Terahertz frequency metrology and sensitivity issues in photomixer spectrometer

L.F. Constantin, L. Aballea, J. Demaison

Abstract—**We propose a compact photomixer spectrometer setup, using 2 laser diodes locked on the resonances of an ultrastable Fabry-Perot cavity, that generates difference-frequency Terahertz-waves with high spectral purity. We investigate a cavity-enhanced spectroscopic technique that would increase the spectrometer sensitivity proportionally with the cavity finesse. We demonstrate a broadband Terahertz Fabry-Perot cavity with microstructured metallic-grid mirrors that have a finesse of 50 at 1,2 THz. Its quality factor is greater than 10^4 in the 0,6-2 THz spectral range.**

Index Terms—Photomixing, Terahertz, Cavity-enhanced spectroscopy, Submillimeter wave resonators, Frequency measurement, Q factor

I. Introduction

IN the last decades different coherent sources operating at terahertz frequencies were developed in order to bridge the electromagnetic gap between electronics and photonics. Among them, the optically-pumped gas lasers are covering the spectral range between 300 GHz to 3 THz with a dense grid of emission lines. The frequency tuning over a discrete spectral range can be ensured via the sideband generation technique, which made these sources suitable for high resolution molecular spectroscopy experiments.

The generation of terahertz-wave with a broad tuning bandwidth [1] was demonstrated from the difference-frequency of two detuned near-infrared diode lasers that are heterodyned in a non-linear element, the photomixer. We will focus in this paper on the design of a terahertz spectrometer relying on photomixers and frequency stabilised laser diodes and will approach sensitivity and Terahertz frequency metrology issues. An important theme in modern spectroscopy is the use of an extended interaction length between matter and the electromagnetic field confined inside a high finesse cavity for an increased detection sensitivity. For example, the noise-immune, cavity-enhanced, optical-heterodyne molecular spectroscopy technique allowed reaching in the infrared range an ultimate fractional detection sensitivity of 5.10^{-13} [2]. Another important theme is the application of frequency metrology for precision measurements. For example, infrared molecular lines with kilohertz-level linewidth have been measured using an intracavity Ramsey separated fields spectroscopy technique [3]. This paper will thus explore some possible ways to extent these potentialities of optical cavities for precision measurements in the far-infrared domain.

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The authors are with the Centre National de la Recherche Scientifique, Laboratoire de Physique des Lasers, Atomes et Molécules, Université Lille 1, 59655 Villeneuve d'Ascq, France. Correspondence should be addressed to L.F. Constantin, phone: +33(0)3 20434790, fax: +33(0)3 20337020; e-mail: FL.Constantin@univ-lille1.fr.

II. Difference-frequency generation of ultra-stable Terahertz-wave

The non-linear element used for difference-frequency generation is a low-temperature-grown gallium arsenide layer (2 μm thickness). It was grown on semiinsulating GaAs substrate using molecular beam epitaxy and was subsequently annealed at IEMN. A pump-probe photoreflectivity experiment allowed us to estimate the photocarriers lifetime for different growth and anneal conditions. Electron-beam lithography and lift-off process were used to define a Ti:Au broadband spiral antenna with interdigitated electrodes (width 0.2 μm, periodicity 1,8 μm) over a 8 μm x 8 μm active area. Two monochromatic beams of near-infrared radiation, focused on the electrodes, modulate the conductance of the substrate. Upon biasing the photoconductor, THz currents generated in the photoconductor flows through the antenna that radiates THz radiation. A hyperhemispherical silicon lens collimates Terahertz radiation that can be further detected with an InSb bolometer. The possible ways for the optimisation of the output power of GaAs THz photomixer source [4] are essentially achieving low photo-carrier recombination times and low RC antenna constant and using a substrate with high thermal dissipation. The generated power is expected in the μW range up to 1 THz [5].

We propose a new setup for the photomixer spectrometer, shown in Figure 1, that allows synthesizing any difference frequency in the far-infrared domain from two diodes laser that are locked on different longitudinal modes of a high finesse Fabry-Perot cavity.

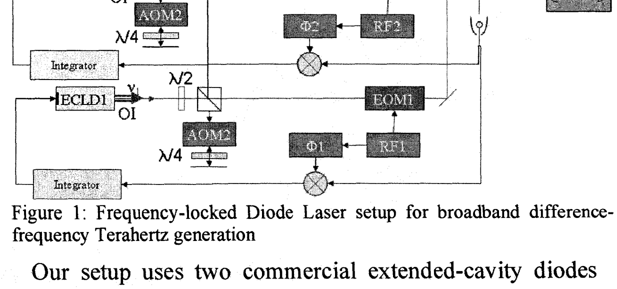

Figure 1: Frequency-locked Diode Laser setup for broadband difference-frequency Terahertz generation

Our setup uses two commercial extended-cavity diodes laser in a Littrow configuration operating around 830 nm with an optical power of 100 mW. They can be tuned electrically over 30 GHz without mode-hops and mechanically over more than 15 nm. We recorded the beatnote between the free-running diode lasers (Figure 2) that have a Lorentzian lineshape. The FWHM linewidth of each laser is thus 1,5 MHz.

