THE MARRIAGE OF SPECTROSCOPY AND DYNAMICS: CHIRPED-PULSE FOURIER-TRANSFORM MM-WAVE (CP-FT-MMW) SPECTROSCOPY IN PULSED UNIFORM SUPERSONIC FLOWS

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A new experimental scheme is presented that combines two powerful emerging technologies: chirped-pulse Fourier-transform mm-Wave spectroscopy and pulsed uniform supersonic flows. It promises a nearly universal detection method that can deliver quantitative isomer, conformer, and vibrational level specific detection, characterization of unstable reaction products and intermediates, and perform unique spectroscopic, kinetics, and dynamics measurements.

Chirped-pulse Fourier-transform microwave (CP-FTMW) spectroscopy, pioneered by Pate and coworkers, allows rapid acquisition of broadband microwave spectrum through advancements in waveform generation and oscilloscope technology. This revolutionary approach has successfully been adapted to higher frequencies by the Field group at MIT.

Our new apparatus will exploit amplified chirped pulses in the range of 26-40 GHz, in combination with a pulsed uniform supersonic flow from a Laval nozzle. This nozzle source, pioneered by Rowe, Sims, and Smith for low temperature kinetics studies, produces thermalized reactants at high densities and low temperatures perfectly suitable for reaction dynamics experiments studied using the CP-mmW approach. This combination of techniques shall enhance the thousand-fold improvement in data acquisition rate achieved in the CP method by a further 2-3 orders of magnitude. A pulsed flow alleviates the challenges of continuous uniform flow, e.g. large gas loads and reactant consumption rates. In contrast to other pulsed Laval systems currently in use, we will use a fast piezo valve and small chambers to achieve the desired pressures while minimizing the gas load, so that a 10 Hz repetition rate can be achieved with one turbomolecular pump.

The proposed technique will be suitable for many diverse fields, including fundamental studies in spectroscopy and reaction dynamics, reaction kinetics, combustion, atmospheric chemistry, and astrochemistry. We expect a significant advancement in the ability to detect absolute populations of complex reaction products under near-nascent conditions, providing the powerful method of reaction dynamics with a universal spectroscopic probe capable of capturing the details of complex chemistry for specific product isomers and conformers.