HIGH-SENSITIVITY ALL-OPTICAL ULTRAFAST SPECTROSCOPY OF COLD MOLECULAR BEAMS

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Time-resolved nonlinear spectroscopy techniques, such as transient absorption spectroscopy and 2D spectroscopy, are routinely used to study ultrafast dynamics. Due to the limited sensitivity of these techniques, they are most commonly applied to optically thick samples, such as solid and liquid solutions. Using a frequency comb laser and optical cavities, we present a technique for performing ultrafast optical spectroscopy with high sensitivity, enabling work in dilute gas-phase molecules and clusters. Resonantly enhancing the probe pulses, we have demonstrated transient absorption measurements with a detection limit of $\Delta\mathrm{OD} = 2 \times 10^{-10}$ $(1 \times 10^{-9}/\sqrt{\mathrm{Hz}})$[a]. Resonantly enhancing the pump pulses allows us to produce a high excitation fraction at a high repetition rate, so that signals can be recorded from samples with OD as low as 10^{-8}, or column densities $< 10^{10}$ molecules/cm^2. This sensitivity enables ultrafast spectroscopy in dilute molecular beams, where cold isolated designer molecules, radicals, and clusters can be produced that do not exist in solution.

In this talk, I will discuss the basic principles of cavity-enhanced ultrafast spectroscopy, initial one-color demonstration experiments, and the development of widely tunable cavity-enhanced ultrafast spectrometers[b] operating in the ultraviolet, visible, and infrared. Finally, I will discuss progress on the development of cavity-enhanced 2DIR spectroscopy for the study of hydrogen-bonded clusters[c].

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[a] M. A. R. Reber, Y. Chen, T. K. Allison, Optica 3, 311 (2016)

[b] Y. Chen, M. C. Silfies et al., Appl. Phys. B. 125, 81 (2019); M. C. Silfies et al. arXiv:2001.10680 (2020)

[c] T. K. Allison, J. Phys. B. 50, 044004 (2017)