaj et al., Nat. Commun. 12, 6047 (2021)
[4] L. Xu et al., arXiv:2308.03935 (2023)

Invited Talk SYCC 1.4 Wed 12:30 Paulussaal
Cold and controlled collisions using tamed molecular beams — •Sebastiaan van de Meerakker — Radboud University Nijmegen, the Netherlands

The study of molecular collisions with the highest possible detail has been an important research theme in physical chemistry for decades. Experimentally, the level of detail obtained in these studies depends on the quality of preparation of the collision partners before the collision, and on how accurately the products are analyzed afterward.

Over the last years, methods have been developed to get improved control over molecules in a molecular beam. With the Stark decelerator, a part of a molecular beam can be selected to produce bunches of molecules with a computer-controlled velocity and with longitudinal temperatures as low as a few mK [1]. The molecular packets that emerge from the decelerator have small spatial and angular spreads, and have almost perfect quantum state purity. These tamed molecular beams are excellent starting points for high-resolution crossed beam scattering experiments.

I will illustrate the possibilities this new technology offers to study molecular collisions with unprecedented precision and at low collision energies. I will discuss our most recent results on the observation of scattering resonances [1], as well as bimolecular dipole-dipole collisions at collision energies down to 0.1 K obtained by merged beam configurations [2].

[1] T. de Jongh et al., Science 368, 626 (2020) [2] G. Tang et al., Science 379, 1031 (2023)