

Mid-IR frequency laser control with a comb for H_2^+ spectroscopy

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Abstract. We will report our recent progress toward H_2^+ spectroscopy by use of a SI-referenced Mid-IR source laser. H_2^+ molecular ions are very interesting candidates to improve the determination of fundamental constants, such as the proton to electron mass ratio m_p/m_e and search for new physics beyond the standard model. At LKB, an erbium fibered frequency comb is phase locked to the LNE-SYRTE frequency standards thanks to the T-REFIMEVE network. By sum frequency generation in a AgGaSe₂ crystal between a CO₂ laser and an output of the comb at 1895 nm, a shifted frequency comb centered at 1560 nm is generated. The latter is then mixed with the original one to generate a beatnote used to stabilise the Mid-IR laser. As a first application, a narrow saturated absorption line in formic acid has been extensively studied. Pressure, power and modulation depth shifts and broadenings have been evaluated, leading to a determination of its central frequency at a sub ppt (10^{-12}) resolution, high enough for H_2^+ spectroscopy and fundamental constant determination.

1 Introduction

High precisions molecular spectroscopy is a very useful tool to test our theoretical understanding of atomic and molecular structure, and in particular the validity of Quantum Electrodynamics (QED) calculations. It can lead to improved determination of certain fundamental constants that enter as parameters in the theoretical models. In this context, H_2^+ or HD^+ molecular ions are very promising candidate for fundamental metrology because they are simple calculable systems with transition frequencies that can be determined to the 11th or even 12th decimal. Their ro-vibrational levels have very long lifetimes, and consequently they offer many possible ultra-narrow infrared transitions, which frequencies may be measured with very high accuracies. Moreover, these frequencies are sensitive to the proton to electron mass ratio m_p/m_e and could thus be used to improve its determination at the 0.01 ppb level.

2 H_2^+ spectroscopy setup

Our experimental setup aims at a high-precision measurement of the $(v=0, L=2) \rightarrow (v'=1, L'=2)$ Doppler-free two-photon transition in H_2^+ . A linear Paul trap allows to confine both H_2^+ and Be^+ ions, installed in a ultrahigh-vacuum vessel. A 313 nm laser is used to cooled the Be^+ ions and thus the H_2^+ ions with the well know sympathetic cooling technique. The generation of a state selected molecular ions is done by a Resonance-Enhanced MultiPhoton Ionization (REMPI) process from a H_2 molecular beam and a pulsed laser at 303 nm. A Mid-IR

frequency stabilized quantum cascade laser at 9.17 μm is then used to probe the two-photon vibrational transition. Finally, a pulsed laser at 213 nm allows to detect the transition by selective dissociation of the $v'=1$ level with a method called Resonance-Enhanced Multiphoton Dissociation (REMPD). The spectroscopic signal thus consists in a loss of H_2^+ ions. Figure 1 aims at illustrating these different steps.

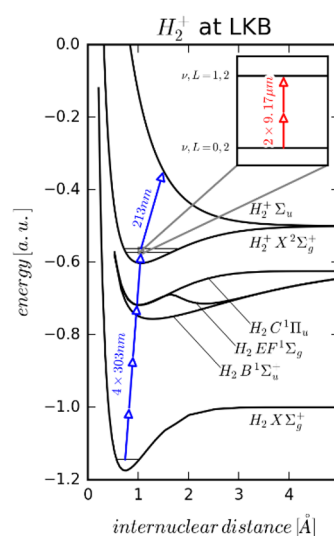


Fig. 1. Energy level of H_2 .

3 Mid-IR experimental setup

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The work presented here focusses on the stabilisation of the mid-IR laser and first tests on formic acid (HCOOH) saturation spectroscopy. Several parts of the experimental setup have been already described in [1]. The laser source is a single mode CO₂ laser operated on the 9R(42) line. The beam is split into three parts, one probing the molecular transition, one for frequency control against a frequency comb and one to control the frequency of a quantum cascade laser (not used in this work). The CO₂ laser frequency is controlled using a piezo ceramic with two zones, a short one for fast control with about 10 kHz bandwidth and a long one with ≤ 1 kHz bandwidth for long term stabilization. The saturated absorption is performed in a 1.2 m long symmetric Fabry-Perot cavity installed in a vacuum vessel. The cavity resonance frequency is modulated at 33 kHz and locked on resonance with the CO₂ laser frequency using a first harmonic phase sensitive detection technique on the signal transmitted by the cavity.

