Enhancement of molecular dispersion spectral signatures in mode-locked lasers

J. Mandon[1], G. Guelachvili[1], I.T. Sorokina[2], E. Sorokin[3], V. Kalashnikov[3], N. Picqué[1],

1. Laboratoire de Photophysique Moléculaire, CNRS, Bâtimnet 350, Université Paris-Sud, 91405 Orsay, France

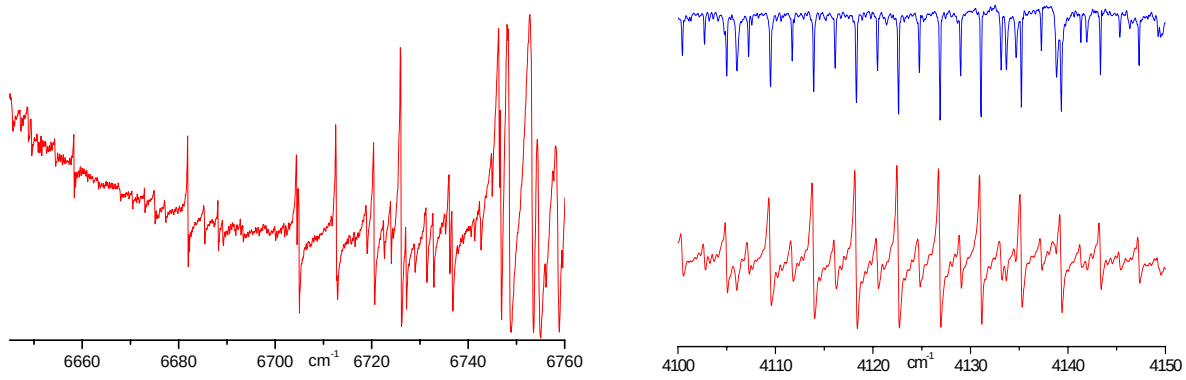
2. Department of Physics, Norwegian University of Science and Technology, N-7491 Trondheim, Norway

3. Photonics Institute, TU Vienna. Gusshausstrasse 27/387. A-1040 Vienna, Austria.

Dispersion determination of transitions of gas-phase molecules have been poorly investigated due to the experimental complexity of their measurements. Broadband traditional measurements can be obtained from dispersive Fourier transform spectroscopy [1]. The interferograms with and without sample inserted in one arm of the Michelson interferometer are compared in a two-step approach. This procedure has been mostly practiced in the less-demanding far-infrared region. However, more stringent requirements on instrumental parameters made the technique not widely used. Frequency modulation tunable laser spectroscopy [2] also provides high-resolution dispersion-related information, but on restricted spectral bandwidth. More recently, different broadband techniques [3-6], mostly based on frequency combs [3-5], have been demonstrated, leading new interest to dispersion measurements, providing altogether amplitude and phase information.

In this contribution, we present a new approach to dispersion spectroscopy which combines high resolution, broad spectral bandwidth and high sensitivity. It consists in inserting the gas sample of interest inside a femtosecond laser cavity and measuring the resulting spectrum, preferably with a Fourier transform interferometer. Dispersion-enhanced molecular transitions appear on the laser emission envelope.

We have given the initial experimental evidence of this approach with two different infrared mode-locked femtosecond lasers based on 1.5 µm Cr^{4+}:YAG and a 2.4 µm Cr^{2+}:ZnSe oscillators. As shown on Figure 1, first results are obtained at near atmospheric pressure, with residual-water-vapour or acetylene transitions. The spectra shown on Figure 1 exhibit a signal-to-noise ratio enhancement of about 50, when compared to traditional low-pressure absorption spectra. These results are satisfactorily reproduced by the numerical model for femtosecond oscillators in which a narrow-width chromatic dispersion is introduced. Experimental investigation with the molecular sample conditioned at low pressure into an evacuated oscillator is underway.



(a) **(b)**

Fig. 1. Experimental results showing the enhancement of molecular dispersion-like transitions inside a femtosecond laser cavity. (a) The fs laser is a Cr^{4+}:YAG oscillator at atmospheric pressure and the transitions are due to residual water vapour. (b) Upper trace: C_2H_2 (22 hPa) conventional absorption spectrum at 0.12 cm^{-1} resolution using a Cr^{2+}:ZnSe laser as a light source [7].Lower trace: The fs laser is a Cr^{2+}:ZnSe oscillator filled with nitrogen (900 hPa) and acetylene (133 Pa) and the transitions are due to the acetylene $\nu_1+\nu_5^1$ band.

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