NEW POSSIBILITIES FOR FREQUENCY STABILIZATION OF FAR INFRARED LASERS

R. Bocquet,[1,*] M. Yu. Tretyakov,[*,†] A. F. Krupnov,[*,†] S. A. Volokhov,[*,†] and L. Poteau*

**Laboratoire de Spectroscopie Hertzienne, Centre d'Etudes et de Recherches Lasers et Applications, Université des Sciences et Techniques de Lille, 59655 Villeneuve d'Ascq Cedex France*

†Permanent address: Applied Physics Institute of the Russian Academy of Science, 46 Ulyanova Street, 603600 Nizhnii Novgorod GSP-120 Russia

Received May 8, 1996

keywords: far infrared laser, submillimeter wave, frequency stabilization

abstract: All spectrometers based on the use of a far infrared laser present the drawback of the laser frequency uncertainty. Even if the knowledge of the laser frequency is very precise, a shift of the laser cavity length is always present, limiting the spectrometer frequency accuracy to 500 kHz.
The goal of this paper is to give a solution to this drawback in demonstrating the feasibility of a frequency stabilization of the far infrared laser.

[1] e-mail: *bocquet@lsh.univ-lille.fr*

1. Introduction

Since 1990, we have been using at Lille (France), a far infrared (FIR) spectrometer for spectroscopic applications (1). This experiment consists in mixing the output radiation from an FIR laser with the radiation from a synthesizer, operating in the range 2-18 GHz, to produce sidebands in the FIR region. These sidebands with a tunability of 32 GHz serve as a source for the spectrometer. The drawback of this experiment is the poor knowledge of the FIR laser frequency. The uncertainty of the measured absorption line frequency is in fact proportional to the laser line frequency uncertainty. Figure 1 shows an example of a FIR laser gain curve.

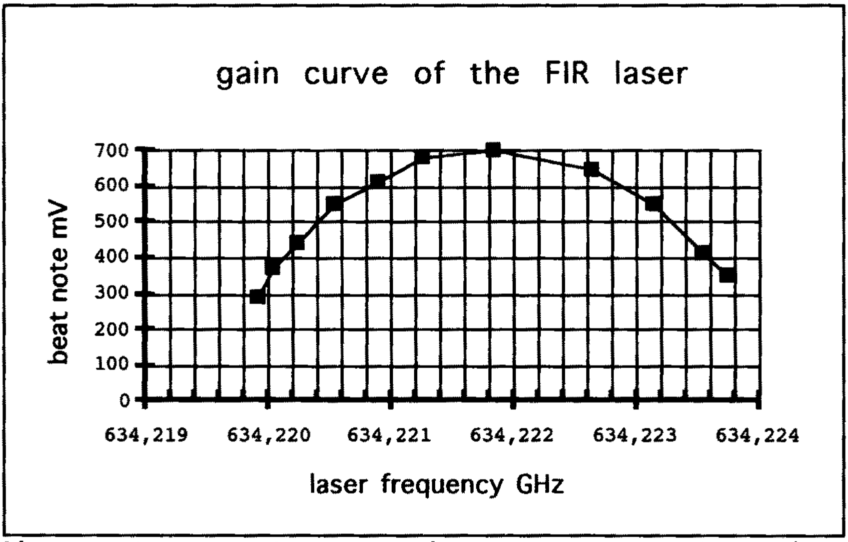

figure 1: FIR laser gain curve for the vinyl fluoride line pumped by the 10P20 line

