MEASURING ROTATIONAL SPECTRA IN EXCITED VIBRATIONAL MODES: A NEW TECHNIQUE BASED ON A QUANTUM CASCADE LASER-PUMPED MOLECULAR LASER

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Measuring the rotational spectra of a molecule in excited vibrational modes can be challenging due to the low thermal population of these rotational levels and the weakness of the underlying absorption lines. We recently demonstrated a quantum cascade laser (QCL)-pumped molecular laser (QPML) concept in which virtually any rotational transition of any excited vibrational mode of virtually any molecule with a permanent dipole moment and a vapor pressure can be made to lase by pumping its corresponding ro-vibrational transition with a frequency-tunable QCL. Here, we show how the frequency of those QCL-pumped rotational transitions in excited vibrational modes may be measured with accuracy comparable to transitions in ground vibrational levels using high precision modulation spectroscopy techniques. We applied this concept to nitrous oxide (N_2O), and measured the individual frequencies of 20 lines in the v3 vibrational mode (2225 $cm^{-1} \approx$ 10 kT) by means of heterodyne spectroscopy with an experimental uncertainty of 200 kHz when lasing and without modulation, improving to 50 kHz with QCL-enhanced absorption or amplification with modulation. We report the measured spectra and the fitted rotational constants B_3 and DJ_3 corresponding to this vibrational mode.