MAR17-2016-030092

Abstract for an Invited Paper
for the MAR17 Meeting of
the American Physical Society

A Few Atoms Too Many: Unravelling Molecular Complexities with Frequency Comb Spectroscopy

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Cavity-enhanced frequency comb spectroscopy[1] has blossomed into a widely versatile tool[2], allowing for trace gas sensing, transient absorption spectroscopy, and the study of buffer gas cooled molecules[3]. This technique offers the unique and simultaneous blend of broad spectral bandwidth, high sensitivity, and high spectral resolution. Recently, we have applied this technique to the important OH+CO→H+CO_2 reaction, which has long been studied due to its importance in atmospheric and combustion environments[4]. Using this technique in the mid-IR, we simultaneously monitor the real-time concentrations of the initial reactants, intermediate transient species, and final products, including for example *trans*-DOCO, *cis*-DOCO, OD, and CO_2 from the deuterated reaction OD+CO→D+CO_2. By determining the time dependencies of these transient molecules, we directly quantify fundamental rate constants and branching yields for the first time. This talk will cover our application of the frequency comb to chemical kinetics as well as the characterization of large molecules in a cold Helium buffer gas environment. Finally, I will discuss the extension of the frequency comb beyond 6 microns.

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[1] M. J. Thorpe et al., Broadband cavity ringdown spectroscopy for sensitive and rapid molecular detection. Science 311, 1595-1599 (2006).

[2] F. Adler et al., Cavity-enhanced direct frequency comb spectroscopy: technology and applications. Annu. Rev. Anal. Chem. 3, 175-205 (2010).

[3] B. Spaun et al., Continuous probing of cold complex molecules with infrared frequency comb spectroscopy. Nature 533, 517-520 (2016).

[4] B. J. Bjork et al., Direct Frequency Comb Measurement of OD + CO $\rightarrow DOCO Kinetics. Science 354, 444-448 (2016).$