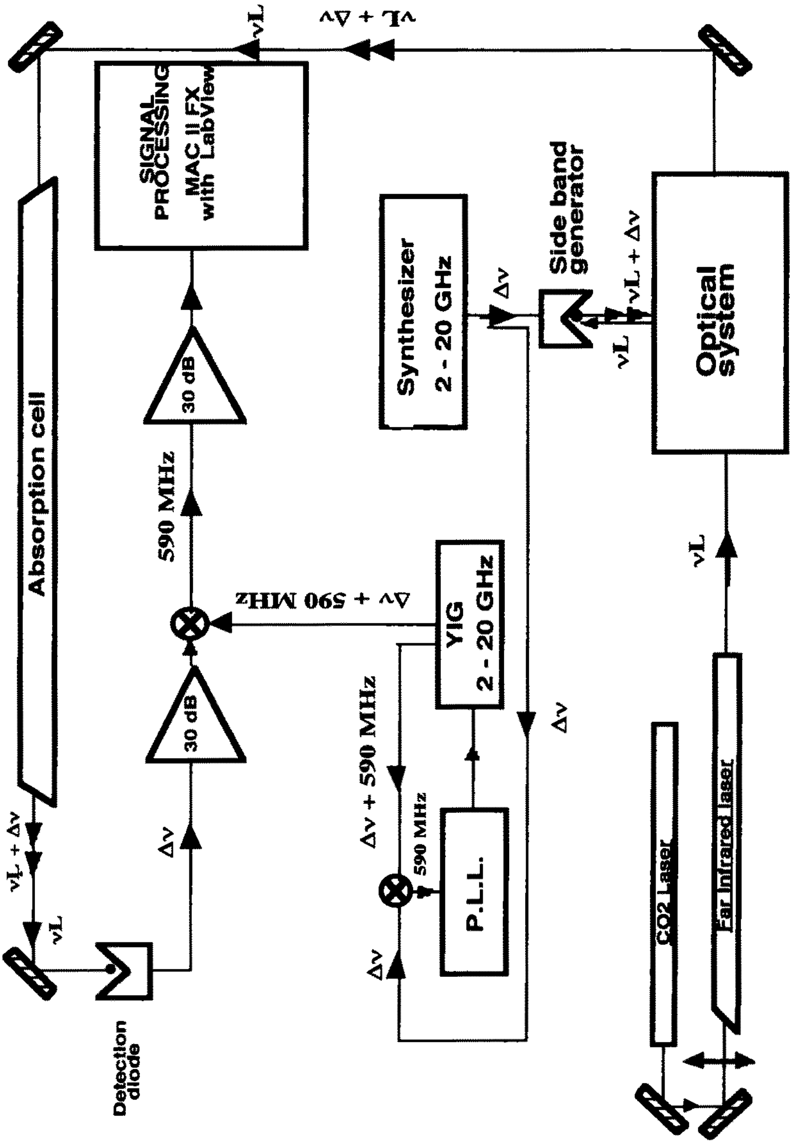

Figure 2: Tunable Far Infrared Spectrometer

One can see on this characteristic example, a bandwidth of the order of 3.5 MHz. This means that we can tune the laser radiation frequency over this range. One can also imagine the difficulty to tune the laser cavity length just at the top of the gain curve. We can reach in these conditions, an accuracy of the order of 500 kHz for the absorption line frequency measurements (2).
The goal of this short paper is to show the possibility of a frequency stabilization of the FIR laser to increase the frequency accuracy of the experiment.

2. Experimental set-up

Figure 2 shows the scheme of the FIR heterodyne sidebands spectrometer used at Lille. Briefly, we generate sidebands by mixing the FIR laser frequency with a synthesized frequency in the range 2-18 GHz. A classical heterodyne detection is used. The first intermediate frequency, in the range 2-18 GHz, is obtained by mixing the unmodulated part of the laser with the sidebands. A second downconversion is realized to have a fixed intermediate frequency at 590 MHz. Interested readers can find more details in ref(1).
Figure 3 shows the added system which permits to generate the sidebands and to measure the laser frequency simultaneously. We used a mixer developed at the Institute of Applied Physics, Nizhnii Novgorod, with a construction similar to the multiplier described in (3). A planar Schottky diode is fitted at a 8 mm focal point of a parabolic mirror, focusing the FIR laser radiation on the diode.

The HP synthesizer in the range 2-18 GHz is coupled to the diode via a coaxial cable to generate sidebands.