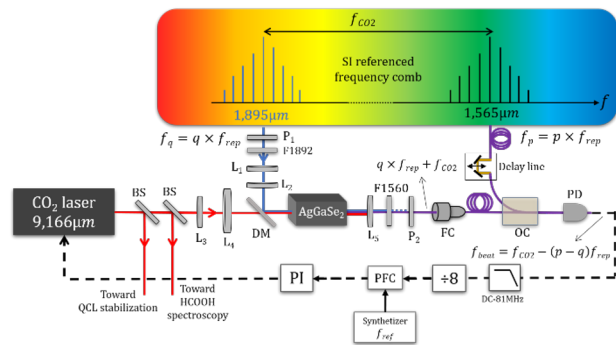


Fig. 2. Experimental setup used to stabilise the CO₂ laser to the LNE-SYRTE referenced frequency comb. BS: Beam Splitter, DM: Dichroic mirror, OC: Optical Coupleur, PD: PhotoDetector, PFC: Phase/Frequency comparator.

The CO₂ laser frequency is referenced to the Système International (SI) following a technique described in [2] and illustrated in Figure 2. In brief, a 1.56 μm frequency comb is optically locked to the 1542 nm ultrastable signal synthesized at LNE-SYRTE laboratory and disseminated through the T-REFIMEVE network. An additional 1895 nm output of the comb is focused and overlapped with the CO₂ laser in a non-linear AgGaSe₂ crystal. Sum frequency generation of the 1895 nm comb and the CO₂ laser gives a shifted comb centered around 1560 nm. This shifted comb is then combined with the original one, after passing through a delay line to temporally match the pulses, in order to generate a beat between the CO₂ laser and the comb. This beat signal is then used to phase-lock the CO₂ laser. In practise, the beat signal is bandpass filtered around 64 MHz, divided by 8 and compared to a RF synthesizer with a phase-frequency comparator to get an error signal. The output of a proportional-integrator system is sent to the fast piezo of the laser and a second integrator stage output is applied on the slow one.

The CO₂ laser is frequency shifted by 128 MHz and matched to a Fabry Perot cavity containing a low HCOOH pressure (about 1 μbar or 0.1 Pa) to perform spectroscopy. The synthesizer used to phase lock the CO₂ laser to the comb is frequency modulated at 700 Hz with a modulation depth from 3 to 5 kHz resulting in an effective

frequency modulation of the CO₂ laser from 24 to 40 kHz due to the divider by 8. The saturated absorption signal is detected using a third harmonic phase sensitive detection.

4 First Results

An example of saturated absorption spectra obtained is presented in Figure 3. The data are both fitted with a theoretical model that takes into account the effect of frequency modulation (S3 model) [3] and intra-cavity intensity modulation (S1 + S3 model) [4]. S_n with $n=1,3$ corresponds to first respectively third derivative of a modulated Lorentzian profile. We have systematically studied the saturated absorption line for three powers, three modulation depths and four pressures. This allows to extract the de-perturbed central frequency of the line with a sub ppt (10^{-12}) resolution, the light shift and pressure shift as well as the pressure broadening of the line.

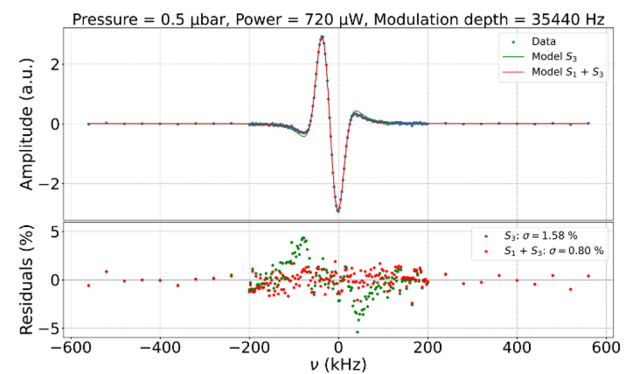


Fig. 3. Saturated absorption spectra of the $J''=21, K_a''=2, K_c''=20, J'=21, K_a'=3, K_c'=19$ HCOOH detected in third harmonic fitted by derivatives of modulated Lorentzian profiles. $\nu=0$ corresponds to 32 708 392 MHz.

5 Conclusion

Our SI-referenced spectrometer at 9.17 μm allows frequency measurement below the ppt level, a high enough resolution for H₂⁺ spectroscopy and fundamental constant determination.

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