From Ion Imaging to Pure Rotational Spectroscopy: Probing the Dynamics of Multichannel Reactions

The first part of this presentation will be dedicated to the dynamics of chlorine atom reactions with saturated and unsaturated hydrocarbons, an important class of reactions in atmospheric and combustion processes. Reactive scattering distributions of the hydrocarbon radical products were measured in a crossed-beam apparatus coupled with dc slice ion imaging. I will underline the typical distributions associated with direct and indirect mechanisms in order to demonstrate that at low collision energies, Cl reactions with alkenes are dominated by addition-elimination reactions for which the frustrated π adducts must undergo near-dissociation before eliminating HCl. These results suggest that roaming radical dynamics, now widely recognized in unimolecular reactions, may play a central role in a broad class of bimolecular reactions as well.

The second part will be focused on a new experimental approach for probing the dynamics of chemical reactions, namely ‘Chirped-Pulse in Uniform Flow’. This approach employs chirped-pulse Fourier-transform microwave (CP-FTMW) spectroscopy to probe photolysis and bimolecular reaction products that are thermalized at low rotational temperatures (~20K) in uniform supersonic flows. This combination delivers broadband spectra with MHz resolution and allows monitoring, on the μs timescale, of the appearance of transient reaction products. I will illustrate the design and performance of this experimental set-up and discuss a number of challenges related to the collisional environment of the flow. A detailed study on the CN + C3H4 reaction will then be presented. The results show that the combination of these two well-matched techniques also provides insight into the vibrational and rotational relaxation kinetics of the nascent reaction products.

Overall, this work opens the door for new applications in molecular physics and laboratory astrophysics, some of which will be briefly introduced.