The emitted laser radiation is coupled to optical isolators and a pair of anamorphic prism pairs. A $\lambda/2$ waveplate associated with a polarising beamsplitter cube transmits a

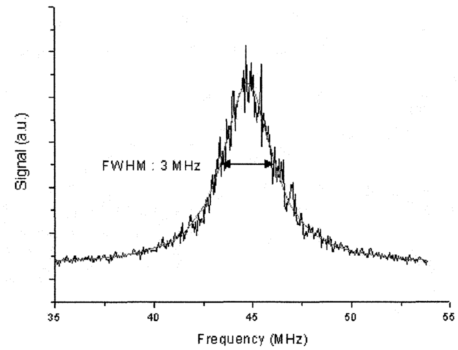

Figure 2: Beatnote of the free-running diodes laser

small amount of diode laser radiation to the frequency locking system. We will use the Pound-Drever-Hall technique [6] that operates a heterodyne detection of the high-frequency phase modulated sidebands which are reflected by the Fabry-Perot cavity using an avalanche photodiode. It generates an error signal which is insensible to the laser intensity noise and that have a bandwidth greater than the linewidth of the cavity. The error signal can be used to lock the diode laser on the top of the Fabry-Perot resonance by a slow lock-loop, with the PZT that tune the diode laser cavity length, and a fast-loop, with a high bandwidth direct current control of the diode laser.

We developed a ULE etalon with finesse greater than 10^4 and a 750 MHz free spectral range. This etalon is placed in a temperature-controlled evacuated chamber (pressure less than 10^{-5} Pa) and supported through a 3-level mechanical isolation stage that efficiently diminish the acoustic noise of the cavity that limits the spectral purity and the ultimate frequency stability of the locked diode laser.

The main part of the diode laser power is reflected by the polarising beamsplitter to a set of double-passed acousto-optic modulators, that have a bandwidth larger than one half of the free spectral range of the cavity, that allows continuous tuning of the difference-frequency without causing the laser beams to deviate. The two frequency-shifted laser beams are superposed, injected in a polarisation-maintaining optical fiber - that guarantee spatial overlapping of heterodyned laser beams - and finally coupled to the photomixer. The synthesised terahertz frequency can be estimated by the sum of an integer multiple of the free spectral range of the Fabry-Perot cavity and the two acousto-optic frequency shifts. Absolute frequency calibration of the spectrometer can be further derived from the measurement of molecular lines [7].

III. Projection for an Intracavity Photomixer Spectrometer

The main advantage of the photomixer spectrometer - its broadband tunability over the THz domain - is counterbalanced by a relatively small power level available that limits the ultimate sensitivity in direct absorption spectroscopy experiments. We propose the design of an intracavity photomixer spectrometer where THz radiation is coupled to a high-finesse Fabry-Perot cavity containing the molecular species of interest. This system represents the extension at THz frequencies of intracavity microwave spectroscopy experiments using the high-sensitivity Flygare FTMW technique [8] in the time domain. We will discuss a key issue of such system that is obtaining a high quality factor of the resonator over a broad frequency range and we will overview some potential detection schemes.

A. Cavity-Enhanced Spectroscopy

We will discuss the cavity-enhanced spectroscopy by considering the absorbing molecules placed in a Fabry-Perot cavity obtained by aligning two mirrors at a separation d comparing to the cavity axis. The transmission T, the absorption A and the reflection R coefficients of the cavity mirrors (R+T+A=1) determinates the finesse of the cavity

$$F = \frac{\pi}{T+A} = \frac{FSR}{FWHM} \quad (1)$$

that corresponds to the ratio of the free spectral range of the cavity to the linewidth of its modes.

If P_i is the incident Terahertz power coupled to the cavity, one can derive how the transmitted power P_t is modified by the absorbing medium:

$$\frac{P_t}{P_i} = \left(\frac{T}{T+A+\alpha d}\right)^2 \approx \left(\frac{T}{T+A}\right)^2 (1 - \frac{2\alpha d}{T+A}) \quad (2)$$

where the absorption coefficient α par unit length of the sample gas is supposed low comparing to the cavity loss ($\alpha d << T,A$). The contrast of the absorption signal

$$\frac{\delta P_t}{P_t} = -\frac{2\alpha d}{T+A} = -\frac{2F}{\pi}\alpha d \quad (3)$$

is enhanced by a factor of $2F/\pi$, compared to the contrast of the absorption signal in a direct absorption measurement in a sample cell of length d.

