

Multimass imaging of molecular fragmentation processes

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The domain of velocity-map imaging is rapidly expanding from highly detailed quantum-state resolved studies of diatomic and triatomic molecules to investigations into the photofragmentation dynamics of much larger molecules, many involving model systems of direct relevance to atmospheric chemistry, astrochemistry, photobiology or organic photochemistry. Such studies present a number of challenges. Diatomic or triatomic molecules generally fragment into only two products, and by characterising one of these products in a state-selective manner and applying basic energy and momentum conservation principles it is generally possible to obtain a fairly complete picture of the photofragmentation dynamics. The key requirement is the existence of a robust state-selective ionization scheme to allow efficient detection of the selected product. In contrast, a much larger molecule is likely to possess numerous fragmentation channels, forming a variety of different products, and even if suitable ionization schemes could be identified, it would quickly become unfeasible to obtain state-resolved images for each and every fragment.

Instead, studies of larger molecules tend to rely on employing a universal (or near-universal) ionization scheme to ionize all products simultaneously, allowing the scattering distribution for each fragment to be recorded, generally at the expense of quantum-state resolution. Images of the scattering distribution for each fragment may be recorded in separate experiments. Alternatively, with the advent of imaging sensors capable of detecting individual particles with nanosecond time resolution¹, images of all fragments may be acquired simultaneously in a single data set. Because multiple fragments may then be detected from an individual parent molecule, a covariance analysis of the data set can reveal the correlated scattering distributions of pairs of photofragments², providing similar information to that obtained in more traditional coincidence measurements.

We have recently carried out a number of studies into photon- and electron-induced processes in a variety of small to medium-sized molecules. These include retrocycloaddition reactions³, McLafferty rearrangements, and photoinduced processes in DNA chromophores and model peptide bonds, as well as ionization and fragmentation processes induced by collisions with electrons⁴. This talk will review recent technical advances in the arena of multimass imaging, including an overview of available detectors, and will present data from a number of these studies.

A large number of researchers have contributed to the work that will be described in this talk. A full list of members of the Pixel Imaging Mass Spectrometry (PImMS) Consortium can be found at <http://pimms.chem.ox.ac.uk/consortium.php>.

References

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