CAVITY-ENHANCED ULTRAFAST SPECTROSCOPY: ULTRAFAST MEETS ULTRASENSITIVE

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Ultrafast optical spectroscopy methods, such as transient absorption spectroscopy and 2D-spectroscopy, are widely used across many disciplines. However, these techniques are typically restricted to optically thick samples, such as solids and liquid solutions. Using a frequency comb laser and optical cavities, we present a new technique for performing ultrafast optical spectroscopy with high sensitivity, enabling work in dilute gas-phase molecular beams. Resonantly enhancing the probe pulses, we demonstrate transient absorption measurements with a detection limit of ΔOD = 2×10^{-10} ($1 \times 10^{-9}/\sqrt{\text{Hz}}$). Resonantly enhancing the pump pulses allows us to produce a high excitation fraction at high repetition-rate, so that signals can be recorded from samples with optical densities as low as OD $\approx 10^{-8}$, or column densities $< 10^{10}$ molecules/cm^2. To our knowledge, this represents a 5,000-fold improvement of the state-of-the-art.

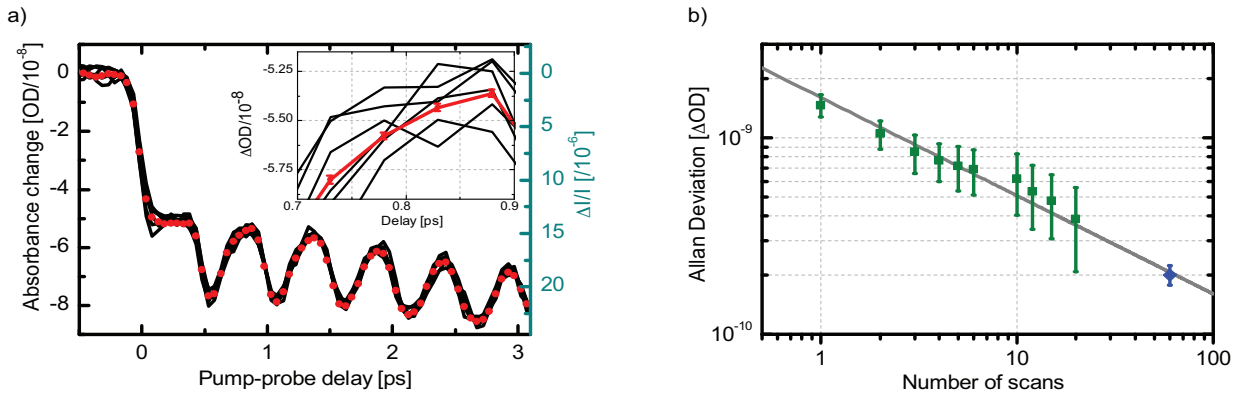

Figure 1: Noise performance of CE-TAS. (a), Transient absorption measurements taken with reduced gas flow and perpendicular polarizations. The red dots represent the average of 60 consecutive scans taken over a 1 hour period. Black curves are every 10th scan from the data set. Inset: Zoom-in around 0.8 ps delay. Error bars represent the uncertainty in the mean. (b), The green squares show the average of the Allan deviations obtained independently for each delay point. Error bars here are the standard deviation (not the uncertainty in the mean) of this ensemble, to represent the spread in the data. The blue diamond is the average of the error bars of (a), along with their standard deviation. The grey line has a slope of -1/2 on the log-log plot, the expected slope for white noise performance