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Vacuum Ultraviolet Photons for the Study of Photodissociation and Bimolecular Reactions

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We have developed a technique called Multiplexed Photoionization Mass Spectrometry for the study of unimolecular and bimolecular chemical reactions. We utilize a low repetition rate pulsed UV laser to initiate a chemical reaction in a collisional environment by breaking a bond in a precursor molecule. The resulting free radicals (resulting from unimolecular dissociation), and the ensuing bimolecular reactions as these radicals react, are probed by extracting a molecular beam sample from the thermal, collisional reaction environment, followed by valence photoionization using quasi-cw, tunable VUV synchrotron radiation. The result is a highly multiplexed experiment (detecting all masses simultaneously), that has high sensitivity (due to single ion counting), is universal (every atom and molecule can be ionized, no exceptions), time-resolved, and sensitive to molecular structure through the photoionization spectra that are obtained. I will give a brief overview of this technique, and compare it to time-resolved Photoelectron Photoion Coincidence Spectroscopy (PEPICO), which we are developing as an even more powerful method to study chemical reactions. Finally, I will discuss the advantages and disadvantages of three regimes of VUV ionization sources: low repetition rates (< 200 Hz), medium repetition rates (0.1 –1 MHz), and quasi-cw repetition rates (> 100 MHz). VUV free electron lasers that achieve moderate spectral resolution, with rapid and wide tunability, at medium repetition rates are the least available source today, but have distinct promise in the study of gas phase chemical physics.

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