HIGH-RESOLUTION SPECTROSCOPY OF COLD SAMPLES IN SUPERSONIC BEAMS USING A QCL DUAL-COMB SPECTROMETER

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Optical frequency comb spectroscopy has proven a very useful tool for high resolution molecular spectroscopy. Frequency combs based on quantum cascade lasers (QCL) offer the possibility to easily explore the mid-infrared spectral range (4-12 μm), but suffer from very large repetition frequencies (~ 10 GHz) which make them seemingly unsuitable for high resolution spectroscopy.

Here, we present techniques to overcome this limitation. We have employed the combined advantages of high temporal ($< 4\ \mu$s) and high spectral resolution to measure the IR spectra of CF_4 and $CHCl_2F$ in pulsed, skimmed supersonic beams. The low rotational temperature of the beams and the narrow expansion cone after the skimmer enabled the recording of spectra of cold samples with high resolution. The spectra cover the range from 1200 to 1290 cm^{-1} and the narrowest lines have a full width at half maximum of 15 MHz, limited by the Doppler effect. The results demonstrate the potential of QCL dual-comb spectroscopy for broadband ($> 60\ \mathrm{cm}^{-1}$) acquisition of spectra at high resolution (< 15 MHz) and high sensitivity in the mid-infrared range. The power of the new technology is demonstrated by comparison with previous results on these molecules obtained by FTIR and diode laser spectroscopy of seeded cw and pulsed supersonic jets.[a]

a) M. Snels and M. Quack, J. Chem. Phys.1991,95,6355, M.Snels, V. Horka-Zelenkova, H. Hollenstein, M. Quack ,'High resolution FTIR and diode laser spectroscopy of supersonic jets', in Handbook of High-resolution Spectroscopy, Eds.; M. Quack and F.Merkt , Vol. 2, pp. 1022-1067, Wiley, Chichester, 2011; M. Caviezel, V. Horka, Z. Guennoun , G. Seyfang, M. Quack, unpublished manuscript (in preparation for Mol Phys.)