Although cavity finesse is a key element in improving the detection sensitivity, the role of T and A is not symmetric: one should optimize the transmission T over the mirror loss A in order to increase the absolute value of the transmitted signal. Finally, for a higher absorption coefficient ($\alpha d \sim T,A$) one should optimise the mirror parameters individually in order to maximise the molecular absorption signal.

Direct [1] and homodyne [9] detection schemes have been demonstrated with a photomixer-generated Terahertz-wave and can be further adapted to a cavity-enhanced photomixer spectrometer. The latter technique is closer to the original ideas of the Fourier-transform microwave spectrometer [8], where the radiation from free-induction decay of molecules is detected by an antenna and is processed through heterodyne mixing. A homodyne detection used for a cavity-enhanced photomixer spectrometer will allow phase-coherent detection of the molecular signal, although the photoresponse of LT-GaAs could be a limiting factor to the overall detection sensitivity of the system. Commercial InAs bolometers with 1 MHz bandwidth, low detection noise (NEP=2 pW.Hz$^{1/2}$) providing a high responsivity (4.10^4 V/W) could be promising for a sensitive direct detection scheme using high-frequency modulation techniques.

B. High finesse mirrors for the THz domain

Electroformed metal meshes have been used as reflecting elements for Fabry-Perot cavities in the far-infrared domain [10]. The flatness of such reflective material should be better than λ/F, in order to limit the cavity diffraction losses, that could be a critical factor for high-finesse resonators. Recently, multi-layer mirrors made of silicon wafers separated by empty gaps have been demonstrated [11] and used in the p-Ge laser cavity but accurate measurement of the Q-factor of the cavity is lacking.

We developed a prototype of high finesse mirror consisting in a metal-mesh deposited on a silicon substrate. High resistivity silicon provides low absorption in the far-infrared domain and have a relatively high index of refraction n=3,42. Electron-beam lithography followed by a metal lift-off process was used to define a Ni-Au (thicknesses: 100Å-100nm) one-dimensional metallic microstructuration on a silicon wafer, shown in Figure 3. The reflection and the absorption coefficients of the

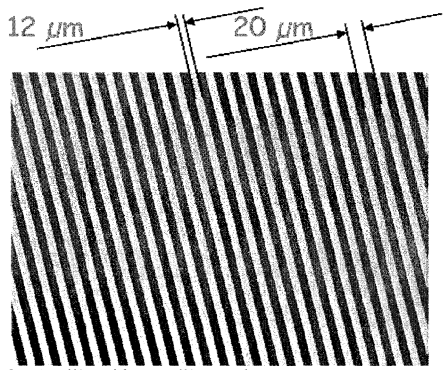

Figure 3: Metallic grid on a silicon substrate

dielectrically-backed mesh can be modelised from a transmission line formalism [12] that take into account the geometry of the structure and its resistive losses. At wavelengths longer than the periodicity of the stripes, the radiation with the electric field polarised along the metallic stripes (inductive grid) is strongly reflected, while the radiation with a crossed polarisation is transmitted (capacitive grid). The transmissivity of the inductive grid increases when the laser wavelength decreases to the periodicity of the grid. Because of interference effects in the high-index silicon substrate, a modulation is superposed on the transmission of the dielectric-backed grid. These features can be observed in Figure 4 that shows the numerical simulation of the transmissivity of a silicon wafer 1.2 mm thick with an inductive grid that have a width of 12 µm and a periodicity of 20 µm of its gold metallic stripes.

We characterised the far-infrared polarisation proprieties of this element at different emission lines of an optically pumped molecular laser [13]. The linearly polarised laser radiation is incident at normal angle on the silicon wafer fixed in a rotation mount. The transmitted beam is detected with a bolometer using an amplitude modulation technique.

