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Cavity-Enhanced Ultrafast Spectroscopy[1]

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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 $\Delta\text{OD} = 2\times10^{-10}$ ($1\times10^{-9}/\sqrt{\text{Hz}}$). 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 $\text{OD} \approx 10^{-8}$, or column densities $< 10^{10}$ molecules/cm^2. To our knowledge, this represents a 5,000-fold improvement of the state-of-the-art.

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