

## MEASUREMENT AND ASSIGNMENT OF METHANE HOT-BAND TRANSITIONS USING CAVITY-ENHANCED FREQUENCY COMB DOUBLE RESONANCE SPECTROSCOPY: EXTENDING TO HIGHER J STATES.

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Accurate assignments of highly-excited molecular vibrational states are needed for the verification of theoretical predictions of high-temperature spectra observed in astrophysics and combustion environments. We previously demonstrated optical-optical double resonance spectroscopy with a continuous wave  $3.3\ \mu\text{m}$  pump and a  $1.67\ \mu\text{m}$  centered frequency comb probe, which allows simultaneous measurement of multiple hot-band transitions from a selectively populated state of  $\text{CH}_4$  with sub-Doppler resolution[1]. The probe spectrum is dispersed using a Fourier transform spectrometer with comb-mode-limited resolution, and interleaving of spectra measured with different  $f_{\text{rep}}$  produces a final spectrum with 2 MHz point spacing. More recently, we implemented an enhancement cavity for the comb probe that improved the absorption sensitivity by more than two orders of magnitude. In addition, the use of both co- and counter-propagating probe light eliminated errors in the final state term value due to errors in the Lamb-dip locking point of the pump, which limited the accuracy of the earlier single-pass measurements. Here we use this cavity-enhanced system to measure  $3\nu_3 \leftarrow \nu_3$  methane transitions from higher J states with much-improved frequency and relative intensity accuracy, which allows unambiguous assignments of the final states using two independent methods. The first method relies on combination differences in probe spectra observed when the pump is sequentially Lamb-dip locked to P, Q, and R  $\nu_3$  band transitions from a fixed rotational level of the ground state. We also observe spectra with probe polarization both parallel and perpendicular to that of the pump light and compare the intensity ratio with predictions for the case of a strongly saturated pump. The two experimental methods of final state assignment agree, and are largely confirmed by theoretical predictions from the TheoReTS database. I. A. Foltynowicz, et al., *Phys. Rev. Lett.* 126, 063001 (2021); *Phys. Rev. A* 103, 022810 (2021).