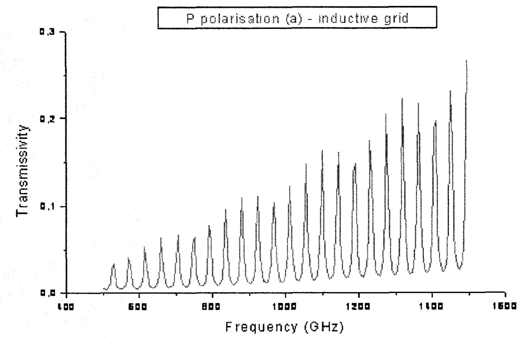

Figure 4: Simulation of the transmissivity of the metallic grid on a Silicon substrate

Figure 5 shows the fractional transmission on different laser lines when rotating the silicon wafer. At 635 GHz laser line, it varies from 70% for the capacitive grid and less than 1%

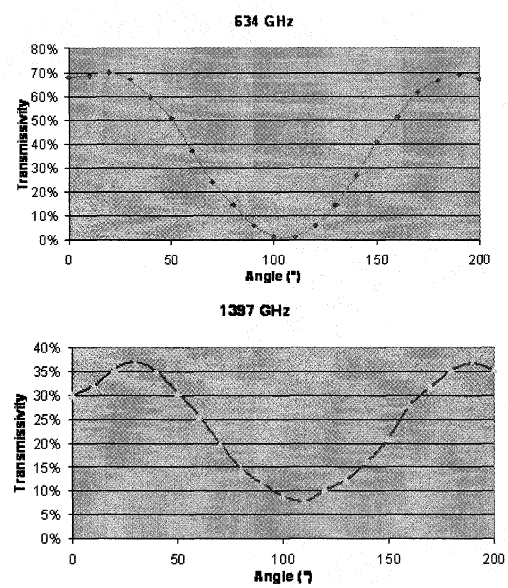

Figure 5: Extinction of linear polarised laser radiation

for the inductive grid. At higher frequency on the 1397 GHz laser line, the fractional transmission of the inductive grid grows up at 7%, while the fractional transmission of the capacitive grid diminish at 36% due to the channel spectrum in the silicon substrate. An anti-reflection coating designed at the specified laser line should limit this parasitic interference effect. The high extinction factor achieved with the inductive grids seems very promising for using them as high-finesse mirrors.

C. A high-finesse Fabry-Perot interferometer with polarisation-dependent mirrors

We set up a prototype of a Fabry-Perot interferometer using two meshes as polarisation-dependent mirrors. The linearly polarised laser beam is collimated, crosses at normal incidence both mirrors that are distanced by 3 cm and the signal transmitted through the cavity is detected with a bolometer. One mirror is mounted on an encoder motor with submicron resolution for cavity tuning. The other is mounted on a PZT ceramic. A modulation is applied on the PZT and lock-in detection is performed on the signal detected on the bolometer.

The cavity was operated in a "high-finesse" setup by aligning the mirror stripes parallel to the laser polarisation. Figure 6 shows the first derivative of the cavity modes interrogated with the 1267 GHz laser line. The finesse of the resonator is calculated by dividing the λ/2 periodicity of the fringes to the peak-to-peak width of the resonance, multiplied by a $\sqrt{3}$ factor due to the first derivative of its Lorentzian lineshape.

We measured a finesse of 50 that corresponds to a quality factor of 1.2 10^4 of our Terahertz tuneable resonator at 1.2 THz. An estimation of the finesse of the Fabry-Perot cavity can be derived from the reflection coefficient of the inductive grid that could be evaluated from previous transmissivity measurements by neglecting the absorption losses. We derive an approximate value of 70 which could point out that the measured finesse could be limited by the plan-plan cavity loss induced by the divergence of the laser beam. We noticed that a high-Q factor of the cavity is maintained over a broad spectral range: for example on the 634 GHz laser line the cavity has a finesse of 150

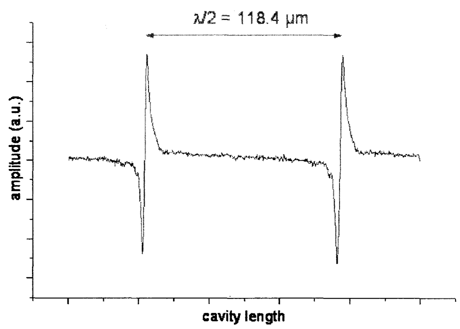

Figure 6: Fabry-Perot cavity modes at 1,2 THz with Q=1.2.10^4

(Q=1.9.10^4) and on the 2522 GHz laser line the cavity have a finesse of 16 (Q=0.8.10^4). These values are comparable with the typical quality factors of microwave cavities used in the high sensitive FTMW spectroscopy technique [8].

A careful design of plan-spherical silicon-backed mirrors, by depositing the grids on concave substrates, associated with the coupling of TEM_{00} mode in a spherical cavity, should improve its quality factor and allow the extension of the length of the interferometer. An efficient coupling of terahertz radiation to such cavity will open the way to intracavity spectroscopy over a broad spectral range.

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