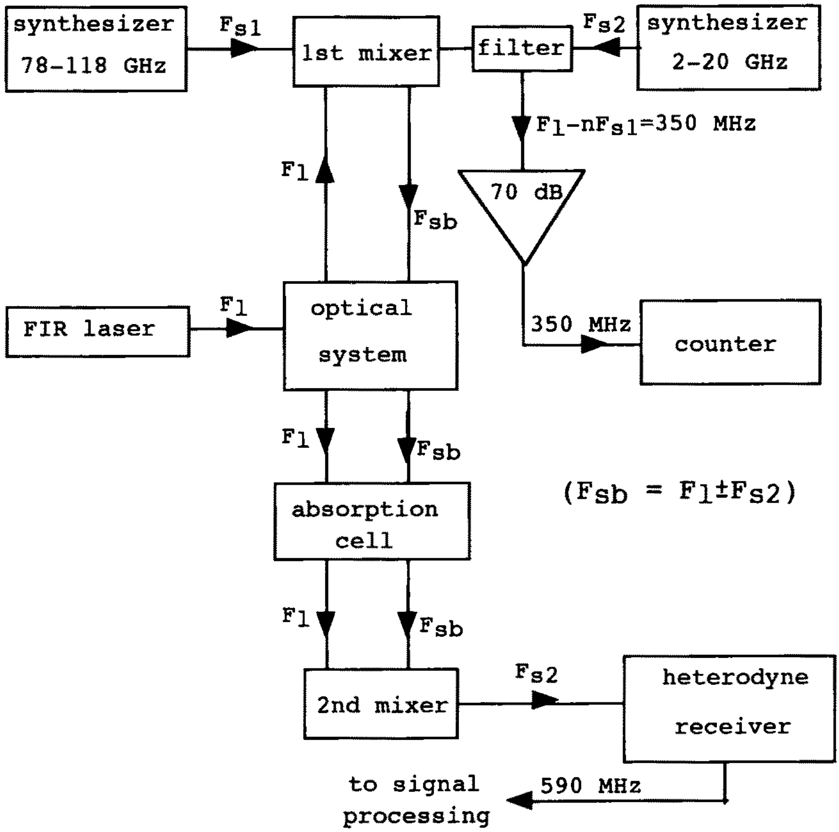

Figure 3: simultaneous sideband generation and FIR laser frequency measurement

A "Kvarz" synthesizer (4) in the range 78-118 GHz is coupled to the diode via a waveguide to produce a beat note near 350 MHz, between a harmonic of its frequency and the FIR laser. After amplification, the beat note may be used for frequency measurements (5) as well as for FIR laser frequency stabilization.

3. Results

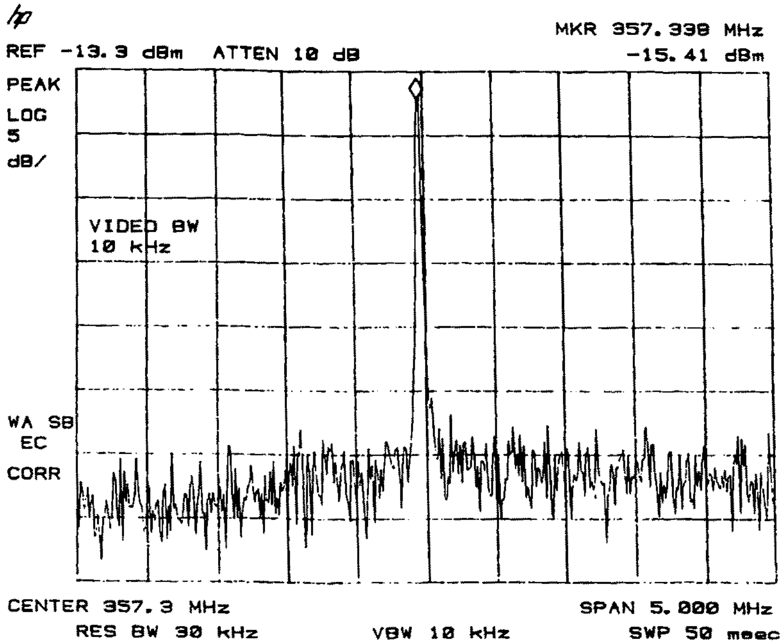

Figure 4: beat note between the laser at 634 GHz and the 7th harmonic of the millimeter wave synthesizer

In order to fulfil our goal, we had to demonstrate the possibility to generate with the same mixer:

- sidebands from the FIR laser radiation to realize a coherent tunable source for spectroscopic applications
- a beat note between a harmonic of the millimeter wave synthesizer frequency and the FIR laser to measure (or stabilize) the FIR laser frequency.

Figure 4 shows the beat note obtained with the CH_2CHF emission line pumped by the 10P20 line of the CO_2 laser. The frequency of the millimeter wave synthesizer F_{s1} was:

$$F_{s1} = 90.654200 \text{ GHz}$$

The beat note is at F_b= 357.338 MHz. This gives a laser frequency F_l:

$$F_l = 7xF_{s1} - F_b \Rightarrow F_l = 634.22207 \text{ GHz}$$

This value is in very good agreement with our previous measurement of this line frequency (1).

