MID-INFRARED DOPPLER-FREE SATURATION ABSORPTION SPECTROSCOPY OF METHANE FOR FUTURE CAVITY-ENHANCED DOUBLE-RESONANCE SPECTROSCOPY INVESTIGATING ITS HIGH POLYADS.

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Understanding the rotational structure of molecular spectra requires high resolution and high-frequency accuracy. Furthermore, a capability of high-speed, wide-range spectral scan is strongly desired. We have developed a mid-infrared Doppler-free saturation absorption spectroscopy apparatus using a continuous-wave optical parametric oscillator (CW-OPO).[a] Here we report a comprehensive spectral scan of the $\nu_3 = 1$ band of methane (CH_4). The absolute frequency calibration was achieved using previously reported transition frequencies determined using optical frequency combs,[b] while a home-build Fabry-Pérot etalon was used for relative frequency calibration. A linewidth of less than 5 MHz has been reached, and the frequency accuracy is estimated to be better than 1 MHz, both of which can be further improved. We have successfully locked the frequency of the OPO to a Doppler-free line of CH_4 using a top-of-fringe locking method. A cavity-enhanced double-resonance spectroscopy apparatus is under construction. It combines the Doppler-free saturation absorption setup and an existing continuous-wave cavity ring-down (CW-CRDS) spectroscopy apparatus. The first mid-infrared photon from the frequency-locked OPO pumps the CH_4 molecule to the $\nu_3 = 1$ vibrational levels, followed by a further excitation to high polyads using a Ti:Saphire ring laser.

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[a] D. B. Foote, M. J. Cich, W. C. Hurlbut, U. Eismann, A. T. Heiniger, and C. Haimberger, "High-resolution, broadly-tunable mid-IR spectroscopy using a continuous-wave optical parametric oscillator" *Opt. Express* **29**, 5295-5303 (2021)

[b] M. Abe, K. Iwakuni, S. Okubo, and H. Sasada, "Accurate transition frequency list of the ν_3 band of methane from sub-Doppler resolution comb-referenced spectroscopy," *J. Opt. Soc. Am. B* **30**, 1027-1035 (2013)