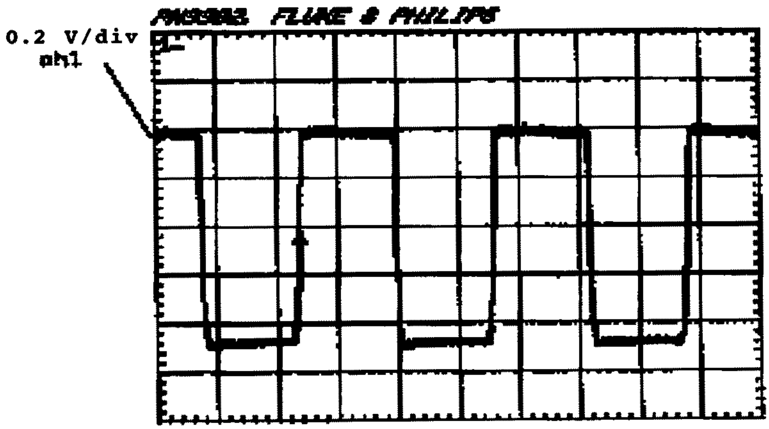

Figure 5: sidebands (4 GHz from the laser at 634 GHz) after the second downconversion at 590 MHz and detection.

Figure 5 shows the detected signal of sidebands at 4 GHz from the laser frequency at 634 GHz. This signal is obtained after 2 downconversions, amplification and detection. The modulation is only used for a visualization of the sideband level and made by chopping the CO_2 laser beam.
The last two figures obtained at the same time demonstrate the feasibility of the experiment.
We have also made a trial of this set up in a real spectroscopic application. Figure 6 shows a record of the H_2S absorption line $(7_{6,1} \leftarrow 7_{5,2})$.

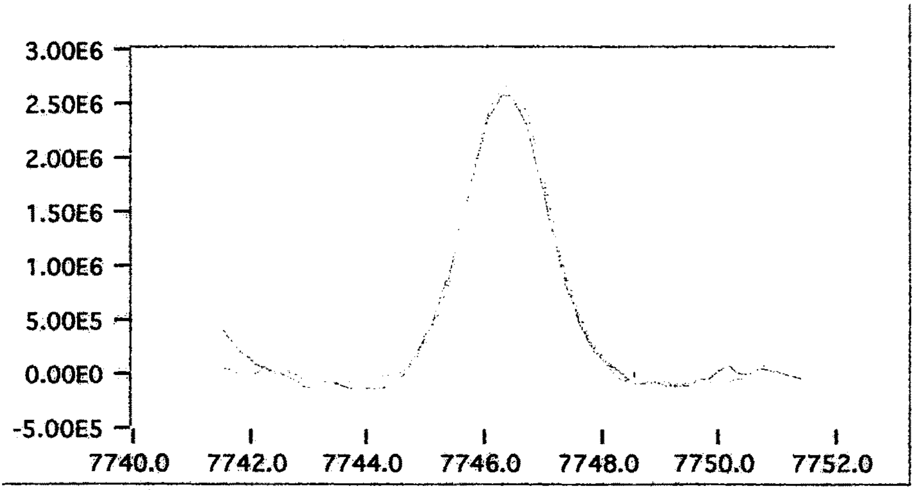

Figure 6: H_2S absorption line at 626.474590(76) GHz

The "y" scale is in arbitrary units and is proportional to the detected power. The "x" scale is in MHz and corresponds to the microwave synthesizer frequency F_{s2}. The two lines correspond to scanning up and down the frequency F_{s2}.

The line frequency is determined by the following formula:

$$F_{line} = (F_{s1} x 7 - F_b) - F_{s2}$$

where:

- F_{s1} is the frequency of the millimeter wave synthesizer

- F_b is the measured frequency of the beat note

-F_{s2} is the measured frequency of the line as shown on figure 6

For precise measurement of this absorption line, we carried out the following statistical procedure. We repeated five times the frequency measurement of the line. For each of them, we detuned the FIR laser cavity and retuned it back to the maximum of laser power. For each measurement, the beat note frequency was measured by a spectrum analyzer. This procedure gave a 76 kHz standard deviation σ, with an average line frequency of 626.474590 GHz.
This measurement is in good concordance with ref(6). This line has been measured in Cologne with a frequency stabilized backward wave oscillator at 626.474625(70) GHz.
With frequency stabilization of the FIR laser, this accuracy could be much better. We have in fact a frequency drift of the FIR laser, due to the heating of the output coupler by the CO_2 laser. This means that the FIR laser frequency was liable to move during the spectroscopic measurement, giving a corresponding frequency uncertainty.

Only the lack of the necessary mechanics and electronics for the servo-control of the FIR laser cavity prevented us from frequency stabilizing the laser.
The feasibility of such an experiment at higher frequencies is supported by figure 7 which presents a beat note between the FIR laser emission line at 1886 GHz and the 19th harmonic of the millimeter wave synthesizer. This FIR laser line corresponds to the HDCO emission line pumped by the 10P10 CO_2 laser line and measured by us in ref(5).

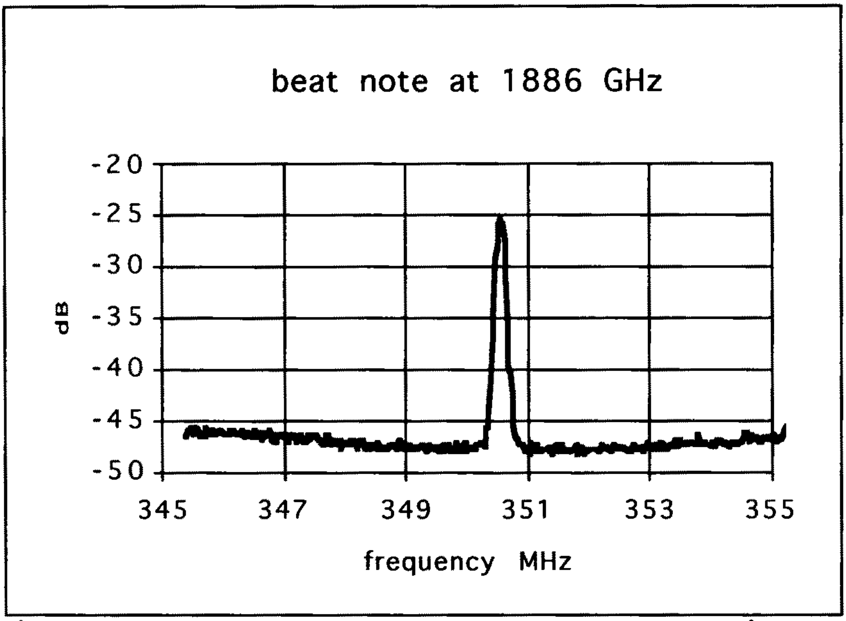

Figure 7: beat note between the HDCO line at 1886 GHz and the 19th harmonic of the millimeter wave synthesizer

4. Conclusion

We have demonstrated the possibility, with the same mixer, to generate simultaneously sidebands from the FIR laser and a beat note between the FIR laser and a harmonic of the millimeter wave synthesizer.
In counting the beat note, we are able to measure the FIR laser frequency and thus reach a frequency accuracy of 76 kHz for the molecular absorption line frequency.
In the future, the beat note will be used to frequency lock the FIR laser. This last step will be reported later; some technical improvements being necessary to realize it.

Acknowledgments

The Laboratoire de Spectroscopie Hertzienne is "Unité de Recherche Associée au CNRS". The Centre d'Etudes et de Recherches Lasers et Applications (CERLA) is supported by the ministère chargé de la recherche, the région Nord-Pas de Calais and the Fonds Européen de Développement Economique des Régions.
This work is part of the programme of the European Community Human Capital and Mobility Network SCAMP (Contract N°. EBRCHRXCT 930157 and its supplementary agreement ERBCIPDCT 940614). M. T. is also grateful to the University of Lille I for financial support. A. K. is grateful to the region Nord-Pas de Calais for financial support.

References

(1) D. Boucher, R. Bocquet, J. Burie, W. Chen, *J. Phys. III France* **4**,1467-1480 (1994)

(2) X. Li, R. Bocquet, D. Petitprez, D. Boucher, L. Poteau, J. Demaison, *J. Mol. Spectrosc.* **172**,449-455 (1995)

(3) A. F. Krupnov, M. Yu. Tretyakov, YU. A. Dryagin, S. A. Volokhov, *J. Mol. Spectrosc.* **170**, 279-284 (1995)

(4) A. F. Krupnov,O. P. Pavlovsky, *Int. J. IR & mmwaves* **15**, 1611-1624 (1994)

(5) Bocquet R., Tretyakov M. Yu., Krupnov A. F., Poteau L., Boulogne O., *accepted for publication Int. J. IR & mmwaves* **17**,6 (1996)

(6) S. P. Belov, K. M. T. Yamada, G. Winnewisser, L. Poteau, R. Bocquet, J. Demaison, O. Polyansky, M. Y. Tretyakov, *J. Mol. Spectrosc.* **173**,380-390 (1995)